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The Influence of Temperature on the Strain-hardening Behavior of Fe-22/25/28Mn-3Al-3Si TRIP/TWIP Steels

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Abstract

The influence of temperature and stacking fault energy (SFE) on the strain-hardening behavior and critical resolved shear stress for twinning was investigated for three Fe–22/25/28Mn–3Al–3Si wt.% transformation- and twinning-induced plasticity (TRIP/TWIP) steels. The SFEs were calculated by two different methods, density functional theory and statistical thermodynamic modeling. The dislocation structure, observed at low levels of plastic deformation, transitions from "planar" to "wavy" dislocation glide with an increase in temperature, Mn content, and/or SFE. The change in dislocation glide mechanisms from planar to wavy reduces the strain hardening rate, in part due to fewer planar obstacles and greater cross slip activity. In addition, the alloys exhibit a large decrease in strength and ductility with increasing temperature from 25 to 200 °C, attributed to a substantial reduction in the thermally activated component of the flow stress, predominate suppression of TRIP and TWIP, and a significant increase in the critical resolved shear stress for mechanical twinning.

stress for mechanical twinning, and other temperature dependent factors which likely play a more dominant role are discussed.

1 Introduction

High-manganese transformation- and twinning-induced plasticity (TRIP/TWIP) steels exhibit an excellent combination of strength and ductility, owing in part to the formation of strain-induced martensite and/or mechanical twinning during deformation [1–4]. The martensite platelets and mechanical twins impede the motion of dislocations gliding on non-coplanar slip systems. As deformation increases, the volume fraction and number density of martensite platelets and/or mechanical twins also increases, resulting in a "dynamic Hall-Petch effect". These strain-induced secondary deformation mechanisms generally occur within specific ranges of the intrinsic stacking fault energy (SFE) and temperature [5,6]. Most studies on high Mn-TRIP/TWIP steels have focused on aspects of their room temperature (RT) deformation, including the role of: Mn content and SFE [7,8], crystallographic texture evolution and its influence on mechanical twinning [1], grain size [9,10], strain rate [11,12], macro alloying additions such as: Al [13], Si [14], Cr+N [15,16], and micro alloying additions such as Nb, Ti, and V [17].

The elevated temperature properties of high-Mn TRIP/TWIP steels from RT to approximately 400 °C are also of scientific and technological interest, in part due to the significant changes in deformation mechanisms that occur when varying the temperature. Several studies have focused on the influence of temperature on SFE, deformation mechanisms, and mechanical behavior [15,18–20]. Increasing the deformation temperature increases the SFE, stabilizes the austenite relative to the ε_{hcp} -martensite phase, and delays the onset or completely suppresses mechanical twinning [18], causing a significant reduction in both strength and ductility. Jung and De Cooman [20] investigated effects of temperature and Al content on the strain hardening and solid solution strengthening in Fe-18Mn-0.6C-XAl steels (X=0, 1.45, and 2.5 wt.%) from -60 °C to 140 °C. The authors reported that work hardening was suppressed with increasing temperature and Al content in each alloy, in part due to an increase in the SFE, which resulted in reduced mechanical twinning. The authors correlated the strain hardening behavior with SFEs calculated by a thermodynamic model [6,21]. Steinmetz et al. [18] recently developed a physics based constitutive model for low SFE metals which incorporates mechanical twinning, twin volume fraction, grain size, dislocation cell boundaries, SFE, and other relevant factors to predict the strain hardening behavior. Their model indicates that a critical stress and a critical dislocation density are necessary for mechanical twins to form, and that increasing the deformation temperature delays the onset of mechanical twinning by: 1) reducing the flow stress, 2) increasing the rate of dislocation annihilation by climb, and 3)

increasing the SFE, which increases the critical stress needed to initiate mechanical twinning. Wong et al. [22] extended and experimentally validated this dislocation density-based crystal plasticity model to incorporate both transformation-induced plasticity and twinning-induced plasticity. They showed that the model was capable of predicting the competition in the activation of the TRIP and/or TWIP deformation mechanisms at different temperatures as a function of the SFE.

Calculations of the elevated temperature SFE have been performed by thermodynamic modeling [6,12,18,21]. These models incorporate a term corresponding to the difference in Gibbs free energy of the facecentered cubic (FCC) and hexagonal close-packed (HCP) phases, as well as parameter for the FCC/HCP interfacial energy. Despite experimental evidence suggesting a significant negative temperature dependence [23], the interfacial energy parameter is often assumed to be a constant value, typically within the range of 8 to 12 mJ m^{-2} [7] and independent of temperature, primarily due to the complexity of experimentally determining this parameter at temperatures other than RT. Determination of the interfacial energy is typically indirect and entails the use of experimental SFE values measured at the appropriate temperature, as well as the difference in Gibbs free energy of the FCC and HCP phases computed by a thermodynamic model, which is sometimes incomplete regarding proper consideration of all the required entropy terms. In addition, the Néel temperature in some high Mn steels is near RT and magnetic contributions may also have influence and complicate efforts to calculate the SFE. As a result, elevated temperature SFEs computed by thermodynamic model may be subject to significant uncertainty. Alloy design procedures for applications involving high temperature behavior of TWIP steels will benefit from an improved understanding of the temperature dependence of the SFE [4]. A recent trend includes incorporation of density functional theory calculations to predict changes in SFE and interfacial energy with temperature [4,24–27].

The aim of the present study is to investigate the effects of temperature and SFE on the deformation behavior of three model Fe-22/25/28Mn-3Al-3Si wt.% alloys from RT to 400 °C, hereafter referred to as the 22Mn, 25Mn, and 28Mn alloys. Of particular interest is the effect of SFE and temperature on the dislocation structure at different levels of strain and the critical stress for mechanical twinning. The SFEs in this investigation are calculated by two independent methods, thermodynamic model and DFT, for correlation to strain hardening behavior. Systematic variation of Mn results in significant changes to the SFE albeit with minimal solid solution hardening [2] (relative to other common alloying elements such as Al, Si and C [2,14,20,28]), enabling easier deconvolution of the effects of SFE and temperature on the deformation behavior.

2 Experimental Procedure

The 22Mn, 25Mn, and 28Mn alloys were induction melted in an Ar atmosphere and cast into ingots. Ascast ingots were thermo-mechanically processed by hot rolling at 1100 °C to produce strips of 3 mm thickness and subsequently by cold rolling to 1.5 mm thickness. The resulting sheet was recrystallized at 900 °C for 30 min, yielding a microstructure with equiaxed grains with average size of 21 µm for each composition, with many grains containing annealing twins. The measured compositions are listed in Table 1.

Table 1 – Chemical compositions of the steels in wt.% unless otherwise specified.										
Designation	Material	Mn	Al	Si	С	O (ppm)	Fe			
22Mn	Fe-22Mn-3Al-3Si	22.2	2.76	2.92	0.0093	<5	Bal.			
25Mn	Fe-25Mn-3Al-3Si	24.7	2.66	2.95	0.0053	<5	Bal.			
28Mn	Fe-28Mn-3Al-3Si	27.5	2.74	2.89	0.0071	<5	Bal.			

