1 Deformation behavior of a NiCo multilayer with a
2 modulated grain size distribution

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9 **Abstract**

In order to gain a fundamental understanding of the deformation behavior of multilayer architectures comprised of a periodic layering of hard and soft structural features, NiCo samples possessing modulated nanocrystalline and coarse-grained microstructures were fabricated using pulsed electrodeposition. Scanning electron microscopy imaging confirmed that the samples possessed an alternating grain size distribution and the desired 1:1 thickness ratio of constitutive layers. Uniaxial tensile testing showed a rule of mixtures relationship in yield and ultimate tensile strengths for the multilayer with respect to reference samples. Notably, an increase in elongation to failure was observed in the multilayer when compared to the nanocrystalline reference. The gains in elongation were found to be associated with improved neck stability due to dislocation activity within the coarse-grained layers. The morphology of the fracture surface, considered together with the available mechanical data, electron micrographs and texture measurements form the basis of a multiscale deformation mechanism which describes a failure pathway for multilayer architectures with nanocrystalline and coarse-grained structural features.

10 *Keywords:* cellular materials, nanostructured materials, plasticity,
11 fracture, mechanical characterization

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1. Introduction

Multilayers (ML)s are a layering of alternating material structures and represent an architecture which may be engineered to improve the relative mechanical properties of the constituent material components [1–4]. In traditional ML architectures, immiscible metallic species are selected for layering such that geometric confinement imposed by layer interfaces is augmented by lattice misfit between constituents, leading to increases in interfacial barrier strength [5, 6]. The layer thicknesses may also be controlled in order to encourage specific dislocation-based strengthening mechanisms. For example, experimental and atomistic simulations have shown that as layer thicknesses reach into the nanometer regime, interfacial confinement effects suppress traditional Hall-Petch hardening and dislocation pile-up phenomena [7–9]. At these length scales, conventional dislocation-based plasticity is supplanted by dislocation confinement and transmission within individual layers, leading to significant gains in mechanical strength that exceed Hall-Petch predictions [7–9]. Once layer thicknesses approach sizes comparable to a dislocation core ($\sim 1\text{--}2\text{ nm}$), interfacial barrier strength has been shown to reduce, promoting interlayer dislocation transmission and, consequently, a reversal of strength improvements from geometric scaling effects [10].

Although the presence of nanoscale geometric features in metallic MLs have led to impressive improvements in strength, as discussed by Anderson et al. [2], confinement from a high density of interfacial misfit dislocations may lead to notable decreases in ductility and a ductile-brittle transition in fracture behavior. For example, uniaxial tensile tests of Cu/Ag MLs show a two-fold decrease in elongation to failure at layer thicknesses of less than 40 nm, when compared against monolithic samples of the same materials [11]. A similar finding was echoed in a tensile investigation of Cr/Cu MLs [12], where the dislocation storing capability of the relatively ductile Cu constituent becomes compromised at layer thicknesses less than 100 nm. In nanoscale Cu/Au and Cr/Cu MLs, these reductions in elongation to failure manifest macroscopically as fracture surfaces exhibiting brittle failure [13]. Ultimately, lattice mismatch at interfacial structures in traditional MLs are generally detrimental to plastic flow, making preservation of the intrinsic ductility of such material architectures problematic. Additionally, in the case of hard-soft nanostructured MLs such as Cr/Cu, material elongation is further handicapped by a significant decrease in the dislocation storage capability of the Cu layer at nanometer thicknesses [13]. In a notable exception to this

property trend, Mara et al. [14] have reported a measurement of ultra-high strength and a ductility of greater than 0.25 true strain in 5 nm Cu/Nb ML pillars under compressive loading. It is unclear from this study, however, if similar strains may be achieved under tensile loading. From a mechanics perspective, the direction of loading relative to the ML stacking direction is also an important consideration. In this work, the referenced literature refers to loadings directed perpendicular to the layering direction in the ML.

The effect of interfaces on strengthening and elongation to failure in MLs may be considered in the greater realm of nanostructured materials. Despite impressive Hall-Petch strengthening properties [15, 16], bulk nanocrystalline metals have also typically suffered from reduced ductility relative to their polycrystalline analogs due to early plastic instability during tensile loading [17, 18]. Recently, monolithic alloys possessing hybrid microstructures comprised of a bimodal distribution of nanocrystalline (NC) and coarse-grained (CG) crystals have been suggested as material architectures possessing both high strength and good ductility [19–22]. This concept of a bimodal grain size distribution may be adopted into the ML architecture, permitting a modulated microstructure with a low interfacial threshold to dislocation mobility. In comparison to traditional MLs, a NC-CG architecture is expected to significantly reduce interfacial lattice misfit and encourage dislocation activity between constituent layers, thereby providing an avenue for assessment of deformation evolution in modulated microstructures. A recent study from Kurmanaeva et al. [23] examined the mechanical properties of a NiFe ML. Compared to a NC reference, however, the ductility of the ML was reported to be in fact slightly reduced. This surprising finding was found to be caused by the combination of FCC and BCC phases in the sample microstructures [23], further implicating the influence of lattice misfit on sample ductility. This result underscores the need for a monophase material, from which the impact of grain fidelity modulation and implementation of the ML architecture may be directly assessed.

