A simplified model connecting lattice distortion with friction stress of Nb-based equiatomic high-entropy alloys

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ABSTRACT

We conducted systematically tensile tests with a series of Nb-based equiatomic alloys with single-phase, body-centered-cubic (bcc) structure. By subtracting all possible strengthening contributions, the intrinsic strength (or lattice friction stress) of each alloy was extracted. It was found that lattice friction stress scaled linearly with the lattice distortion in these Nb-based alloys. A simple model was developed to interpret this result. Strengthening effects of lattice distortion and dislocation core width between bcc Nb-based and face-centered-cubic (fcc) Ni-based equiatomic alloys were also compared. It was demonstrated that the enhanced strength in high-entropy alloys was attributable to the lattice distortion.

IMPACT STATEMENT

Lattice friction stress in equiatomic alloys correlates linearly with the lattice distortion, quantitatively defined as the average lattice strain. A simple physical model was developed to interpret this result.

Over the past decade, multi-principal elements (MPE) alloys, or high-entropy alloys (HEAs) \cite{1,2}, have increasingly attracted research attention due to their unusual mechanical properties, such as high strength/hardness \cite{3}, good wear resistance \cite{4}, and excellent thermal stability \cite{5}. Lattice distortion has been widely recognized as one of the main contributing factors for the observed unusual mechanical properties \cite{6}. Various characterization methods, including neutron diffraction \cite{7,8}, synchrotron X-ray diffraction \cite{9}, and high-resolution transmission electron microscopy \cite{10}, have been employed to study lattice distortion in these HEAs. For instance, Tong et al. \cite{9} evaluated local lattice distortion in some fcc HEAs (i.e. NiCoCr, NiCoFeCr and NiCoCrFeMn) using pair distribution function analysis from synchrotron X-ray data. Subsequently, several statistical models, based on atomic size difference, were also proposed to describe lattice distortion, such as the \( \delta \)-parameter \cite{11} and \( \alpha_2 \)-parameter \cite{12}. Recently, Okamoto et al. \cite{13} used the first-principles to calculate the average root-mean-square atomic displacements (MSAD)\(^{1/2} \) of the constituent atoms in the Cantor alloy and found an empirical correlation between (MSAD)\(^{1/2} \) and the yield strength of HEAs. These models, together with the advanced characterization techniques, provided useful assessment and direct visualizations of lattice distortion, but a physical-based quantitative connection between the atomic-scale lattice distortion and macroscopic materials properties, in particular, the mechanical properties, is still missing.

Recently, in a study of fcc Ni-based equiatomic alloys, we reported that lattice distortion, defined as the lattice parameter variation caused by elemental substitution, was linearly dependent on the lattice friction stress \cite{14}. However, a physical picture for this result was not given. Here, we model the correlation as follows.

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Figure 1. (a) Lattice distortion can be achieved either via chemical substitution or mechanical dilation/contraction. (b) Lattice constants of the initial and final (under a mean hydrostatic stress $\sigma_m$) unit cells are $a_0$ and $a$, respectively.

Imagine a bcc cell, for example, Nb, with a lattice constant of $a_0$. To build a multi-component bcc-HEA from this cell, we begin to substitute Nb atoms one-by-one with other transition elements (such as Ti, Zr, and Hf). Since the crystal structure is fixed and each newly added atom has a size and charge different from Nb, the cell will gradually distort as a result of the substitution. After finishing the substitution, the lattice constant of the cell becomes $a$, as schematically illustrated in Figure 1(a). Alternatively, we can imagine the cell is mechanically strained (dilating or contracting) isotropically from the initial lattice constant $a_0$ to the final $a$. The two routes, one is chemical substitution and another mechanical straining, produce the same dilated or contracted cell. Let us assume the stored energy in the final cell is purely mechanical and all other energy forms, such as magnetic and electronic, are insignificant. In other words, from the energy viewpoint, the final cell produced via either processing route is essentially the same.