Table 1 Chamical compositions of the steals in wt % u

Sub-sized flat tensile specimens with a 20 mm gauge length and 5 mm width were cut from the sheet in the direction parallel to the rolling direction using electro-discharge machining (EDM). Specimens of each alloy were strained in tension at 25 °C, 100 °C, 200 °C, 300 °C, and 400 °C at a rate of 4 x 10⁻⁴ s⁻¹. Tests at 25 °C and 400 °C were interrupted at 0.03, 0.1, 0.18, 0.34, 0.44 and 0.47 plastic true strain for microstructural characterization by light optical microscopy (LOM) and transmission electron microscopy (TEM). Specimens for LOM were prepared by standard metallographic procedures, with an automatic polishing system. Low loads were used during this process, and successively decreased from 25 N to 10 N for the final polish. The final polish utilized a suspension of $0.05 \,\mu m$ colloidal silica for chemical-mechanical polishing to ensure a flat surface with minimal deformation due to metallographic preparation. A 10% Nital solution was used to etch the polished specimens. For TEM, disks 3 mm in diameter were cut from the gauge length of specimens deformed to 0.03, 0.1, 0.18 and 0.34 true strain using EDM. The 3-mm disks were mechanically polished to approximately 100 μ m thickness and then jet electro-polished to electron transparency with a TenuPol-5 using a solution of 70% methanol and 30% nitric acid at -30 °C. Microstructures were analyzed with a Philips CM20 TEM operated at 200 kV.

Sample preparation for X-ray diffraction (XRD) experiments involved mechanical polishing through 1200 grit SiC paper and a final chemical polishing to remove the damaged surface layer (samples were dipped into baths of hydrofluoric acid, hydrogen peroxide, and de-ionized water in a 1:50:50 mixture, respectively for approximately 5 min). The temperature-dependent XRD patterns were collected using an X-ray diffractometer (PANAlytical X'Pert MPD Pro) with Cu K α radiation ($\lambda_{K\alpha 1} = 1.54060$ Å and $\lambda_{K\alpha 2} = 1.54443$ Å), equipped with an Anton Paar XRK-900 heating stage and an Oxford PheniX Cryogenic Stage. Prior to each XRD measurement at each temperature, 3 minutes of waiting time was set for temperature equilibration. The 2θ scanning range, step size and scan rate for each measurement was 30 ° to 120°, 0.0167113° and 0.107815 deg/s, respectively. The XRD measurements which used the heating stage were performed in an Ar environment from 25 °C to 600 °C and the measurements which used the cryogenic stage were conducted under vacuum from -250 °C to 25 °C. The first five allowable austenite peaks were identified for each XRD scan. Austenite lattice parameters were calculated using Bragg's law and the Nelson-Riley function to account for larger sensitivities of the lattice constant at higher

diffraction angles [29]. Specifically, the angle of every K α 1 diffraction peak was fit at full width half max using PAN Analytical Data Viewer software and a linear regression of calculated lattice constants was used to find the intercept of all diffraction angles for a given alloy and temperature.

3 Results

3.1 X-ray Diffraction and Stacking Fault Energy Calculation

This section discusses lattice parameter measurements and SFE calculations. The DFT SFE calculations used lattice parameter measurements as inputs. Figure 1 shows the lattice parameter increases with Mn content (also see Figure S 1). The lattice parameter decreases approximately linearly with temperature from 600 °C to 200 °C (Figure 1, dashed green line) but non-linearly below about 200 °C. The values of instantaneous coefficient of thermal expansion (CTE) are similar in the range of 250 °C to 550 °C (Figure S 2, dashed green box) but decrease significantly with decreasing temperature below 250 °C in each alloy. According to recent work [30], the lattice parameter of high Mn Fe-Mn-C austenitic steels is significantly less sensitive to temperature in the antiferromagnetic state (below the Néel temperature) relative to the paramagnetic state. Estimates of the Néel temperatures of the present 22, 25, and 28Mn steels by empirical relationships yield -6, 9, and 25 °C [7], respectively, suggest the temperature dependence of the CTE may in part be associated with magnetic affects and the antiferromagnetic to paramagnetic transition.



Figure 1 - Austenite lattice parameter determined from XRD measurements of the 22, 25, and 28%Mn alloys plotted as a function of temperature. Error bars are ± 0.001 Å.

The stacking fault energy is obtained using the axial next nearest neighbor Ising (ANNNI) model, where only energies of HCP and FCC bulk structures are required. Since the main goal of the DFT calculations is to capture the complex chemical composition and the magnetically disordered state, the exact muffin tin orbital (EMTO) method is employed in combination with the local density approximation (LDA) of the electronic structure [31], for each of the phases. The shortcoming of this approach is that it does not give direct access to the vibrational entropy. Instead, the influence of temperature is studied by enlarging the lattice constant (thermal expansion) and by considering magnetic and configurational contributions to the entropy. The austenite lattice constant measured by XRD at a given temperature is also used in the ab initio calculations to calculate the magnetic moment at a given temperature. Since the Néel temperature is below 25 °C and our focus is on room temperature and above, the magnetism in the DFT calculations is considered in the disordered local moment (DLM) [32–34] approach for all three alloys. Explicit supercell calculations would give access to local relaxations of atomic positions and magnetic moments, but previous calculations for ternary systems (Fe-Mn-Al, Fe-Mn-Si) [35] indicated that the corrections are small. The temperature-dependent magnetic moment $[\mu(V(T))]$ yields magnetic entropy (see Equation (1)) and Figure 2a) and is used in the ab initio free energy calculations. Figure 2a shows the relationship between lattice constant and magnetic moment for Mn and Fe atoms arranged in FCC and HCP structures. Ab initio free energies are used to compute a SFE based on the difference between HCP bulk energy per atom at the meta-stable FCC lattice constant and the FCC bulk energy per atom at the stable FCC lattice constant (see Figure 2b). Since all curves for a given alloy were nearly the same in character, the SFE of only one alloy (22Mn) is presented for clarity in Figure 2b.

$$S_{mag.} = k_B \ln(1 + \mu(V)) \tag{1}$$



Figure 2 - (a) The magnetic entropy contribution to SFE based on crystal structure and elemental contribution for a given lattice constant. (b) Energy per atom contributions calculated at -273 °C (0 K) for the Fe-22Mn-3Al-3Si alloy using DLM-ANNNI calculations. A gray shading was placed to highlight the range of lattice constants measured by XRD.

Figure 3 shows the SFE of the 22Mn, 25Mn, and 28Mn calculated as a function of temperature as calculated by this DFT approach. The result is compared to an empirical thermodynamic model developed in previous work [7]. Both methods of calculation consistently indicate a positive dependence of the SFE on temperature. In contrast to the example of Ni, where such an agreement has not been achieved [36], this gives confidence to the quality of the empirical model. However, the magnitude of the temperature dependence is significantly different. DFT predicts the SFE increases on average by 0.041 mJ m⁻² °C⁻¹ over the range of 25 to 400 °C for all three alloys, while the thermodynamic model predicts the SFE increases by approximately 0.18 mJ m⁻² C⁻¹. Each method has various uncertainties that may account for some of these differences. Most importantly, the DFT approach incorporates experimentally measured lattice constants to account for the temperature dependence of the SFE, but the vibrational entropy contribution to the SFE was not considered, while it has shown to be a significant contributor to the SFE in pure FCC metals such as Al, Cu, and Ni [36]. Nonetheless, the increase in SFE with temperature determined by DFT is relatively close to that of 0.063 mJ m⁻² C⁻¹ reported for an Fe-20Mn-4Cr-0.5Si-0.5C wt.% alloy determined by experimental observations of changes in the geometry of extended dislocation nodes with temperature [37]. The thermodynamic model has been added because it contains the vibrational entropy implicitly, but in this case a primary source of uncertainty is the interfacial energy. The thermodynamic model uses interfacial energies, determined at RT, that are assumed to remain constant with changes in temperature [7]. However, some combined experimental and thermodynamic approaches have suggested that the interfacial energy parameter is in fact dependent on temperature, and may even decrease