In an effort to understand the intrinsic deformation behavior of ML structures comprised of hard and soft features, the current study investigates the mechanical properties of a Ni₈₀Co (wt. %) ML with an alternating grain size distribution. The NiCo system was selected for study as the metallic species exhibit solid solubility in a FCC unit cell for material chemistries greater than 40 wt. % Ni [24] and NC NiCo is known to possess enhanced thermal stability [25]. The purpose of this study is two-fold: firstly, improvements in material plasticity achieved from a combination of NC and CG microstruc-

87 tures are examined; secondly, the unique architecture of the NiCo ML offers
 88 the opportunity to study the deformation behavior of relatively soft coarse-
 89 grained microstructures confined by hard nanocrystalline structural features.
 90 Due to its monolithic construction, deformation may proceed in the hard-soft
 91 ML without inhibition to dislocation activity from interfacial lattice misfit.
 92 In an effort to rigorously assess the deformation behavior of the ML archi-
 93 tecture, microscale architectural features and plasticity must be considered.
 94 Therefore the current study investigates both micro and macroscale deforma-
 95 tion events in order to provide a comprehensive multiscale characterization
 96 of the mechanical response of MLs with modulated microstructures.

97 2. Methods and Materials

98 Free-standing NiCo plates were fabricated at Integran Technologies Inc.
 99 using pulsed electrodeposition (PED) on an inert stainless steel cathode from
 100 a proprietary electroplating solution and process. This procedure is known
 101 to create high quality samples and the PED process is described in greater
 102 detail in [26–28]. CG and NC layers were deposited by modifying the cathodic
 103 pulse shape to achieve the desired microstructure. The PED parameters were
 104 varied to create a ML with a 1:1 CG-NC thickness ratio. Bulk CG and NC
 105 sheets were also fabricated to serve as a comparative reference for mechanical
 106 testing. Bath chemistries were maintained to produce a nominal Ni₈₀Co (wt.
 107 %) alloy composition. Following PED, the sheets were mechanically stripped
 108 from the stainless steel cathode. Tensile coupons with a 100 mm length, 10
 109 mm grip width, 3.5 mm grip fillet radius, and a nominal gauge dimension
 110 measuring 33 mm long (L_o) by 3 mm wide (W_o) were cut from the bulk
 111 sheets using a waterjet. The resulting coupons were deburred and ground
 112 to at least a grit of 600 with SiC abrasive papers to remove any obvious
 113 surface flaws from the gauge length. The coupons all possessed thicknesses
 114 (t_o) ranging from 0.5 – 0.7 mm. A small taper was present in the tensile
 115 coupon cross-sections due to the waterjetting process, creating a slightly
 116 trapezoidal gauge area. All measurements reported in this study use the
 117 caliper dimension for calculation where applicable. Deviations in nominal
 118 measurements due to tapering of the gauge area are rather low and were
 119 measured to be in the range of 3–5% for the specimens examined in this
 120 study. It should be noted that the effects of this measurement error did not
 121 alter the statistical significance of the presented measurements.

122 The as-deposited microstructure of the NiCo ML was imaged using scan-
 123 ning electron microscopy (SEM) and PED alloy chemistries were confirmed
 124 with energy dispersive X-ray (EDX) analysis using a Hitachi SU 3500 in-
 125 strument. For SEM imaging, samples were progressively ground to a grit
 126 of 1200 and then polished using 50 nm colloidal silica suspension. Vibra-
 127 tory polishing in colloidal silica suspension was then performed for 4 hours
 128 as the final polishing step. Colloidal silica residue was removed through Ar
 129 ion milling at a 6kV accelerating voltage with a gracing angle of 8° for 10
 130 minutes. At low magnification (Figure 1a), it can be seen that the NC and
 131 CG layers appeared to have a near equal thickness throughout the full sam-
 132 ple cross-section. The orientation of the tensile coupon dimensions relative
 133 to the features of the ML are indicated in the figure. At higher magnifica-
 134 tions (Figures 1b and 1c), a clear interface between NC and CG layers was
 135 observed, and the elongated morphology of the CG microstructure became
 136 evident. Based on the collected micrographs, the CG and NC layers were
 137 measured to be in the range of 5–10 μm thick. An asymmetric grain mor-
 138 phology associated with the PED growth direction (image down) of the CG
 139 layer was visible at high magnification (Figure 1c). The facets of the termi-
 140 nal structures created a characteristic roughness in the ML interface on the
 141 order of 1–2 μm , which has been previously observed in Fe MLs [29]. EDX
 142 analysis confirmed that the PED sample chemistries were near target, with
 143 Co compositions of 19, 17, and 20 wt. % measured for the CG, ML, and NC
 144 samples, respectively. Note that, previous studies of NiCo manufactured by
 145 a similar PED procedure report sulphur and carbon concentrations on the
 146 order of several hundred ppm [25].

