Ultrastrong nanotwinned pure nickel with extremely fine twin thickness

Fenghui Duan¹, Yan Lin¹, Jie Pan¹*, Lei Zhao², Qiang Guo²*, Di Zhang², Yi Li¹*

The strength of nanocrystalline and nanotwinned metals stops increasing or even starts decreasing when their grain size or twin thickness is below a critical size—a phenomenon known as Hall-Petch breakdown—which hinders the attainment of ultrahigh strength. Here, we report continuous strengthening in nanotwinned pure Ni with twin thicknesses ranging from 81.0 to 2.9 nm. An unprecedented strength of 4.0 GPa was achieved at extremely fine twin thickness of 2.9 nm, which is about 12 times stronger than that of conventional coarse-grained nickel. This ultrahigh strength arises from the excellent stability of twin boundaries and their strong impedance to dislocation motion. In particular, we find that secondary nanotwins are activated to sustain plastic deformation, which also contribute to the high strength. These results not only advance the understanding of the strengthening mechanisms in nanotwinned metals but also offer an alternative pathway to develop engineering materials with ultrahigh strength.

RESULTS

Microstructures of fabricated NT-Ni

High-purity (99.75 weight %; table S1) bulk Ni specimens (300 μm in thickness) containing a high density of nanoscale twin lamellae embedded within nanometer-sized columnar grains were synthesized by DC electrodeposition in a citrate bath (see Materials and Methods for details). By regulating the nickel and citrate ion contents in the electrolyte, we succeeded in refining the average twin thickness from 81.0 to 2.9 nm while maintaining the highly ordered, parallel nature of the twins (fig. S1). The average twin thickness is correlated with the average columnar grain width; a smaller columnar grain width is accompanied by a finer average twin thickness (table S2). All NT-Ni specimens exhibit the same strong (111) crystallographic texture (fig. S2). For simplicity, each NT-Ni specimen is identified by its average twin thickness (λ); for example, the specimen with λ = 2.9 nm is referred to as NT-2.9.

Figure 1A shows the typical three-dimensional microstructure of the as-deposited NT-2.9 specimen. Plan-view TEM image shows equiaxed nano-sized grains, while cross-section images reveal elongated columnar grains embedded with nanoscale growth twins (fig. S3). The columnar grain width is between 14 and 48 nm, with an average value of 28 nm (fig. 1B). The twin thickness shows a narrow distribution ranging from 0.5 to 15 nm (fig. 1C). Magnified

1Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, 72 Wenhua Road, Shenyang 110016, China. 2State Key Lab of Metal Matrix Composites, Shanghai Jiao Tong University, 800 Dongchuan Road, Shanghai 200240, China.

*Corresponding author. Email: jiepan@imr.ac.cn (J.P.); guoq@sjtu.edu.cn (Q.G.); liyi@imr.ac.cn (Y. Li)
TEM micrographs (Fig. 1, D and E) indicate that most of the twin planes have coherent TBs (CTBs). In addition to these CTBs, a small fraction (less than 2%) of incoherent TBs and stacking faults (SFs) are also observed. The x-ray diffraction (XRD) pattern of the NT-2.9 specimen indicates a (111) out-of-plane crystallographic texture and the presence of a faint (200) peak (Fig. 1F); this is consistent with the TEM results showing that the long axis of the columnar grains and the normal direction of the (111) twin plane are parallel to the growth direction.

Nanotwin formation mechanism

Introducing nanoscale growth twins, especially those with extremely fine spacings, is difficult in Ni because of its high SF energy (SFE) of 128 mJ/m² (14). Nanotwin formation mechanism during electrodeposition is a fundamental issue, which has not been clearly elucidated until now. However, according to our experimental observations, the nanotwin formation mechanism of our NT-Ni can be interpreted from two aspects: First, high deposition rate favors the formation of high-density nanotwin structure; second, the tunability of average twin thickness is closely related to the generation of internal tensile stress in the deposit.