In the case of via mechanical route, we consider the elastic deformation of a cubic cell (see Figure 1(b)). The linear ($\delta$) and volumetric ($\Delta$) strains are $\delta = (a - a_0)/a_0$ and $\Delta = (\Omega - \Omega_0)/\Omega_0 \approx 3\delta$, respectively, where $a$ and $a_0$ are the lattice constants of the HEA and baseline Nb, and $\Omega$ and $\Omega_0$ are cell volumes of the HEA and baseline Nb, respectively. The mean hydrostatic stress, $\sigma_m$, required to cause such deformation is

$$\sigma_m = B \cdot \Delta \approx B \cdot 3\delta \quad (1)$$

where $B$ is the bulk modulus of the alloy. In other words, an internal stress is stored in the final cell after elemental substitution and this stress is linearly proportional to the lattice distortion $\delta$. In fact, we have previously reported this correlation in the study of fcc Ni-based equiatomic alloys [14]. It is particularly noted that the currently defined lattice distortion is essentially the average lattice strain, which is similar to the average (MSAD)$^{1/2}$ defined by Okamoto et al. [13].

In the current study, we only consider the onset of yielding and there are several pieces of evidences indicating yielding in HEAs is asymmetrical. For example, yield strength of the Cantor alloy (NiCoCrFeMn) in tension and compression are 316 and 445 MPa, respectively [15], and our recent stress–strain measurements from a bcc-HEA (NbTiZrHf) also confirmed yielding asymmetry (unpublished private data, an extension of Ref. [16]). These results led us to believe that yielding of HEAs follows a Mohr-Coulomb criterion, not von Mises. In other words, yielding of HEAs is pressure-dependent. Consequently, although hydrostatic stress created by lattice dilation/contraction cannot cause dislocations to move, it can affect the effective shear stress, thus the onset of dislocation motion.

To validate the above model, we carry out systematic experiments to measure the lattice friction stresses in pure Nb and a series of bcc Nb-based equiatomic alloys (from binary to quinary) and correlate them with their respective lattice distortion. For comparison, the relationship between lattice friction stress and lattice distortion in fcc Ni-based equiatomic alloys is also discussed. We will show that there exists evidently a quantitative correlation between friction stress and lattice distortion. This result offers an explanation for the unusually enhanced strength in HEAs.

Ingots of pure Nb and Nb-containing equiatomic alloys listed in Table 1 were prepared by arc-melting mixtures of high-purity (> 99 wt.% constituents elements in a Ti-gettered argon atmosphere. To promote chemical homogeneity, these ingots were re-melted at least eight times and, then, injected into a copper mold ($10 \times 10 \times 60$ mm$^3$) with water cooling. Phases microstructure of the cast alloys were examined using X-ray diffraction (Cu K$_\alpha$ radiation, MXP21VAHF) and scanning electron microscopy (ZEISS Supra 55) and results indicated that all samples are bcc single-phase with an average grain size of about 100 μm. Tensile tests were performed on a CMT4105 universal tensile testing.
Table 1. Summary of lattice constant (a), lattice distortion (δ), Young’s modulus (E), 0.2% offset yield strength (σy), the lattice friction stress (σf), corresponding σf/E, and the normalized dislocation core width (w/b) of bcc-structured, Nb-based equiatomic alloys.