147 The texture and grain size of the CG layers were characterized using
 148 electron backscatter diffraction (EBSD) mapping. EBSD maps were col-
 149 lected with a step size of 300 nm and an indexing of over 85% was achieved.
 150 Due to the high number of interfaces in the PED samples, a conservative
 151 interpolation algorithm was utilized to smoothen EBSD results, which had
 152 a negligible impact on the orientation distribution of the collected measure-
 153 ments. According to the collected EBSD maps (Figure 2), the morphology of
 154 the CG layers was columnar, with major and minor grain axes measurements
 155 in the range of approximately 5–8 and 3–5 μm , respectively. For EBSD anal-
 156 ysis, grain boundaries were defined as adjacent regions with a greater than
 157 10° misorientation. Figure 2a,b presents the sample texture in the t_o (growth
 158 orientation) and L_o directions and the corresponding orientation distribution
 159 functions of the collected EBSD data is provided in Figure 2c,d, respectively.

Figure 1: Low (a), intermediate (b) and high (c) magnification micrographs of the as-deposited ML revealed through backscatter SEM imaging. The ML lacked any observable porosity, and possessed a periodic spacing and 1:1 thickness ratio of NC and CG layers. The orientation of tensile coupon dimensions relative to the SEM image is indicated in (a). The scale bars represent lengths of 100 (a), 20 (b) and 5 μm (c). The growth direction of the ML from PED is indicated in (a).

160 According to the EBSD texture data, the CG layer possess a strong $\langle 100 \rangle$
 161 fiber texture along the growth orientation (Figure 2c), which is common in
 162 PED materials [30] and was reported in NiFe MLs [23]. Given this orienta-
 163 tion restriction, the EBSD pole figures along the loading axis (L_0) show a
 164 preferred $\langle 0kl \rangle$ orientation (Figure 2d).

165 The grain size of the NC structure was characterized using X-ray diffrac-
 166 tion (XRD) peak broadening analysis with a Rigaku MiniFlex 600 Benchtop
 167 instrument under Cu- $K\alpha$ radiation ($\lambda = 0.154 \text{ nm}$). In order to sample the
 168 cross-sectional dimension of the NC layer, the XRD spectra were collected to
 169 measure texture along the loading axis. According to XRD analysis (Figure
 170 3), the NC reference sample possessed a texture with the (111) peak exhibit-
 171 ing the strongest diffraction condition and the (111) (200) and (220) peaks
 172 had relative intensities of 100, 59 and 46, respectively. In comparison to a
 173 random texture (Ni powder), which possess relative peak intensities of 100, 42
 174 and 21 for the first three peaks [31], the NC sample exhibited some texturing

Figure 2: EBSD maps collected showing the CG texture along the growth orientation (a) and loading axis (b), which fall along the t_o and L_o directions, respectively. The CG microstructure appeared to have a columnar morphology with many grains transcending the entire layer. (c,d) The respective orientation distribution functions calculated for the EBSD patterns in (a,b) indicated a strong $\langle 100 \rangle$ fiber texture along the growth direction (a) and a $\langle 0kl \rangle$ texture along the loading direction (b). The colormap in (c) corresponds to the inverse pole figure of the EBSD data in (a,b). The scale bars represent lengths of $5 \mu\text{m}$.

175 in the (200) and (220) directions. Using the Scherrer formula, an estimate
 176 of a 17 nm average grain size for the NC microstructure was obtained af-
 177 ter correction for instrument peak broadening. This grain size is typical for
 178 NiCo electrodeposits synthesized using comparable plating conditions [25].
 179 Additionally, XRD analysis of the CG reference revealed a pronounced (200)
 180 peak, with relative intensities of 38, 100 and 35 for the (111) (200) and (220)
 181 peaks, respectively. These spectra are in good agreement with EBSD results
 182 in Figure 2b for the CG layer in the ML. Secondary phases were not detected

Figure 3: XRD spectra of the NC and CG reference samples. In contrast to the strong (200) texture in the CG sample, a (111) dominated texture was observed in the NC reference. The XRD spectra refer to texture measurements along the loading axis.