Electrodeposition is a nonequilibrium process, and the widespread formation of twinned nuclei is favorable when the relative radius difference between critical sizes of defect-free nucleus (r*_{perfect}) and a twinned nucleus (r*_{twin}) can be minimized (15). Higher deposition rate (~current density) induces the smaller relative radius difference (detailed derivation is provided in the Supplementary Materials), leading to the formation of NT structure (fig. S4). For our Ni with high SFE, only when the deposition rate is higher than 13 nm/s do we observe columnar grains embedded with nanoscale twin lamellae in the as-deposited Ni (fig. S4). In contrast, when the deposition rate is below 13 nm/s, only equiaxed nano-sized grains were observed. This experimental observation confirms the above-mentioned theory and is also consistent with previous reports (15–17), wherein nanotwins were formed under the condition of a high deposition rate. Note that the critical deposition rate of 13 nm/s for nanotwin formation in Ni is much higher than that (0.6 to 8 nm/s) in Cu (1, 15), suggesting that higher deposition rate is needed to favor the formation of high-density nanotwins in metals with higher SFE.

Internal tensile stress was reported to be present during the deposition of NT-Cu; the formation of twins helped relax the stress and thus lower the total energy of the deposited samples (18). In other words, the stress-relaxed NT-metals is energetically more stable than the highly stressed deposits without twins. It also indicates that the higher tensile stress state may assist the formation of finer twin-spaced nanotwins. We calculated the internal tensile stress...
using the classic Stoney equation (19) and found that the calculated internal stresses of NT-Ni specimens with $\lambda = 2.9$, 4.4, 16.5, and 81.0 nm are approximately 500, 340, 90, and 20 MPa, respectively (fig. S5). This negative correlation between tensile stress and twin thickness confirms our speculations. Note that the generation of internal tensile stress in our deposits was realized by adding a specific chemical additive (2-butyne-1,4-diol) in the bath (20) and internal tensile stress levels were regulated by changing the concentration ratio of citrate and nickel ion. The lower concentration ratio of citrate and nickel ion usually results in higher internal tensile stress and thus finer twin thickness.

Hydrogen was additionally reported to promote twin nucleation (21). The absorbed hydrogen ions at the cathode surface lead to a reduction of SFE and an increase in the cathodic overpotential, both of which may contribute to the high density of nanotwin formation in Ni.

**Continuous strengthening**

To evaluate the mechanical properties of NT-Ni with extremely fine twin thickness, we conducted uniaxial compression tests on micro-pillars having a diameter of ~1.3 μm. The loading direction was normal to TBs. Figure 2 presents the true stress-strain curves for NT-2.9 and NT-6.4 specimens, together with those for NG-Ni and coarse-grained (CG) Ni with grain sizes of 20 nm and 80 μm obtained from (22) for comparison. The NT-6.4 specimen exhibits a flow stress of 2.9 GPa at 2% plastic strain (here, we assign the flow stress of 2.9 GPa at 2% plastic strain (here, we assign the flow stress for data fitting here is $\lambda = 2.9$ nm. Note that the relationship between hardness and strength, i.e., the Tabor factor for NT-Ni, is in the range of 2.1 to 2.4, smaller than the empirical value of 3. This is mainly attributed to the strongly textured structure (10, 36) and the high internal tensile stress in the deposits (37, 38). After annealing at 523 K, internal tensile stresses are released and GBs relax (5), leading to further enhancement in the hardness. A maximum value of ~9.5 GPa is achieved for the NT-2.9 specimen (table S2). This continuous strengthening and hardening with decreasing twin thickness are in stark contrast to the softening observed in NG-Ni-Mo, NG-Ni-W, and NT-Cu (Fig. 3 and fig. S7), for which both the hardness and yield strength decrease when the grain size or twin thickness is below a critical value (typically 10 to 15 nm) (2, 5, 39).

**Fig. 2. Mechanical properties of NT-Ni pillars.** Uniaxial true stress-strain curves for pillars showing that the flow stress at 2% plastic strain in the NT-2.9 and NT-6.4 specimens is 4.0 and 2.9 GPa, respectively. The true stress-strain curves for NG- and CG-Ni from (22) are also presented for comparison. The red square, orange circle, and blue and black triangles denote the flow stresses at 2% plastic strain for the four samples. The inset displays a schematic of the compression test that was carried out on 1.3-μm-diameter NT-Ni specimens.