significantly with increasing temperature in the Fe-Mn system [23]. The latter suggests the temperature dependence of the SFE calculated by the thermodynamic model using constant interfacial energy may overestimate the actual temperature dependence in the Fe-Mn-Al-Si system, although further investigation would be required. In an attempt to overcome uncertainty in the interfacial energy parameter, some authors have used a hybrid approach [18], using DFT to calculate the FCC/HCP interfacial energy term and the standard thermodynamic model to calculate the chemical and magnetic contributions to the difference in Gibbs free energy of the FCC and HCP phases, $(\Delta G_{chem}^{fcc \rightarrow hcp} + \Delta G_{Mag}^{fcc \rightarrow hcp})$. The SFE of the Fe-22Mn-0.6C wt.% steel, displayed as the black line in Figure 3, determined using the hybrid approach, exhibits a similar temperature dependence to values calculated by thermodynamic model in the present work. The latter approach has also been used to calculate the SFE of high Mn TWIP steels as a function of Mn content at RT [25]. Finally, it is clear from the data in Figure 1 and Figure S 2 that anti-ferromagnetic ordering begins to influence the temperature dependence of the lattice parameter below approximately 200 °C. In the thermodynamic model [7], the magnetic contribution to the Gibbs energy of a phase is described by the model proposed by Inden [38] as modified by Hillert and Jarl [39]. The model results in a gradual reduction in the magnetic contribution to the Gibbs energy above the calculated Néel temperatures, which in this case are 25 °C or below. In the model, the magnetic contribution to the Gibbs energy is significant up to approximately 100 °C, and remains present up to about 200 °C, which is qualitatively consistent with the apparent range of temperature that the magnetic contributions influence the lattice parameter, as shown in Figure 1. It is also notable that the significant differences in temperature dependencies of the SFE values calculated by different methods, DFT and thermodynamic model, shown in Figure 3, are useful in clarifying the role of SFE and its relative importance on specific phenomena related to the activation of mechanical twinning at elevated temperatures, namely the critical resolved shear stress for mechanical twinning, which is discussed further in section 4.3.



Figure 3 - SFEs for the Fe-22/25/28Mn-3Al-3Si alloys calculated by thermodynamic model [7] and DFT. The SFEs of an Fe-22Mn-0.6C calculated by Steinmetz et al. [18] and an Fe-20Mn-4Cr-0.5S-0.5C steel experimentally measured by Remy [37] are provided for comparison.

3.2 Microstructural Evolution

3.2.1 Optical Microscopy

Figure 4a, b, c, and d show optical micrographs of the 28Mn alloy deformed to 0.1, 0.18, 0.34, and 0.44 true strain at room temperature, respectively. Mechanical twinning is the dominant secondary deformation mechanism in this alloy at RT [8] and the etched specimens reveal bundles of mechanical twins. After 0.1 true strain at RT, mechanical twinning is only apparent from the optical micrographs in a small percentage of grains, indicated by red arrows. After 0.18 strain at RT, most grains exhibited evidence of primary mechanical twinning occurring on a single slip system while a relatively small percentage of grains displayed mechanical twinning activity in two independent slip systems, indicated by blue arrows in Figure 4b. In specimens deformed to 0.34 true strain, nearly all grains exhibited mechanical twinning and many show mechanical twinning on multiple slip systems and the grains themselves are noticeably elongated in the direction of tensile deformation. In contrast to the RT deformation, no evidence of secondary deformation mechanisms is observed after 0.1 and 0.34 plastic true strain at 400 °C in the 28Mn alloy, as shown in Figure 5a and b, respectively. However, slip traces are

observed in Figure 5b (indicated by red lines), but these features are more diffuse and less well defined compared to the mechanical twins observed at 0.34 true strain at RT (Figure 4c).



Figure 4 - Optical micrographs of the 28Mn alloy after deformation to (a) 0.1, (b) 0.18, (c) 0.34, and (d) 0.44 plastic true strain at RT (the horizontal of the image is along the rolling and tensile directions). The red and blue arrows indicate mechanical twinning activity in single and multiple slip systems, respectively.



Figure 5 - Optical micrographs of the 28Mn alloy after deformation to (a) 0.1 and (b) 0.34 true strain at 400 °C (the horizontal of the image is along the rolling and tensile directions). The red arrows indicate slip traces.

3.2.2 TEM - 0.03 true strain – RT and 400 $^{\circ}$ C

Figure 6a and b show representative bright-field (BF) TEM micrographs of the dislocation structure of the 22Mn alloy after 0.03 true strain at RT from two different grains. The micrographs reveal a planar dislocation structure with relatively large stacking faults bounded by partial dislocations. The features are present on two different slip systems. The extension of the stacking faults typically ranges between 50 nm to greater than 1 μ m, which is one to two orders of magnitude greater than the equilibrium spacing of 5 to 16 nm, measured previously [7]. In many cases, a leading or trailing partial dislocation is located several hundred nm from other defects, as illustrated by green arrows in Figure 6a. However, the leading or trailing partial dislocations are often impeded by the stacking faults on a non-coplanar slip system, as indicated by red arrows, and in some cases, the stacking faults are intersected, revealing direct evidence that the leading partial has penetrated the stacking fault on the non-coplanar slip plane (indicated by blue arrows in Figure 6a). Qualitatively, these observations suggest that the stacking faults act as impediments to dislocations gliding on non-coplanar slip planes in this alloy (22Mn) at RT and at low strains. Figure 6c and d show the dislocation structure after deformation to 0.03 plastic true strain at 400 °C, revealing a tangled dislocation structure with significant curvature. While the majority of dislocations reveal no extension in the BF micrographs, some dislocations do exhibit some extension (see orange arrows), typically around 100 nm or less, but the extension observed after deformation at 400 $^{\circ}$ C is much smaller than the extension observed after this strain at RT.



Figure 6 – BF TEM micrographs of the 22Mn alloy after deformation to 0.03 true strain at (a,b) RT and (c,d) 400 °C. See text for explanation of arrows.

The BF images in Figure 7 show the dislocation structure of the 28Mn alloy strained to 0.03 true strain at RT (a and b) and 400 °C (c and d). The dislocation structures after deformation at RT and 400 °C are similar, exhibiting tangled dislocations with significant curvature which are primarily constricted, except for a few dislocations that show a minor amount of extension (indicated by orange arrows). It is important to note that the dislocation structure in the 28Mn alloy after deformation to 0.03 true strain at RT and 400 °C are similar to the dislocation structure of the 22Mn after deformation to 0.03 true strain at 400 °C (Figure 6c and d), but considerably different than that of the 22Mn after RT deformation to 0.03 true strain.



Figure 7 - BF TEM micrographs of the 28Mn alloy after deformation to 0.03 true strain at (a,b) RT and (c,d) 400 °C. The orange arrows indicate extended dislocations.