183 in the XRD spectra of the CG and NC PED samples, which is consistent with
 184 previous studies [25].

185 Room temperature ($\sim 20^\circ\text{C}$) mechanical measurements were collected via
 186 uniaxial tensile testing at a cross-head velocity of 1 mm/min (5×10^{-4} /s). In
 187 order to avoid errors associated with machine compliance, an extensometer
 188 with a 25.4 mm gauge was used. All tensile coupons were observed to fail
 189 within the extensometer gauge length. Additionally, digital image correlation
 190 (DIC) in combination with a camera system was utilized to provide local
 191 strain mapping during tensile necking. The camera system was configured
 192 to capture photographs at a frequency of 1 Hz, which corresponded to one
 193 photograph per 5×10^{-4} increment of engineering strain. Fractography of

194 failed tensile coupons was performed using SEM imaging and trenching of
 195 the ML fracture surface to reveal NC and CG features was achieved with a
 196 focused ion beam (FIB) milling instrument under protective tungsten layer
 197 deposition.

198 3. Results and Discussion

199 3.1. Tensile properties and fractography of the PED samples

200 Figure 4 presents the engineering stress-strain curves (σ , ϵ) of representa-
 201 tive CG, ML and NC samples. The yield (σ_{YS}) and ultimate tensile strengths
 202 (σ_{UTS}) of the CG, ML and NC samples were measured to be 365 ± 48 , 664
 203 ± 15 and 1017 ± 30 MPa and 557 ± 11 , 1112 ± 24 and 1622 ± 39 MPa,
 204 respectively. σ_{YS} was determined using the 0.2% strain offset definition and
 205 error is reported here as a 95% confidence interval ($n = 3$). In terms of
 206 elongation to failure, the CG and NC reference samples exhibited the ex-
 207 pected elastic-plastic material responses, with evidence of necking occurring
 208 in both specimens. Due to the limited dislocation storage capacity of the NC
 209 microstructure, the ductility of the NC sample was substantially less than
 210 the CG reference. The post-yielding elongation of the PED samples may be
 211 divided into two categories, with elongation occurring prior to and after peak
 212 load σ_{UTS} , representing the uniform (ϵ_u) and non-uniform (ϵ_{nu}) deformation
 213 of the sample [32]. Based on this classification, ϵ_u and ϵ_{nu} values of $0.113 \pm$
 214 0.007 and 0.049 ± 0.009 (CG), 0.042 ± 0.003 and 0.027 ± 0.015 (ML), and
 215 0.037 ± 0.004 and 0.020 ± 0.006 (NC) were measured for the PED samples,
 216 respectively. As expected, the CG reference possessed the highest uniform
 217 elongation. Additionally, the ML sample possessed only marginally higher
 218 uniform elongation when compared against the NC reference, highlighting
 219 the role of the NC layer in the initiation of necking.

220 Although measurements indicated the ML sample possessed superior non-
 221 uniform deformation characteristics, when compared to the NC reference, it
 222 is difficult to draw strong conclusions from these values due to the inher-
 223 ently large error in end-of-life measurements of elongation. Therefore, planar
 224 images of fracture surfaces were collected and the reduction in area ratios
 225 (R_a) for the tested specimens were compared. R_a is defined here as the ratio
 226 of original area before deformation to final fracture area, as projected onto
 227 the plane formed by W_o and t_o . Figure 5 presents representative fracture
 228 surfaces of the PED samples. Comparatively, the CG sample exhibited the
 229 largest degree of necking, whereas the NC reference did not appear to have a

Figure 4: Uniaxial tensile response of the PED samples. In terms of yield and ultimate strengths, the ML sample appeared to follow a rule of mixtures behavior. The elongation to failure of the ML sample was also higher with respect to the NC reference. Notably, non-uniform elongation appeared to be improved with respect to the NC specimen, which indicated a better developed and stable neck. The callouts on the stress-strain curve correspond to loading stages in the DIC analysis presented in Figure 8.