**Fig. 3. Continuous strengthening in NT-Ni.** Variation in the yield strength with average grain size or twin thickness for Ni and Mo-microalloyed NT-Ni (1.3 at. %), along with literature data directly obtained by tensile and compression tests for electrodeposited (ED) Ni, Ni pillars, ED NT-Ni (22, 24–33), and NT-Cu (2). Continuous strengthening behavior extending to twin thickness of 2.9 and 1.9 nm is observed in the as-deposited NT-Ni and Mo-microalloyed NT-Ni specimens, respectively. Conversely, softening behavior, i.e., decreasing yield strength with decreasing grain size or twin thickness, is observed in the as-deposited NT-Cu when the average twin thickness is below 10 to 15 nm. The Hall-Petch relationship used for data fitting here is $\sigma_y = 0.267 \text{ GPa} + 6.233 \text{ GPa} \times d^{-1/2}$, where $d$ is the grain size or twin thickness and $\lambda$ is the twin thickness (39, 55).
Microstructure evolution and strengthening mechanisms

To explore the mechanisms that are responsible for this continuous strengthening, we characterized the microstructures of the NT-2.9 specimen after deformation. In the deformed region of the NT-2.9 pillar after ~3% plastic strain (Fig. 4A), we found that the high density of nanotwins was retained and that the average twin thickness was $3.4 \pm 1.9$ nm (inset in Fig. 4B), which is nearly identical to the value of $2.9 \pm 1.9$ nm before deformation. This observation indicates a high stability of nanotwins in Ni, in line with the absence of detwinning in compressed NT-N\textsubscript{84}Mo\textsubscript{11}W\textsubscript{5} alloys (34), but in contrast to the intensive detwinning behavior previously reported in columnar-grained NT-Cu loaded at a 72° incline with respect to the TBs (13). This high nanotwin stability arises from the suppressed activity of twinning partial dislocations along TBs, i.e., the detwinning process. On the one hand, the nucleation and slip of twinning partial dislocations along TBs were not favored, as the resolved shear stress on twin planes is negligible under normal loading (10). On the other hand, the high SFE of Ni could also play an important role in hindering the detwinning process (40). In low-SFE metals, twinning partial dislocations easily nucleate, and their glide on the twin planes results in detwinning and subsequent softening (8, 41). In contrast, in Ni with high SFE, partial dislocations tend to constrict into perfect dislocations (40, 42, 43), as evidenced in fig. S8. Perfect dislocations gliding on the twin planes cannot lead to the elimination of TBs. As detwinning is suppressed, the dense TBs can play a vital role on impeding dislocations, which is an essential prerequisite for continuous strengthening.

We also investigated the interactions between TBs and dislocations using TEM. Plenty of dislocations with Burgers vectors inclined to TBs [hard mode I dislocations, see definition in (10)] are observed within twin lamellae and/or at TBs (Fig. 4C and S8). In particular, we observed partial dislocations with the Burgers vectors of a/6<112> in hard mode I, as shown in Fig. 4 (C and D). Hard mode I dislocations are known to be the dominant deformation mechanism in highly oriented nanotwins when the loading direction is perpendicular to the TBs, leading to continuous strengthening with decreasing twin thickness (10). In addition, we found the presence of secondary nanotwins (Fig. 5, A to D), which initiate at the intersection of TB and column GB (Fig. 5B), run across several TBs, and are finally blocked by primary TBs (Fig. 5, C and D). Magnified TEM observation (Fig. 5B) shows that several SFs appear at the front of secondary nanotwin, which indicates that the formation of secondary nanotwins may originate from the simultaneous and cooperative activation of different Shockley partial dislocations (hard mode I) on neighboring parallel glide planes (44). The above observations confirm that the strengthening mechanism in the sub–10-nm regime of NT-Ni is mainly attributed to the effective barrier against the motion of hard mode I dislocations by the primary TB. Moreover, secondary nanotwins create obstacles to impede dislocation motions, providing additional strengthening in the NT-Ni (44, 45).
DISCUSSION
Secondary nanotwins or hierarchical nanotwins are easily formed in metals or alloys having low SFE, such as Ag \((\gamma_{\text{SF}} = 16 \text{ mJ/m}^2)\) and twinning-induced plasticity steel \((\gamma_{\text{SF}} = 20 \text{ to } 50 \text{ mJ/m}^2)\) \((45, 46)\). However, in metals with high SFE, such as Ni, the formation of secondary nanotwins is unexpected because of the higher energy barrier associated with the emission of a partial dislocation from GB or TB and the subsequent extending of the SF ribbon across the grain. However, the activation of partial dislocations has a strong dependency on the boundary spacing. According to a previous prediction, partial dislocations are preferentially activated over perfect dislocations when the boundary spacing is below a critical value \((17 \text{ nm for Ni}) \) \((47)\). In our NT-2.9, the TB spacing, 2.9 nm, is much smaller than the critical value, whereas columnar grain size, 28 nm, is larger than the critical value. Partial dislocations are therefore generated in hard mode I within extremely fine nanotwins, while perfect dislocations tend to be constricted into leading and trailing partial dislocations as they are constrained within larger columnar grains.