| Alloys          | Processing | a (Å) | δ     | E2 (GPa) | σy (MPa) | σf (MPa) | σf/E (10^-3) | w/b | Ref. |
|-----------------|------------|-------|-------|----------|----------|----------|---------------|-----|------|
| Nb              | AC         | 3.307 | 0     | 103      | 188 ± 7  | 130      | 1.3           | 1.25| Present work |
| NbTi            | AC         | 3.292 | −0.0047 | 92      | 354 ± 14 | 309      | 3.4           | 1.09| Present work |
| NbTiZr          | AC         | 3.404 | 0.0292 | 81      | 749 ± 11 | 706      | 8.7           | 0.94| Present work |
| NbTiHf          | AC         | 3.389 | 0.0246 | 94      | 613 ± 9  | 567      | 6.0           | 0.99| Present work |
| NbZrHf          | AC         | 3.490 | 0.0551 | –       | –       | –       | –             | –   | –    |
| NbTiZrHf        | AC         | 3.451 | 0.0433 | 83      | 783 ± 17 | 739      | 8.9           | 0.94| Present work |
| NbTiZrHf        | AC         | 3.443 | 0.0410 | 83      | 879      | 830      | 10.0          | 0.92| [17] |
| NbTaTiZr        | AC         | 3.359 | 0.0155 | 112     | 876 ± 27 | 826      | 7.4           | 0.97| Present work |
| NbTaTiHf        | AC         | 3.368 | 0.0185 | 110     | 762 ± 10 | 712      | 6.5           | 0.99| Present work |
| NbTaZrHf        | AC         | 3.446 | 0.0421 | 103     | 1046 ± 15 | 997     | 9.7           | 0.93| Present work |
| NbTaTiZrHf      | AC         | 3.421 | 0.0344 | 114     | 1142 ± 23 | 1091    | 9.6           | 0.93| Present work |
| NbTiZrHf        | CR + A     | 3.419 | 0.0338 | 114     | 830      | 779      | 6.9           | 0.98| [18] |
| NbTiZrHf        | CR + A     | 3.404 | 0.0292 | 114     | 1145     | 1067     | 9.4           | 0.93| [19] |
| NbTiZrHf        | CR + A     | 3.402 | 0.0286 | 114     | 1114     | 1048     | 9.2           | 0.93| [20] |
| NbTaZrHf        | CR + A     | 3.406 | 0.0298 | 114     | 940      | 892      | 7.9           | 0.96| [21] |

aAC: As-Cast; A: Annealed; CR: Cold Rolled.
bMeasured from elastic portion of tensile stress–strain curves.

Figure 2. Lattice distortion (δ) in bcc Nb-based (also listed in Table 1) and fcc Ni-based equiatomic alloys.

Before analyzing the data in Table 1, it is first pointed out that, in the current bcc equiatomic Nb–Ta–Ti–Zr–Hf alloy system, only Nb and Ta are bcc-metals with a nearly identical lattice constant (Nb: 0.3294 nm, Ta: 0.3303 nm [23]), while all the other constituent elements are hcp, instead. Furthermore, Nb and Ta both belong to group-VB elements in the Periodic Table, their chemical specificities (e.g. charge transfer and electronegativity) are also similar. It is, therefore, reasonable to treat these alloys as a series of Nb-based equiatomic alloys. In other words, Nb can be considered as the baseline of the equiatomic Nb–Ta–Ti–Zr–Hf alloy series and lattice distortion can subsequently be computed; δ values for these Nb-based bcc-equiatomic alloys are now included in Table 1.

A graphic comparison of the lattice distortion in the current Nb-based and the previous Ni-based equiatomic alloys [24] is shown in Figure 2. Except for binary NbTi which has a contracting lattice strain, all other bcc Nb-based and fcc Ni-based alloys show a dilating lattice strain. For the convenience of illustration, the x-axis is plotted with increasing number of constituent elements. It is evident in Figure 2 that lattice distortion does not simply increases with increasing number of constituent elements in either bcc Nb-based or fcc Ni-based HEAs. In actuality, ternary alloy NbZrHf has the largest lattice distortion in the Nb-based alloys. It is also apparent that lattice distortion in the bcc Nb-based alloys is far larger than that in the fcc Ni-based alloy, especially in the ternary and quinary systems. This result is consistent with the ab initio calculations performed by Song et al. [25], who concluded that lattice distortion in bcc refractory HEAs (e.g. NbZrHf, NbTiZrHf, and NbTaTiZrHf) was significantly
higher than that in fcc HEAs (e.g. NiCoFe, NiCoFeCr, and NiCoCrFeMn).

Lattice distortion in HEAs can result in a strengthening effect. To evaluate this effect, the nature of yield strengths of the current Nb-based equiatomic alloys in Table 1 must first be clarified. Yield strength of an alloy \( \sigma_y \) generally consists of two parts (intrinsic and extrinsic),

\[
\sigma_y = \sigma_f + \Delta\sigma_s \tag{2}
\]
or

\[
\sigma_f = \sigma_y - \Delta\sigma_s \tag{3}
\]

where \( \sigma_f \) is the friction stress (or intrinsic strength) and \( \Delta\sigma_s \) refers to all possible strengthening contributions, including the solid-solution strengthening, dislocation strengthening, grain-boundary strengthening, and precipitation strengthening. However, it is noted that these strengthening mechanisms are all assumed absent in the model (see Figure 1) and the internal stress described in the model is actually the friction stress. In contrast, the experimentally measured yield strengths listed in Table 1 possibly contain contributions from the above mechanisms. To deduce the friction stress, it is necessary to subtract their contributions. For brevity and clarity, a summary of the calculated strengthening contributions is presented in Table 2.