230 significant neck. The ML sample showed an intermediary fracture behavior,
 231 with the development of a neck visible in the SEM image. Based on image
 232 analysis, the CG, ML and NC samples were found to have area reductions of
 233 12.2 ± 1.1 , 2.0 ± 0.2 and 1.5 ± 0.2 , respectively. From these calculations, the
 234 ML sample was found to possess a statistically significant ($p \leq 0.05$) larger
 235 reduction ratio than the NC reference. Furthermore, the effects of cross-
 236 sectional dimensions on the ability of the coupon to form a well-developed
 237 neck cannot explain the relative increase in R_a for the ML sample [33]. On
 238 the other hand, the R_a of the ML structure was still considerably lower (~ 6
 239 times) than the CG sample, which suggested that the fracture behavior of
 240 the ML was influenced significantly by the presence of the NC layer. Careful
 241 inspection of the ML fracture surface (Figure 5c) revealed a periodic stack-

Figure 5: SEM micrographs of the CG (a), NC (b), and ML (c) fracture surfaces which were captured from the perspective of the loading axis. (d) A high magnification SEM image of the periodic features observed in the ML fracture surface. The coarse and fine dimpled structures were formed by microvoid coalescence fracture modes in the CG and NC layers respectively. The scale bars are 1 mm in (a)–(c) and 5 μm in (d).

ing of microscopic features which transcended the relief cross-section. A high magnification SEM image of the ML fracture surface (Figure 5d) revealed a sequence of coarse and fine dimpled structures which corresponded to microvoid coalescence failure modes in the CG and NC layers respectively. A recent study of hard-soft Cr/Cu MLs has also reported similar microscopic features along the fracture surface, with the protrusions being identified as the ductile Cu layers [12].

249 A summary of the collected mechanical properties of the PED samples is
 250 provided in Figure 6 and Table 1. The measured values of σ_{YS} and σ_{UTS} were
 251 in reasonable agreement with rule of mixtures (ROM) predictions of 691 and
 252 1090 MPa, respectively. While elongation was somewhat improved in the ML
 253 sample, it fell short of ROM behavior. Tensile failure is complicated process
 254 which is driven by the evolution of the stress-state from a global-uniaxial
 255 to a localized-multiaxial condition, and therefore a simple ROM behavior
 256 for elongation of the ML cannot be assumed. A detailed investigation of
 257 the development of strain localization and plastic interaction between con-
 258 stituents is required in order to understand deformation behavior in the ML
 259 architecture.

260 3.2. Uniform deformation hardening behavior and strain localization during 261 necking

262 In order to understand uniform deformation behavior during tensile load-
 263 ing, the hardening response of the PED samples was examined. As shown in
 264 Figure 7, the tangent moduli of the ML and NC samples decayed in a linear
 265 manner, conforming to Stage III work hardening, whereas the CG sample
 266 exhibited both linear Stage III and near-asymptotic Stage IV behavior [34].
 267 The tangent modulus (Θ) is presented here as the ratio of differential true
 268 stress (σ_t) and true strain (ϵ_t) and the data in the figure is plotted until the
 269 point of peak load in each sample respectively. The absence of Stage IV in
 270 the ML tensile data suggested that NC layer work hardening mechanisms
 271 were dominant during uniform elongation and that contributions from the
 272 CG layer were negligible prior to necking. Considering the microstructure
 273 architecture of the ML and the weakest-link nature of tensile loading, this
 274 behavior was expected and is a consequence of the uniaxial stress-state prior
 275 to neck formation.

276 The non-uniform deformation behavior during necking was captured us-

Table 1: A summary of the mechanical measurements extracted from tensile testing of the PED samples.

PED Sample	σ_{YS} (MPa)	σ_{UTS} (MPa)	ϵ_u	ϵ_{nu}	R_a
CG	365 ± 48	557 ± 11	0.113 ± 0.007	0.049 ± 0.009	12.2 ± 1.1
ML	664 ± 15	1112 ± 24	0.042 ± 0.003	0.027 ± 0.015	2.0 ± 0.2
NC	1017 ± 30	1622 ± 39	0.037 ± 0.004	0.020 ± 0.006	1.5 ± 0.2

Figure 6: A summary of the mechanical measurements collected from the PED samples. Error bars represent a 95% confidence interval ($n = 3$) and the dashed lines indicate ROM predictions. The star markers indicate statistically significant measurements ($p \leq 0.05$).

ing DIC analysis. An image of a representative tensile coupon speckled for
 DIC analysis is presented in Figure 8a and Figure 8b presents line-scans of
 the strain profile along the normalized gauge length for each of the PED
 samples. The relative positions of the strain stages in each of the line-scans
 are indicated in the stress-strain curves of Figure 4. The selected line-scans
 present the strain localization during the following deformation stages: post-
 yield uniform elongation; at peak load; after strain localization within tensile
 the neck; and immediately prior to coupon failure. In general, the progres-
 sion of deformation in the PED samples was consistent. In each of the DIC
 tensile tests, the coupon failed within the gauge dimension of the sample. As
 anticipated from tensile data, each sample experienced a period of uniform
 elongation which transitioned to high strain localization just prior to failure.
 In the CG reference, fracture proceeded through strain development within
 a local neck. At strain stage 2, there were several local maximums along the
 gauge length. The largest of which developed into the localized neck. This
 behavior is well-known for materials with high strain hardening capabilities
 [35], where potential necks may stabilize during mechanical loading. In con-
 trast to the CG reference, the NC and ML samples appeared to experience