An analytical model has been developed by Zhu et al. \((48)\) to describe the nucleation and growth of secondary twins in NG face-centered cubic metals. According to this model, the calculated critical yield stress for the twin nucleation in NT-Ni specimen is about 6.2 GPa (see the Supplementary Materials for details). Considering that stress concentration is commonly generated at GBs during deformation, this high critical yield stress value for the twin nucleation can be readily reached in the NT-2.9 specimen with high yield stress of 4.0 GPa and thus induce the secondary twinning. On the basis of this model, we also found that the critical resolved shear stress for twin nucleation is smaller than that to activate full dislocation at extremely fine twin thickness (fig. S10), consistent with our observation that secondary twinning is one of the dominant deformation mechanisms.

In addition, our experimental results show that secondary twins are only formed in NT-Ni with an extremely fine twin thickness, rather than those with a large twin thickness \((\text{e.g., } \lambda = 81.0 \text{ nm})\). This indicates that a transition exists in the strengthening mechanism of NT-Ni, from dislocation pile-up at a large twin thickness to secondary nanotwin strengthening at an extremely fine twin thickness.

Before concluding, we note that the observed continuous strengthening can be further extended to a twin thickness of 1.9 nm. This finer twin thickness is realized by microalloying Mo in Ni, which slightly decreases the SFE and promotes the formation of nanotwins. In this Mo-microalloyed NT-Ni \([1.3 \text{ atomic } \% \text{ (at } \%) \text{ Mo; see fig. S11}]\) with \(\lambda = 1.9 \text{ nm, its } \sigma_c \text{ (under normal loading conditions)}\) is 4.4 GPa (fig. S12), which is 0.4 GPa higher than that of the NT-2.9 specimen, and also lies on the extrapolated Hall-Petch line (fig. 3).

The strength of our Mo-microalloyed NT-Ni is 1.0 GPa higher than that of NT-Ni0.3Mo14W2.4 film with nearly the same \(\lambda \) \((1.8 \text{ nm})\) \((35)\). The results \((\text{extending of strengthening to the twin thickness of 1.9 nm})\) are indicative of that the higher strength even to the ideal theoretical limit may be achieved at angstrom-scale twin thickness.

In summary, our NT-Ni with extremely fine twin thickness, achieved via DC electrodeposition, exhibits a strength of 4.0 GPa—greater than those of known pure Ni. This strength derives from the continuous strengthening extending to the finest twin thickness \(2.9 \text{ nm}\). The continuous strengthening behavior originates from the excellent stability of TBs and their effective barrier on the nucleation and motion of dislocations, plus secondary nanotwins as new obstacles to further impede dislocation motions. The present study not only provides an opportunity for the synthesis of NT-structures with extremely fine twin thicknesses in high-SFE metals but also clarifies that Hall-Petch strengthening can be extended to extremely fine structure dimensions via structural constructing in nanostructured metals. Our work reveals a new practical strategy for the design of ultrastrong and ultrahard materials for potential applications in micro-electromechanical system and surface coatings.