3.2.3 TEM - 0.1 to 0.34 true strain – RT, 200 °C, and 400 °C

The microstructural evolution from 0.1 to 0.34 plastic true strain at RT for the 22Mn, 25Mn, and 28Mn alloys is reported in detail in previous work [8] and only summarized here. The prior work shows the RT microstructure is significantly influenced by Mn content and strain. The 22Mn alloy exhibits planar dislocation glide, suppression of dislocation cross slip, and ε - and α -martensite transformation as the dominant secondary

deformation mechanisms at RT, with the formation of thin ε -martensite lath structures on multiple slip systems forming from the nearly the beginning of plastic deformation. In contrast, the 28Mn alloy exhibits more frequent cross slip and wavy dislocation character, strain induced ε - and α -martensite transformation is predominately suppressed, and mechanical twinning begins to impact the strain hardening behavior at approximately 0.1 plastic true strain. Mechanical twinning generally occurs in TWIP steels first in grains oriented favorably with respect to the tensile axis [9]. In the 25Mn alloy, both ε -martensite formation and mechanical twining are present at plastic true strains of 0.1 and above at RT. The BF TEM images in Figure 8a, c, and e depict the microstructure of the 25Mn alloy after deformation to 0.1, 0.18, and 0.34 true strain at RT and are compared to micrographs of the dislocation structure obtained at 400 °C (Figure 8b, d, and f). The BF TEM image in Figure 8a, recorded a few degrees of the [110] zone-axis, shows extended dislocations along with a bundle of mechanical twins on a single slip system oriented diagonally on the image, as confirmed by the selected-area electron diffraction (SAED) pattern in the inset recorded on the [110] zone-axis. The BF TEM image shown in Figure 8c was also recorded slightly off the [110] zone-axis in a two-beam condition, revealing the presence of ε -martensite lath structures in the edge on orientation (aligned approximately parallel to the horizontal of the image) and extend dislocations gliding on a non-coplanar slip system. The BF TEM image in Figure 8e was recorded slightly off the [111] zone in a two-beam condition, showing a high density of dislocations and further evidence of ε -martensite, with $(0001)\varepsilon$ parallel to the beam.

BF TEM micrographs of the dislocation structure of the 25Mn alloy deformed at 400 °C to 0.1, 0.18, and 0.34 true strain are shown in Figure 8b, d, and f, respectively. The dislocation density progressively increases with increasing strain and the dislocations in the 25Mn alloy appear predominately constricted in BF imaging. No evidence of secondary deformation mechanisms was observed in the 25Mn alloy at these strains at 400 °C. The dislocation structure of the 22Mn alloy appeared similar except that some dislocations were extended. No evidence of secondary deformation mechanisms was observed after deformation of the 22Mn alloy at 400 °C. The TEM imaging suggests a complete suppression of secondary deformation mechanisms in these alloys at 400 °C. As such, further TEM examination was conducted on the 22Mn alloy after deformation at the intermediate temperature of 200 °C to maximum uniform elongation (0.37 plastic true strain). The high density of dislocations at this level of strain makes a clear interpretation of the dislocation substructure difficult. Nonetheless, a relatively small amount of mechanical twins are observed in some grains at this strain, such as those depicted in Figure 9, but no evidence of ε -martensite from SAED patterns. This observation suggests that the type of secondary deformation mechanisms occurring in the 22Mn alloy changes from α/ε -martensite formation at RT to mechanical twinning as the temperature is increased to 200 °C, but also that the intensity of secondary deformation mechanisms is reduced at the higher temperature.



Figure 8 - BF TEM micrographs of the 25Mn alloy after deformation to 0.1 (a,b), 0.18 (c,d), and 0.34 (e,f) plastic true strain at (a,c,e) RT and (b,d,f) 400 $^{\circ}$ C.



Figure 9- BF TEM micrographs of the 22Mn alloy recorded a few degrees off the [110] zone showing a relatively high dislocation density and mechanical twinning after deformation to 0.37 plastic true strain at 200 °C.

3.3 Strain Hardening Behavior

The temperature dependence of the strength (yield and ultimate tensile) and ductility (uniform and total elongation) are shown in Figure 10a and b, respectively. Increasing the test temperature from 25 °C to 100 °C results in a substantial drop in the yield and ultimate tensile strengths of ~65 and 125 MPa, respectively, in the three alloys. The drop in strength is accompanied by a moderate decrease in ductility in the case of the 22Mn and 25Mn alloys and a more intense decrease for the 28Mn alloy. Further increase in the test temperature from 100 °C to 200 °C causes additional significant decreases in the mechanical properties. The values for strength and ductility were observed to be similar, regardless of alloy, for testing temperatures of 200, 300 and 400 °C.



Figure 10 - (a) 0.2% offset yield strength and UTS and (b) maximum uniform (solid lines) and failure elongation (dashed lines). All graphs present data from 25 °C to 400°C. The error bars in (a) are approximately the size of the symbols. The RT data in (a) and (b) as well as the elevated temperature yield strength data in (a) are from [8] and are shown here for comparison.

Figure 11a, b, and c show the true stress and strain hardening rates ranging from 25 to 400°C vs. plastic true strain for the 22Mn, 25Mn and 28Mn alloys, respectively. Using Considère's instability criterion, RT values of the true ultimate tensile strength of 1172, 1136, and 1106 MPa and true strain values at the onset of necking of 0.54, 0.57, and 0.56 were determined for the 22Mn, 25Mn, and 28Mn alloys, respectively. The three alloys display multi-stage strain hardening behavior during tensile deformation. The 22Mn alloy exhibits three distinct strain hardening plateaus at RT, which are discussed along with additional details regarding the RT strain hardening behavior of the three alloys in [8].

Figure 11 shows that increasing the test temperature from 25 °C to 100 °C significantly reduces the strain hardening in the range of true strain from 0 to 0.1 for the 22Mn and 25Mn alloys compared to a relatively small decrease in the 28Mn alloy (denoted by " Δ (d σ /d ϵ)" in Figure 11a, b, and c). At 100°C, the strain hardening rate curves of the 22Mn, 25Mn, and 28Mn experience an inflection at true strains of approximately 0.23, 0.25, and 0.27, respectively, indicated by purple arrows in Figure 11. These inflection points likely correspond to the true strain and true stress at which mechanical twinning begins to influence tensile deformation for each alloy at 100°C. At test temperatures of 200°C and above, the strain hardening rate curves also experience an inflection, but downward in this case, resulting in a significantly faster decrease of the strain hardening rate with further strain relative to 100 °C.



Figure 11 - True stress and strain-hardening rate vs true strain for the (a) 22Mn, (b) 25Mn and (c) 28Mn alloys. All graphs present data from 25 °C to 400°C. The RT strain-hardening data in (a), (b) and (c) are from [8] and are shown here for comparison to the elevated temperature data.

4 Discussion

4.1 Influence of Temperature on Microstructure and Strain Hardening Behavior – 0 to 0.1 True Strain

The strain hardening rate in stage 1 exhibits a rapid decrease of about 1000 MPa with increasing plastic true strain from 0 to approximately 0.1 in each alloy and at all test temperatures. The strain hardening rate in this stage is typically strongly influenced by dynamic recovery processes (e.g., cross slip, dislocation annihilation, and the formation of low-energy dislocation structures [40–43]), which are in turn influenced by temperature, SFE, and the lattice friction stress [44], among other factors. Cross slip enables screw dislocations to cross to planes more favorably oriented for slip, to avoid obstacles, and also facilitates the annihilation of screw dislocations [40], which increases dynamic recovery and lowers the strain-hardening rate.