Figure 7: Hardening data for the PED samples up until the peak load. All samples exhibited a linear decay in the tangent modulus, which was consistent with Stage III hardening. The CG sample also showed asymptotic Stage IV hardening. This behavior was absent in the ML sample, which indicated that the CG layer did not contribute significantly to deformation behavior before necking.

295 a mixed mode of deformation behavior, with diffuse and local necks evident
 296 in the strain maps. In order to examine the relative size of macroscale de-
 297 formation features, a methodology that tracks changes in the strain profiles
 298 during loading was proposed. The size of the diffuse (L_{dn}) and local necks
 299 (L_{ln}) were quantified by considering the spatial and temporal derivatives of
 300 the DIC line-scans. Excluding the shoulder, the diffuse neck represented the
 301 length of the tensile coupon where the tangent of the strain profile deviated
 302 from zero (i.e. $|d\epsilon/dx| > 0$). Similarly, the local neck was identified near the
 303 point of fracture when the temporal derivative of strain increased significantly
 304 from zero (i.e. $d\epsilon/dt > 0$). The degree of strain localization (ϵ_{ln}) was then
 305 calculated as the difference from the peak strain to the baseline at the inci-
 306 dence of the local neck. The definition of these measurements is illustrated
 307 in Figure 8c and the corresponding derivatives were calculated for each of the
 308 PED sample line-scans presented in Figure 8b. The calculated quantities of
 309 L_{dn} and L_{ln} were normalized against W_o and these measurements, along with
 310 ϵ_{ln} , are provided in Table 2. From this analysis, the CG sample possessed the

Figure 8: (a) A photograph of a tensile coupon speckled for DIC measurements. The normalized length (x/L) was defined as indicated in the image. (b) The strain profiles along the gauge length of the PED samples at various stages: 1) during uniform elongation; 2) at peak load; 3) at the initiation of localized necking; and 4) immediately prior to fracture. The relative positions of these stages are indicated on the stress-strain curves provided in Figure 4. (c) Measurements of the diffuse (L_{dn}) and local necks (L_{ln}) were defined by considering the spatial and temporal derivatives of the strain profiles in (a) for the ML sample. The strain localization in the local neck (ϵ_{ln}) was defined as the peak strain subtracted from the baseline of the local neck length. The scale bar in (a) is 5 mm.

311 highest degree of strain localization. Notably, the strain localization in the
 312 ML sample was much larger than the NC reference, indicating a significant
 313 contribution from the CG layers to deformation behavior within the tensile
 314 neck. It should be noted that a distinction between the local and diffuse
 315 necks could not be established for the CG sample. The CG measurements
 316 are therefore considered as localized neck values.

317 In order to compare the local necks in the PED samples, DIC strain maps
 318 were collected just prior to fracture (Figure 9). The colormap is normalized
 319 with respect to maximum engineering strain within each of the PED sam-
 320 ples and the final dimensions of each tensile coupon are scaled to fit the
 321 image frame. As anticipated from tensile stress-strain data, the CG sample
 322 appeared to fail by conventional necking mechanisms. Conversely, strain lo-
 323 calization in the ML and NC samples was confined to a band rotated 55°
 324 to the loading axis. Macroscopic shear banding at this angle is a commonly
 325 reported fracture mechanism in bulk nanocrystalline metals [36–38] and is
 326 a result in the collapse of dislocation storage capacity within nanoscale mi-
 327 crostructures [17, 18]. It is interesting to note that although the ML sample
 328 appeared to fail macroscopically by NC dominated shear banding, the extent
 329 of strain localization within the band was substantially higher than the NC
 330 reference, indicating a significant contribution to material plasticity from CG
 331 layers (Figure 8b).

332 3.3. Multiscale deformation mechanism within the tensile neck

333 In order to gain insight into the interplay between fracture morphology
 334 and microscale plasticity within the tensile neck of the ML samples, SEM im-
 335 ages captured perpendicular to the loading axis of the coupon are provided
 336 in Figure 10. The low magnification images presented in Figures 10a and
 337 10b are mating fracture surfaces and examination of the images revealed a
 338 number of interesting features. As in DIC analysis of the planar dimensions,

Table 2: The width-normalized lengths of the diffuse and local necks, and strain localiza-
 tion in the PED samples.