MATERIALS AND METHODS

Materials preparation
The NT-Ni specimens with the plane dimensions of 20 mm × 10 mm and a thickness of approximately 300 μm were synthesized using conventional (DC) electrodeposition. The plating bath was a novel citrate electrolyte composed of NiSO4·6H2O \((60 \text{ to } 300 \text{ g/liter, NiCl}_2·6\text{H}_2\text{O} (10 \text{ g/liter, Na}_3\text{C}_6\text{H}_5\text{O}_7·2\text{H}_2\text{O} (60 \text{ to } 120 \text{ g/liter, sodium saccharin (1.0 g/liter, and 2-butyne-1,4-diol (0.64 g/liter, maintained at a pH value of 6.4 to 7.0 and temperature of 338 ± 2 K. A pure nickel sheet with the dimensions of 20 mm × 10 mm × 1 mm was used as the cathode, and the initial current density is 40 to 100 mA/cm².

The Mo-microalloyed NT-Ni alloys with thickness of 20 to 50 μm were also fabricated using conventional DC electrodeposition. The plating bath was composed of NiSO4·6H2O \((60 \text{ g/liter, Na}_3\text{C}_6\text{H}_5\text{O}_7·2\text{H}_2\text{O} (80 \text{ g/liter, Na}_2\text{MoO}_4·2\text{H}_2\text{O} (0.5 \text{ g/liter, sodium saccharin (1.0 g/liter, and 2-butyne-1,4-diol (0.64 g/liter). The deposition was maintained at a pH value of 7.0 to 9.0 by adding 10% NaOH. The deposition temperature was chosen to be 313 K.

Chemical composition analysis
The contents of light elements—H, C, N, O, and S—in the as-deposited NT-Ni specimens were analyzed using a combination of simultaneous carbon/sulfur determinator (CS600, LECO) and oxygen/nitrogen/hydrogen analyzer (TCH-600, LECO). The contents of other elements were also quantified using inductively coupled plasma optical emission spectroscopy (ICP-OES; OPTIMA 8300DV). The carbon and sulfur in our specimens (table S1) originate from the additives in the plating bath, i.e., 2-butyne-1,4-diol and sodium saccharin, and their contents in NT-Ni specimens are consistent with those reported previously in \((49)\). Energy-dispersive x-ray spectroscopy (EDS) equipped in an FEI Talos F200X TEM was used as a supplemental method to characterize the elemental distribution at GBs in the as-deposited NT-2.9 specimen. EDS line scans showed that carbon and sulfur were uniformly distributed in the sample with no obvious segregation at the GBs (fig. S13).

Structural characterization
The crystal orientation of the as-deposited NT-Ni specimens was determined by XRD \((\text{Bruker D2})\). Plan-view and cross-section images of the sample microstructures were examined with a Tecnai G2 F20 transmission electron microscope operated at 200 kV. Plan-view orientation mapping was conducted using NanoMEGAS \((\text{Brussels, Belgium})\) hardware and the software package provided for the TEM system \((50)\). High-resolution TEM \((\text{HRTEM})\) observations were carried out using a Tecnai F30 and a double aberration-corrected Titan Cubed Themis G2 300 TEM operated at 300 kV. To prepare TEM samples in plan view, the deposited NT-Ni specimens were removed from the substrate and then mechanically polished to a thickness of ~15 μm, followed by ion milling at 173 K \((\text{Gatan 691})\).
For cross-section imaging, samples were prepared according to the following procedures: (i) electrodeposition of a protective CG-Ni coating on the surface, (ii) extraction of cross-section foils by wire cut, (iii) mechanical polishing to ~15 μm in thickness, and (iv) thinning via a precision ion polishing system (Gatan 691) at 173 K. TEM and HRTEM specimens were prepared after compression using FEI Scios Dual-Beam focused ion beam (FIB) operated at a voltage of 30 kV with the cross-section at a tilt angle of 52° relative to the Ga ion sputtering direction. The specimens were further milled using ion milling to remove possible damage from Ga beam.