The strain hardening rates at RT in the strain range of 0 to 0.1 true plastic strain are greatest in the 22Mn alloy but decrease with increasing Mn content as shown in Figure 11. The decrease in strain hardening rates with increasing Mn content is primarily attributed to changes in the mechanisms of dislocation glide, as illustrated in representative TEM images of the 22 and 28Mn samples deformed to 0.03 plastic true strain (Figure 6 and Figure 7, respectively). The 22Mn alloy deformed at RT to 0.03 true strain exhibits dislocation slip active in one or two slip systems in all grains. The dislocations exhibit a large degree of extension (separation of partial dislocations), which is characteristic of a low SFE, and limits dislocation cross slip. The extended dislocations also act as planar obstacles to dislocations gliding on non-coplanar slip systems, (Figure 6a and b).

The wavy dislocation structure in the 28Mn alloy (Figure 7a and b) suggests greater dislocation mobility and more favorable cross slip. As previously noted [8], the friction stress in these alloys is likely to be similar at a given temperature, as Mn tends to have a relatively small influence on solid solution strengthening in austenitic TWIP steels [2]. Therefore, the large increase in SFE from the 22Mn to the 28Mn alloy, from 15 to 39 mJ m⁻², is likely the primary cause of changes in dislocation structure, greater propensity for cross slip, and decrease in the strain hardening rate at RT from 0 to 0.1 plastic true strain. Finally, the dislocation structure of the 25Mn alloy after deformation to 0.03 plastic true strain exhibits both planar and wavy characteristics, described in more detail in [8].

With increasing temperature from RT to 100 °C, the strain-hardening rates (in the range of 0 to 0.1 true strain) decrease by a relatively large amount in the 22Mn alloy, more moderately in the 25Mn alloy, while the decrease in the 28Mn alloy is limited, as shown by $\Delta(d\sigma/d\varepsilon)$ indicators in Figure 11. Interestingly, further increases in temperature from 100 to 400 °C have no significant influence on the strain hardening rates (from 0 to 0.1 true strain) of the three alloys. TEM micrographs of the 22Mn alloy after deformation to 0.03 true strain at 400 °C, Figure 6c and d, show microstructures comprised primarily of dislocations exhibiting no observable extension (in BF imaging mode). Some of these dislocations exhibit large curvature and display a relatively small amount of extension (some of which are indicated by orange arrows), typically of no more than 250 nm. This dislocation structure differs significantly from the highly planar dislocation structure observed in the 22Mn alloy deformed at the same value of true strain at RT. The latter suggests that the 22Mn alloy transitions from planar dislocations gliding on non-coplanar slip systems, to wavy dislocation glide at 400 °C, where dislocations are largely constricted and able to cross slip to avoid obstacles. This transition in dislocation glide mechanisms may be largely responsible for the concomitant reduction in strain hardening rate with increasing temperature in the 22Mn alloy.

Unlike the 22Mn alloy, the dislocation structures of the 28Mn alloy after 0.03 true strain at RT (Figure 7a and b) and 400 °C (Figure 7c and d) are quite similar, with the majority of dislocations displaying no observable extension and some with a high degree of curvature. In addition to similar dislocation structures after 0.03 true strain at RT or 400 °C, the strain hardening rates from 0 to 0.1 true strain at these temperatures are also nearly identical, despite a large increase in temperature from RT to 400 °C, and a concomitant increase in SFE above 39 mJ m⁻² at RT, as illustrated in Figure 3. This observation reinforces the above conclusion that changes in the mechanisms of dislocation glide (planar, wavy, or a mix of the two) may significantly influence the strain-hardening behavior at low strains, and in the absence of such a change in the 28Mn alloy with increasing temperature, the strain hardening rates remain similar in the strain range of 0 to 0.1.

4.2 Influence of Temperature on Microstructure and Strain Hardening Behavior – 0.1 True Strain to Failure

This section is structured to first discuss aspects of the strain hardening behavior in the range of 200 $^{\circ}$ C to 400 °C where secondary deformation mechanisms are limited, then 100 °C where secondary deformation mechanisms are clearly active, and then finally RT where intense activity of secondary deformation mechanisms occurs during straining. The yield strength, UTS, and elongation in the range from 200 °C to 400 °C exhibit minimal differences over this temperature range between the three alloys, as shown in Figure 10. The yield strength of these alloys can be approximated from the athermal and thermal contributions [8], with the thermally activated contribution only becoming significant as temperatures are decreased below approximately 200 °C. Above 200 °C, the athermal components, comprised mainly of solid solution strengthening and Hall-Petch strengthening [20,45], dominate and exhibit only weak temperature sensitivity. The strain hardening rate and flow stress for each alloy at each temperature from 200 °C to 400 °C are also nearly identical (Figure 11a, b, and c). The strain hardening rates in this range of temperature can be approximated by 4 different stages: stage 1 occurring from the onset of plastic deformation to ~ 0.1 true strain, stage 2 from ~ 0.1 to ~ 0.25 true strain, stage 3 from ~ 0.25 to 0.375 true strain, and stage 4 from ~ 0.375 true strain to failure. These stages are labeled on Figure 11b for the 25Mn alloy. Stage 1 is characterized by a sharp decrease in the work hardening rate, which as explained before is associated with dynamic recovery processes [8,40–43] including cross slip, the formation of low energy dislocation structures like cells and tangles, and also the transition from elastic to plastic deformation [19]. In stage 2, the work hardening rate continues to decrease but at a significantly slower rate relative to stage 1. The dislocation density increases significantly during deformation in this stage at 400 °C, as shown for the 25Mn alloy at true strains of 0.1 and 0.18 in Figure 8b and d, respectively, and no evidence of secondary deformation mechanisms are observed for the three alloys. The onset of stage 3 at temperatures of 200 °C to 400 °C coincides with another inflection in the strain hardening rate, followed by a more significant decrease in this parameter than observed in stage 2. While work hardening continues to occur in stage 3, and the dislocation density continues to increase, as can be observed qualitatively by comparing Figure 8d and f, the reduced hardening rate suggests that the microstructures ability to trap additional dislocations is becoming weaker, dynamic recovery is becoming enhanced, but the initiation of secondary deformation mechanisms that can enhance and prolong work hardening is not favorable at this stress level and temperature. Finally, stage 4 is characterized by an additional inflection and significant further decrease in the strain hardening rate until failure, indicating the microstructure has lost the capacity to further harden. It should be noted that mechanical twinning was generally suppressed in the range of temperature from 200 to 400 °C, with the exception of a small minority of grains in the 22Mn alloy that exhibited twinning after deformation to maximum uniform elongation (0.37 true strain and a true stress of 708 MPa, Figure 9), and the twinning in these grains was confined only to a single slip system.