PED Sample	L_{dn}	L_{ln}	ϵ_{ln}
CG	-	$\approx 2W_o$	0.18
ML	$\approx 3W_o$	$\approx W_o$	0.11
NC	$\approx 3.5W_o$	$\approx W_o$	0.03

Figure 9: The strain distribution across the (a) CG, (b) ML, and (c) NC tensile coupons immediately prior to fracture. Macroscopic shear banding at 55° from the loading axis was evident in the ML and NC samples. The colormapping represents a normalization of the maximum strains in each corresponding PED sample.

339 the fracture plane also appeared inclined at 55° to the loading axis through
 340 the coupon cross-section. The shear band is therefore inclined at compound
 341 angles of 55° to the loading axis in both the planar and cross-section views,
 342 forming a fracture plane which sectioned the waist of the tensile coupon
 343 along an oblique path (Figure 10b). Consequently, although the fracture
 344 plane initiated at the thinnest cross-section, fracture did not propagate di-
 345 rectly through the waist. This phenomenon was a factor responsible for the
 346 drastic reduction in R_a relative to the CG reference. It is interesting to note
 347 that in the cross-section view the fracture plane appeared to fall along planes
 348 oriented $\pm 55^\circ$ to the loading axis. Conversely, observations from DIC test-
 349 ing showed a single fracture path in the planar view. This behavior was not
 350 observed in the NC sample (Figure 5c) and was unique to the ML coupons.
 351 It is possible that the laminate layering of CG microstructure through the
 352 sample cross-section disrupted the formation of a singular shear band ori-
 353 entation, permitting a mixed fracture plane. On the microscopic scale, the
 354 flow of NC and CG layers into the multiaxial stress field of the localized neck

Figure 10: (a) A low magnification SEM image of the fractured ML sample. The sample appears to have fractured through a shear band oriented at $\pm 55^\circ$ to the loading axis. The orientation of the nominal tensile coupon dimensions relative to the image plane is provided. (b) An SEM image of the mating fracture surface to (a) after polishing. The flow of CG and NC layers into the neck is evident. The fracture path initiated at the thinnest region of the coupon waist and propagated along an oblique path, following the observed shear band. (c) A high magnification SEM micrograph of the CG and NC layers within the diffuse neck. Arrows highlight distortion in the CG microstructure along the interface between layers, suggesting dislocation activity that contributed to the improved necking stability in the ML. The scale bars are 200 in (a) and (b), and $2\mu\text{m}$ in (c).

355 is clearly visible in Figure 10b. A higher magnification micrograph (Figure
356 10c) images the interfacial boundary between layers in the tensile neck. The
357 pronounced refinement and distortion of the CG layer microstructure near
358 the ML interface was likely the result of interfacial dislocation activity which
359 was caused by the multiaxial stress-state resulting from macroscopic strain
360 localization in both the diffuse and local necks. From a macromechanical per-
361 spective, this deformation accommodation in the interfacial structures was
362 believed to contribute to the comparatively increased tensile strain measured
363 within the necking region of the ML (Figure 8b).

364 As observed in reduction area measurements (Figure 5c), the fracture sur-
365 face shown in Figure 10a is populated by a landscape of dimpled CG and NC
366 features. In order to gain insight into the relative topography of these fea-
367 tures, FIB milling was implemented to cut a trench within the fracture plane
368 and image the cross-section of the dimpled features. Figure 11a presents a
369 low magnification SEM image which provides perspective for the higher mag-
370 nification images. The FIB was positioned such that the trench was aligned
371 approximately parallel to the loading axis. The resulting milled surface is
372 imaged in Figure 11b. The axes provided in the figure may be referenced
373 with respect to Figure 11a. Despite the large deformation near the fracture
374 surface, backscatter SEM imaging (Figure 11c) of the trench cross-section
375 revealed a faint channeling contrast in the underlying microstructure of the
376 fracture surface. From these microstructure features, it can be seen that the
377 large dimples correspond to failure in the CG layer whereas the finer dim-
378 ples represent fracture in the NC layer. Note that, the bright layer visible
379 in the image is a surface-conforming protective layer of tungsten which was
380 deposited to prevent damage to the underlying material and also assisted
381 in visualizing the topographical landscape of the fracture surface. Further
382 examination of the trenched cross-section showed that the CG layers have
383 a saw-toothed shape that protruded approximately 2-3 μm above the frac-
384 ture plane. The outline of these features was measured to be approximately
385 45° from the loading axis. Considering the $\langle 100 \rangle$ texture measured in the
386 EBSD data (Figure 2), and the proximity of the observed CG layer fracture
387 angle relative to the localized neck (55° to the loading axis), this fracture
388 orientation likely corresponds to activation of the $\langle 110 \rangle / \{111\}$ slip system,
389 which has been previously reported as a possible deformation mechanism in
390 NiFe MLs [23].