**Internal stress measurement**

To measure the internal stress of NT-Ni specimens, the following procedures are performed. First, flat Ni foils with the thickness of 100 μm were chosen as the substrate. Before electrodeposition, one side of the foil was sealed by silicone sealant. Second, NT-Ni films of ~10-μm thickness with various twin thicknesses were electrodeposited on the other side of the substrate. Subsequently, the curvature of the film-substrate was measured with a surface profiler (KLA-Tencor P7).

The internal stresses in NT-Ni film were calculated using the classic Stoney equation (19)

$$
\sigma = \frac{1}{6} \left( \frac{E_s}{1-\nu_s} \right) \left( \frac{t_i^2}{t_f R} \right)
$$

where $E_s$ and $\nu_s$ are Young’s modulus and Poisson’s ratio of the substrate, respectively; $t_i$ is the substrate thickness; $t_f$ is the film thickness; and $R$ is the radius of curvature of the film-substrate structure. According to Stoney equation, the internal tensile stresses are calculated to be approximately 500, 340, 90, and 20 MPa for NT-Ni specimens with $\lambda = 2.9, 4.4, 16.5, $ and 81.0 nm, respectively.

**Mechanical characterization**

Micropillars of the NT-Ni specimens with ~1.3 μm in diameter were machined from the normal plane of the deposited material using FIB previously described. The diameter-to-height aspect ratio of each micropillar was 2 to 3, and the taper angle from the top to the bottom was less than 2°. Microcompression testing was performed using an Agilent Nano Indenter XP with a flat diamond punch tip. The load and a strain rate of $10^{-3}$ s$^{-1}$. At least three pillars were examined to verify the test data accuracy and scatter. The true stress and true strain were calculated using the method developed by Greer et al. (51)

$$
\sigma_t = \frac{PL_p}{A_0 L_0}
$$

$$
\varepsilon_t = \varepsilon_{el} + \varepsilon_{pl} = \frac{1}{E \frac{L_p}{L_0}} - \ln \left( \frac{L_p}{L_0} \right)
$$

where $P$ is the instantaneous load on the pillar; $L_0$ and $A_0$ are the height and cross-sectional area of the pillar, respectively; $L_p$ is the final height of the pillar; $\varepsilon_{el}$ and $\varepsilon_{pl}$ are the elastic strain and plastic strain, respectively; and $E$ is the elastic modulus of nickel. Here, $A_0 = \frac{1}{2} \pi D^2$, where $D$ is taken as the initial half-height pillar diameter, instead of the diameter at the top of pillar. The true stress-strain curves for NT-Ni specimens only include the data points obtained from the stages of elastic and plastic deformation before shear banding, while those corresponding to the highly localized deformation after shear banding are not shown. Hardness measurements on the as-deposited and annealed specimens were conducted using the Qness Q10A+ Micro-Hardness Tester with a load of 50 g and a dwell time of 10 s. At least 10 indentations were performed to verify the test data accuracy and scatter.

**Geometric phase analysis**

Strain distribution in the deformed region of NT-2.9 specimen was mapped by means of geometric phase analysis (GPA) based on the individual HRTEM image. The GPA, which is based on the formalism given in (52) and implemented in Gatan DigitalMicrograph as a plug-in, was used to calculate the in-plane components of the symmetric strain tensor, $\varepsilon_{ij}$, and the rigid-body rotation, $\delta_{ij}$. Strain maps were plotted with respect to an internal reference lattice by $g_1 = (\overline{111})$ matrix/twin and $g_2 = (200)$ matrix for the matrix and $g_1 = (\overline{111})$ matrix/twin and $g_3 = (\overline{111})$ twin for the twinned area using Lorentzian masks with a diameter of 0.4 nm$^{-1}$ (in reciprocal space). The maximum and minimum strains were set in the range from +10% to −10%, and the rotation angles were set from +5° to −5°.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at [http://advances.sciencemag.org/cgi/content/full/7/27/eabg5113/DC1](http://advances.sciencemag.org/cgi/content/full/7/27/eabg5113/DC1)

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