Reducing the deformation temperature from 200 °C to 100 °C causes remarkable changes in the work hardening behavior near 0.25 true strain. At 100 °C, the three alloys exhibit continued and sustained strain hardening well after ~0.25 true strain, which differs from the behavior at 200 °C and greater, where the work hardening rate dramatically decreases after this level of true strain. This region of continued strain hardening at 100 °C is labeled stage 3a in Figure 11b. The specific onset of stage 3a (see purple arrows in Figure 11) occurs at ~0.23, 0.25, and 0.27, for the 22Mn, 25Mn, and 28Mn alloys, and is caused in part by the influence of mechanical twinning on the strain-hardening rate [18]. The true stress at these levels of strain at 100 °C are 583 MPa, 583 MPa and 625 MPa, respectively. The SFEs calculated by DFT of the 22 and 25Mn alloys are similar at 19 and 24 mJ m⁻² at 100 °C (Figure 3), respectively, while the SFE of the 28Mn alloy is considerably greater at 42 mJ m⁻². The temperature dependence of the SFE calculated by DFT is similar to the measured temperature dependence reported in Ni and Cr containing austenitic stainless steels [46] but different than the SFE values calculated by thermodynamic model in the present work, which predicts SFE values of 28, 35, and 51 mJ m⁻² at 100 °C, respectively. Since Mn mainly affects the strain hardening indirectly through changes in SFE, the increase in SFE with Mn content at 100 °C, calculated by both DFT and thermodynamic model, is predominately responsible for the modestly higher stress levels at which mechanical twinning begins to significantly influence the strainhardening behavior in the 28Mn alloy.

Further reducing the deformation temperature to 25 °C causes the influence of secondary deformation mechanisms on the strain hardening rate to occur at even lower stresses. For instance, the influence of mechanical twinning on the strain hardening rate in the 28Mn alloy is apparent at ~0.1 true strain and a true stress of ~448 MPa (see Figure 4a and Figure 11c) near the transition from stage 1 to 2. Mechanical twinning is also present in the 25Mn alloy near 0.1 true strain in conjunction with ε-martensite formation [8].

4.3 Critical Stress for Mechanical Twinning

This section reviews prior work on determination of the critical stress for mechanical twinning and evaluates potential phenomena that influence the temperature dependence of the critical stress in the present steels. The critical stress for mechanical twinning significantly influences the deformation behavior of TWIP steels as reported in the literature [4,18,47] and shown in the previous section. In predicting the strain hardening behavior of TWIP steels, Steinmetz *et al.* [18] calculated the critical resolved shear stress for twinning, τ_{T} , from Equation (2) for the Fe-22Mn-0.6C TWIP steel, assuming the twin nucleus is the "three layer fault" model proposed by Mahajan and Chin [48]:

$$\tau_{\rm T} = \frac{\gamma}{3b_{\rm p}} + \frac{3Gb_{\rm p}}{L_0} \tag{2}$$

where γ is the SFE, b_p is the magnitude of the partial dislocation Burger's vector, G is the shear modulus, and L_0 is the width of the sessile partial dislocations between pinning points forming the twin embryo [18]. Steinmetz et al. [18] calculated temperature dependent SFE values and an "optimal" L_0 of 260 nm for prediction of τ_T in the Fe-22Mn-0.6C steel. In the model, L₀ is constant with evolution of the microstructure and temperature, but the authors did note that other researchers suggest L_0 may be related to the forest dislocation density and mean free path [49], which is an important point and discussed further below. The calculated values of τ_{T} , at 25 °C, 100 °C, and 200 °C reported by Steinmetz et al. for the Fe-22Mn-0.6C steel [18] are 192 MPa, 203 MPa, and 227 MPa, respectively, as shown Table2, and match well with the manifestation of mechanical twinning in the strainhardening rate obtained from compression tests. In the model of Steinmetz et al. [18], increasing the deformation temperature results in an increase in SFE, which in turn increases $\tau_{T,}$, requiring higher normal stresses to initiate mechanical twinning. The aforementioned fact, combined with decreasing flow stress as temperature increases, significantly delays the onset of mechanical twinning and can result in reduced strain hardening at elevated temperatures. The "three layer fault" model has also been reported to be the dominant mechanism for nucleation of primary twinning at RT by Mahato et al. [47] in an alloy of composition Fe-27Mn-2.5Si-3.5Al wt.%, which is similar in composition to the present alloys. Mahato et al. [47] estimated L₀ to be 200 nm at RT, based on microstructural observations, and calculated a τ_{T} of 201 MPa using Equation 2.

In the present study, the stress for the onset of mechanical twinning (σ_i) was estimated from both microstructural observations of mechanical twinning and/or the inflection point on the strain hardening rate curve, whose appearance is the initial manifestation of mechanical twinning in the present steels and other types of TWIP steels, such as the Fe-22Mn-0.6C wt.% alloy [18]. Figure 11c shows the inflection point on the strain hardening rate curve associated with the initiation of twinning occurs at ~0.1 true strain and 448 MPa in the 28Mn alloy at RT, and evidence of mechanical twinning was clearly observed in the microstructure of samples deformed at this strain by LOM and TEM (Figure 4a and [8], respectively). This indicates that mechanical twinning in this alloy initiates before a stress level of 448 MPa is reached. However, TEM observation (Figure 7a and b) revealed no evidence of mechanical twinning in the 28Mn alloy at 0.03 true strain at a corresponding stress level of 326 MPa. As such, the critical stress (σ_T) for the onset of mechanical twinning at RT in the 28Mn alloy, which occurs between 326 MPa and 448 MPa, is taken as the intermediate value of 387 with an uncertainty of ±61 MPa. A similar approach was utilized to estimate σ_T for onset of mechanical twinning in all three alloys at 100 °C, yielding 522 ±61, 522 ±61 and 564 ±61 MPa for the 22, 25, and 28Mn alloys, respectively, and the 22Mn alloy at 200 °C, 647 ±61 MPa. These values are shown in Table 2. The error associated with these values is due to the uncertainty in identifying the exact onset of mechanical twinning.

The critical resolved shear stress for mechanical twinning (τ_T) is calculated from the following equation:

$$\tau_T = \sigma_T m \tag{3}$$

where the Schmid factor m is assumed to have a value of 0.47 [28], which represents the maximum possible value for mechanical twinning. Mechanical twins typically nucleate first in grains with high Schmid factors for twinning [9]. The values of τ_T are listed in Table 2. At RT, a value of 182 ±29 MPa for τ_T was determined experimentally in the 28Mn alloy, which is similar to the value of 201 MPa calculated for the Fe-27Mn-2.5Si-3.5Al wt.% alloy by Mahato et al. [47]. The dependence of τ_T on temperature and SFE is shown graphically in Figure 12, with SFE calculated by DFT in (a) and by thermodynamic model in (b). In general, the data in Figure 12 suggests that τ_T exhibits a relatively strong dependence on temperature, but a relatively weak dependence on SFE. For instance, the increase in τ_T of the 28Mn alloy from RT to 100 °C ($\Delta \tau_T$) of ~83 MPa can be deconvoluted into two components by the following approximation:

$$\Delta \tau_T = \Delta \tau_{T,Temperature} + \Delta \tau_{T,SFE} \tag{4}$$

where $\Delta \tau_{T,SFE}$ is the variation in τ_T caused by changes in the SFE from modification of the temperature and $\Delta \tau_{T,Temperature}$ is the change in τ_T caused by all other phenomena, unrelated to changes in the SFE, which are influenced by temperature. $\Delta \tau_{T,Temperature}$ is obtained assuming a hypothetical increase in temperature with constant SFE as shown by purple arrows in Figure 12 until intersection with the line connecting the values of τ_T for the 25 and 28 Mn alloys at 100 °C. Figure 12 graphically shows $\Delta \tau_{T,SFE}$ and $\Delta \tau_{T,Temperature}$. Under these simplified assumptions, and accounting for variation in SFE due to different calculation methods, $\Delta \tau_{T,SFE}$ ranges from 6 to 14 MPa and comprises only a relatively small percentage (6 to 17%) of the total increase in τ_T of the 28Mn alloy with increasing temperature from RT to 100 °C. In Figure 12, the uncertainties of $\tau_{\rm T}$ are relatively large. However, the actual values of τ_T for all conditions are likely to shift in the same direction, positive or negative, due to similar systematic methods for determining τ_T , thus having lesser impact on the relative proportions of $\Delta \tau_{T,SFE}$ and $\Delta \tau_{T,Temperature}$. In addition, the values of τ_T of the 22Mn and 25Mn alloys at 100 °C in Figure 12 are greater than that of the 28Mn alloy at RT, despite the latter having a greater SFE (regardless of SFE calculation method), further highlighting the importance of temperature induced effects on $\tau_{\rm T}$ which appear to be separate from changes to τ_T that result from SFE modification. The SFE values calculated by DFT and by the thermodynamic model in this work clearly differ, as illustrated in Figure 3, but in either case the same general conclusion holds true, namely, that $\tau_{\rm T}$ exhibits a strong temperature dependence but relatively modest dependence on the SFE. Furthermore, as previously noted, the SFE calculated by thermodynamic model, shown in Figure 3, is likely to serve as an upper limit of the actual SFE of the alloys, since the interfacial parameter is assumed constant with increasing temperature in the present work, but likely actually decreases with increasing temperature [23], potentially causing the actual SFE to be lower. As such, the proportion of the increase in τ_T with temperature which is due to an increase in SFE is likely to be less than shown in Figure 12b. These findings provide a greater

understanding of the factors contributing to the temperature dependence of τ_T and insight into potential alloy design strategies for effective control of τ_T at temperatures above RT.

The values of $\tau_{\rm T}$, determined experimentally for the present steels, exhibit much greater sensitivity to temperature relative the calculated values of $\tau_{\rm T}$ reported for the Fe-22Mn-0.6C steel [18], which were calculated at RT, 100 °C, and 200 °C, using equation (2), to be 192, 203, and 227 MPa , respectively. The values of $\tau_{\rm T}$ calculated for the Fe-22Mn-0.6C steel assume a constant value of L₀, the width of the sessile partial dislocations between pinning points forming the twin embryo, and exhibit good agreement with experimental observations of the manifestation of mechanical twinning in the strain-hardening response at RT and above [18]. In the present work, the use of equation (2) with a constant value of L_0 results in a poor approximation of τ_T from RT to 200 °C. Alternatively, rearrangement of Equation (2) enables calculation of individual "fitted" L₀ values for the present alloys that would result in accurate calculation of the experimentally determined τ_T . All other variables in Equation (2) were known or could be estimated with reasonable accuracy, such as G [20], permitting the calculation of L₀, the values of which are in Table 2. The calculated values of L₀ decrease with increasing temperature, from 339 nm in the 28Mn alloy at RT to 119 or 152 nm in the 22Mn alloy at 200 °C, depending on whether the SFE value used in the calculation is from DFT or thermodynamic model. While L₀ was not observed experimentally, a calculated decrease in this parameter is also consistent with microstructural observations of higher dislocation densities at the initiation of mechanical twinning at higher temperatures. For instance, the dislocation density in the 28Mn alloy (qualitatively shown in TEM images in [8]) deformed to 0.1 true strain at 25 °C is relatively low compared to the 22Mn alloy deformed to 0.37 true strain at 200 °C (Figure 9).

While the exact mechanism(s) which result in delays in mechanical twinning in the present alloys with increasing temperature (25 to 200 °C) are not explicitly known, the present work provides some insight into this behavior. Increasing the SFE only modestly increases the stress required for the initiation of mechanical twinning and does not by itself explain the large increase in τ_T with increasing temperature. However, at the initiation of mechanical twinning at elevated temperatures, the present results confirm that higher normal stress levels are present (see Figure 11), the friction stress is significantly lower than at RT (as inferred by the large reduction in yield stress with increasing temperature from RT to 200 °C shown in Figure 10a), and the microstructure contains a significantly higher dislocation density relative to RT.

Reducing the friction stress is reported to increase the favorability of cross slip [44], which in turn can reduce local stress concentrations and the propensity for mechanical twin nucleation [18], potentially delaying the initiation of mechanical twinning to higher normal stress levels, which may be needed to facilitate high enough localized stresses for mechanical twinning. As such, the significant decrease in the friction stress in the present alloys from RT to ~200 °C [8] likely increases dislocation mobility and changes the dynamics of the competition between dislocation glide and mechanical twinning. The decrease in friction stress from RT to 200 °C is in part related the presence of high to Si content (3 wt.%), which causes the transition from thermally activated

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dislocation glide to athermal dislocation to occur at about 200 °C in the present alloys compared to ~RT in FeMnC TWIP steels with no Si addition [45]. Practically, this means the flow stress is highly sensitive to temperature below 200 °C in the present steels. In comparison, TWIP steels that contain elevated C or N levels and no significant Si, such as the Fe-22Mn-0.6C and Fe-14Cr-16Mn-0.3C-0.3N wt.% [50] alloys, have higher friction stresses that exhibit only a modest decrease with increasing temperature from RT to 200 °C [45]. The steels with elevated C and/or N tend to exhibit mechanical twinning at higher temperatures (e.g., 250-300 °C [15,18]) compared to the present steels (~200 °C). Increases in the C content of high Mn steels [51] (and other FCC systems [52]) have also been suggested to reduce cross slip via other mechanisms, such as the large positive impact that solute C may have on the constriction energy of screw dislocations [51,53].

A significantly higher dislocation density is also observed at the initiation of mechanical twinning at elevated temperatures compared to RT in the present alloys, as qualitatively observed by comparing TEM images in Figure 8a and Figure 9. Furthermore, simple theories of work hardening, which approximate the flow stress as the sum of the yield stress and a second term proportional to the square of the dislocation density, which may be valid in the early stages of deformation up to the onset of mechanical twinning, also estimate higher dislocation densities at the initiation of mechanical twinning at higher temperatures [54]:

$$\sigma_T = \sigma_{YS} + \alpha G b \rho^{\frac{1}{2}} \tag{5}$$

where in the present analysis, σ_T is the normal true stress at the initiation of mechanical twinning, σ_{YS} is the yield strength, α is assumed constant [55], b is the total Burger's vector, G is the shear modulus, and ρ is the dislocation density. The range of dislocation density expected at the onset of mechanical twinning is estimated using equation (5) for the 28Mn alloy at RT and the 22Mn alloy at 200 °C, and the results are provided in Table 3. While substantial uncertainty exists in the estimation of the dislocation density from Equation (5), the range of dislocation density for the 22Mn alloy at the initiation of mechanical twinning at 200 °C is estimated to be between 5 and 69 times greater than that of the 28Mn alloy at RT. This large estimated difference in dislocation density may also underestimate the actual difference, since α is assumed constant at 0.26 (reported previously for an Fe-31Mn-3Al-3Si steel [55] at RT), but may actually decrease with increasing temperature [43]. The dislocation density at elevated temperatures is also likely greater at equivalent stress levels at or below the onset of mechanical twinning, in part due to the significantly lower friction stress and yield stress at elevated temperatures relative to RT in these alloys, requiring higher dislocation density to achieve equivalent flow stresses. While it is generally thought that a certain dislocation density is required for the necessary stress concentrations and defect structures to form and initiate mechanical twinning, the present results suggest that excessive dislocation density may impede mechanical twinning or even raise the critical stress required for mechanical twinning under certain circumstances. For instance, the data presented in Table 2 suggest that L₀ decreases with increasing temperature, which may be a consequence of the higher dislocation density prior to and