391 The deformation behavior of the NiCo ML is an inherently multiscale phe-
392 nomenon in which the modulation of grain size on the microscale translates

Figure 11: (a) An SEM micrograph of the ML fracture surface taken from a perspective orientation. (b) An image of the trench which was cut using FIB milling. The trench is aligned approximately to the loading axis of the tensile coupon. The orientation of the nominal tensile coupon dimensions relative to the image plane is provided in (a) and (b). (c) A backscatter SEM image of a trench milled into the fracture surface. The protruding structures appear to have a saw-tooth geometry which were inclined at approximately 45° to the loading axis. The scale bars are 1 mm (a), $50\text{ }\mu\text{m}$ (b), and $5\text{ }\mu\text{m}$ (c).

393 into the macroscopically observed fracture geometry. Figure 12 presents a
 394 schematic representation of this multiscale deformation mechanism. In each
 395 test, tensile failure initiated from the surface of the coupon, which was con-
 396 sistent with observations in other mechanical studies of PED materials [39].
 397 Due to the relatively low strain hardening capability of the NC layer, a
 398 macroscopic shear band formed, which developed into a localized neck. The
 399 change from a uniaxial to a multiaxial stress-state caused plastic interaction
 400 between the constituent layers and led to the formation of a shear band,
 401 which activated the $\langle 110 \rangle / \{111\}$ slip system in the CG layer and permit-
 402 ted the development of a significant neck. Continued elongation resulted in
 403 fracturing of both the NC and CG layers by microvoid coalescence. Macro-

Figure 12: The proposed multiscale deformation mechanism for MLs with modulated hard-soft NC and CG layers. Micro- and macroscale deformation phenomena are illustrated schematically during necking and upon ultimate fracture of the ML. Material flowed from the diffuse into the localized neck where fracture initiated along the shear band at the thinnest region. The fracture plane cut across the coupon waist at an oblique angle of 55° through the sample width and thickness. The projection of this fracture plane is the measurement R_a . On the microscale, the activation of the $\langle 110 \rangle / \{111\}$ slip system in the CG layer manifested into the saw-toothed protrusions observed in SEM micrographs of the ML fracture surface.

scopically, the fracture plane initiated at the thinnest region of the loaded cross-section and propagated along the shear band. This behavior caused the waist of the tensile coupon to be cut along an oblique angle oriented at 55° from the loading axis along the width and thickness of the sample. In comparison to the CG sample, the oblique orientation of the fracture path relative to the loading axis was also likely the cause of the drastically reduced R_a . The relatively small area reduction due to uniform elongation is not represented in the schematic. On the microscopic scale, the dimpled fracture surface was populated by periodic saw-toothed protrusions which revealed the topographical landscape of the CG layer. It is expected that the orientation of CG protrusions in the fracture surface is highly texture dependent. A more randomized texture may lead to a less uniform saw-toothed fracture feature, but is not expected to significantly alter the interacting deformation mechanisms observed in the ML architecture.

4. Conclusions

The deformation behavior of a pulse electrodeposited NiCo multilayer with an alternating grain size distribution was studied. Uniaxial testing showed a rule of mixtures yield and tensile strength in the ML relative to CG and NC references. Digital image correlation analysis and examination of the ML fracture surface offered important insight into the micro- and macroscale deformation mechanisms leading to ultimate material failure. Based on these observations, a multiscale mechanism was proposed to describe the deformation behavior in MLs with modulated hard-soft NC and CG layers. Fracture was preceded by the development of a diffuse and local neck within the ML sample. Inside the local neck, a shear band was observed to form. The shear band was oriented at 55° to the loading axis along both the coupon width and thickness. Upon the formation of a shear band, the resulting multiaxial stress state induced significant deformation in the CG layer, improving the neck stability in the ML. Further elongation of the sample resulted in ultimate failure with fracture initiating at the thinnest point of the neck cross-section and propagating along the shear band, which sectioned the coupon waist at an oblique angle relative to the loading axis. High magnification SEM micrographs indicated dislocation activity across ML interfaces, leading to significant material plasticity and improved non-uniform ductility within the sample neck. Furthermore, failure of the CG layers yielded periodic topographical features which protruded from the fracture surface. The

440 saw-toothed topography was created by activation of the $\langle 110 \rangle / \{111\}$ slip
441 system in the ML, which was a consequence of the texture of the CG layer
442 as well as the orientation of a shear band during tensile loading.

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