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at the initiation of mechanical twinning at higher temperatures (indicated in Table 3 and by TEM), and which according to Equation (2), may cause the increase in σ_T with temperature. Other authors have also reported an influence of dislocation density on the critical stress for mechanical twinning, with Kang et al. [54] suggesting that increased dislocation density in fine grained TWIP steels could contribute to higher back stresses compared to relatively coarse grained specimens, which enhances cross slip, and suppresses mechanical twinning. While the present work provides a greater understanding of the factors influencing the temperature dependence of σ_T , additional research is recommended, with a particular focus on experimental observations and theoretical simulations of the temperature dependent defect structures and mechanisms at the micro level which serve to initiate mechanical twinning in Fe-Mn-Al-Si TWIP steels. Table 2 – Normal stress (σ_T) and resolved shear stress (τ_T) determined for the onset of mechanical twinning for the present alloys. Calculated values of τ_T for other TWIP steels from the literature [18,47] are provided for comparison. The intrinsic SFE (γ), shear modulus (G), two terms from equation (2), and the calculated values of L_0 for the present alloys are determined from SFE values calculated by DFT and thermodynamic model.

-		Experiment	ntal Values of	Shear	Parameters and terms calculated using SFE obtained from			Parameters and terms calculated using SFE obtained from				
		Critical -Tw	vinning Stresses	Modulus	DFT			thermodynamic modeling				
Alloy	Temperature (°C)	σ _T (MPa)	τ _T , (MPa)	G (MPa) ^a	γ (mJ m ⁻²)	$\frac{\gamma}{3b_p}$ (MPa)	$\frac{3Gb_p}{L_0} (MPa)$	L ₀ (nm)	γ (mJ m ⁻²)	$\frac{\gamma}{3b_p}$ (MPa)	$\frac{3Gb_p}{L_0} (\text{MPa})$	L ₀ (nm)
Fe-27Mn-2.6Si- 3.3Al [47]	25		201						18.3			200 ^b
Fe-22Mn-0.6C [18]	25		192						28			260 ^c
28Mn	25	387 ±61	182 ±29	72	39 ^d	88	94	339	39 ^d	88	94	339
Fe-22Mn-0.6C [18]	100		203						37			260°
22Mn	100	522 ±61	245 ±29	70.6	19	43	202	155	28	64	181	173
25Mn	100	522 ±61	245 ±29	70.6	24	55	190	165	35	78	167	188
28Mn	100	564 ±61	265 ±29	70.6	42	95	170	184	51	116	149	210
Fe-22Mn-0.6C [18]	200		227						55			260°
22Mn	200	647 ±61	304±29	67.6	23	52	252	119	47	106	198	152

^aG measured previously for the 22, 25, and 28Mn alloys at RT (72 GPa) [7,56]. The temperature dependence is estimated from other high Mn steels containing Al and equates to an ~6% reduction in shear modulus with increasing temperature from 25 to 200 °C [20].

^bObtained by experimental observation in [47]

^cReported in [18]

^dExperimentally determined [7]

Table 3 – Critical stress for the initiation of mechanical twinning (σ_T), yield stress (σ_{YS}), shear modulus (G), magnitude of Burger's vector (b), and constant α used in the estimation of dislocation density, ρ , at the initiation of mechanical twinning from Equation (5) for the 28Mn alloy at RT and the 22Mn alloy at 200 °C.

Alloy	Temperature (°C)	σ _t (MPa)	σ _{ys} (MPa)	G (MPa)	B (nm)	α	ρ (# m ⁻²)
28Mn	25	387 ±61	259	72	0.2560	0.26	2.0x10 ¹⁴ -1.6x10 ¹⁵
22Mn	200	647 ±61	183	67.6	0.2564	0.26	8.0x10 ¹⁵ -1.4x10 ¹⁶



Figure 12 – Influence of intrinsic SFE, as computed by (a) DFT and (b) thermodynamic model, and temperature on the critical resolved shear stress for twinning. The triangle is the 28Mn alloy at RT, the circles are all three alloys at 100 $^{\circ}$ C, and the square is the 22Mn alloy at 200 $^{\circ}$ C.

5 Conclusions

The present study investigated the influence of temperature on the strain-hardening behavior of three Fe– 22/25/28Mn–3A1–3Si wt.% transformation- and twinning-induced plasticity (TRIP/TWIP) steels. The temperature dependent SFE was calculated by two independent methods, DFT and thermodynamic model. Coupling the SFE calculations with quasi-static tensile properties measured from 25 to 400 °C as well as a detailed investigation of the deformation mechanisms with TEM enabled deconvolution of the temperaturedependent effects on SFE and critical stress for mechanical twinning. The following important conclusions were drawn from this work:

- Increasing the temperature from RT to 200 °C resulted in large decreases in yield and ultimate tensile strength in all alloys, attributed in part to a significant reduction in the thermally activated component of the flow stress in this temperature range. Further increases in temperature from 200 to 400 °C result in minimal changes to the strain hardening behavior and flow stress.
- 2. At relatively low strains (0 to 0.1 true strain), the 22Mn alloy, with a SFE of ~15 mJ m⁻² at RT, exhibits planar dislocation glide at RT, with a microstructure consisting primarily of partial dislocations with large

extension and limited evidence of dislocation cross slip. Increasing the Mn content by 6 wt.% (28Mn alloy) results in a transition to from planar to wavy slip and evidence of significantly more cross slip at RT. In addition, increasing the temperature of the 22Mn alloy from RT to 400 °C causes a similar transition in the mechanisms of dislocation glide. In both cases, the transition from planar to wavy dislocation glide is in part influenced by an increase in the SFE and causes a decrease in the work hardening rate.

3. The critical resolved shear stress for the onset of mechanical twinning, estimated from microstructural observations and the strain hardening behavior, increases significantly with increasing temperature until mechanical twinning is suppressed above approximately 200 °C. The resulting increase in SFE with rising temperature contributes only modestly to the increase in critical resolved shear stress for twinning in the present alloys. Other potential mechanism(s) which may cause an increase in the critical resolved shear stress for mechanical twinning with temperature are discussed.

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8 Supplemental Information



Figure S 1 - XRD measurements of the 22Mn, 25Mn, and 28Mn alloys acquired at 25 °C with the first five allowable austenite reflections labeled. The $\{220\}\gamma$ inset is provided to illustrate the effect of Mn content on the shift in diffraction angle and austenite lattice parameter.



Figure S 2 - Values of CTE determined for specific temperature ranges for the 22, 25, and 28Mn alloys.