A particular note about the precipitation strengthening in Table 2 is that, in the current Nb-based equiatomic alloys, precipitates are absent. As mentioned before, no second phase was discernible in these bcc-alloys, based on scanning electron microscope and X-ray diffraction analyses. Lei et al. [16] also confirmed that the as-cast equiatomic NbTiZrHf alloy did not contain any precipitate using neutron diffraction and scanning transmission electron microscopy. Consequently, precipitation strengthening is excluded here.

Another note is about the solid-solution strengthening. As pointed out before [14], contribution from solid-solution strengthening in equiatomic alloys can be, in principle, ignored. According to the traditional solid-solution strengthening theories, when moving dislocations travel through a solvent lattice, they will encounter resistance from local lattice distortion caused by the mismatch between solvent and solute atoms. However, in equiatomic alloys, lattice distortion is quite uniform and global, as long as the constituent atoms are randomly partitioned in the lattice, and they indeed are [32,33]. In this case, dislocations are envisioned to travel through a heavily distorted lattice, which yields a highly distorted Peierls potential landscape. Since the solute content is negligible in these alloys, contribution from solid-solution strengthening can also be excluded.

Using available data for Nb-based equiatomic alloys in Table 2, lattice friction stresses can now be readily deduced from Equation (3) and their values are presented in Table 1. For easy discussion, we express friction stresses in a normalized form \( \sigma_f/E \) and present these values also in Table 1. It is apparent that the normalized friction stresses of bcc Nb-based equiatomic alloys are generally much higher than those of fcc Ni-based equiatomic alloys. In perspective, the normalized friction stresses \( \sigma_f/E \) of some bcc metals, fcc Ni-based and the current bcc Nb-based multi-component \(( \geq 3)\) equiatomic alloys are listed together in Table 3. It is readily seen that \( \sigma_f/E \) of fcc Ni-based alloys are actually close to those of the traditional bcc metals, but \( \sigma_f/E \) of bcc Nb-based alloys are about one order of magnitude higher. This result is consistent with the notion that dislocation motion in bcc Nb-based equiatomic alloys is more difficult than that in the conventional bcc alloys.

It is noted again that, in the current model (see Figure 1), the internal stress is actually the stress increment with respect to the intrinsic stress of the initial `undeformed' cell, which is the pure bcc-Nb. Therefore, to validate the model, we must further subtract the measured lattice friction stresses from the friction stress of Nb, that is, \( \Delta\sigma_f = \sigma_{f,HEA} - \sigma_{f,Nb} \), where \( \Delta\sigma_f \) is

### Table 2. Strengthening effects produced by various mechanisms in the current bcc Nb-based equiatomic alloys.

| Strengthening mechanisms | Solid-solution strengthening | Dislocation strengthening\(^a\) | Grain-boundary strengthening\(^b\) | Precipitation strengthening |
|--------------------------|------------------------------|----------------------------------|-----------------------------------|---------------------------|
| Equation                 | NA                           | \(\Delta\sigma_s = M\alpha Gb\gamma^{1/2}\) [26] | \(\Delta\sigma_{gb} = k_{gb}d^{-1/2}\) [27] | NA                        |
| Calculated value (MPa)   | NA                           | \(\Delta\sigma_s \approx 19–27\)  | \(\Delta\sigma_{gb} \approx 24\)  | NA                        |

\(^a\)M: Taylor factor, \(M = 2.75\) [28]; \(\alpha\): an empirical constant, \(\alpha = 0.4\) [28]; \(G\): shear modulus, \(G = 0.367E\) [29]; \(b\): Burgers vector, \(b = (\sqrt{3}/2)\alpha\); \(\rho\): dislocation density \(\rho \approx 4 \times 10^{15} \text{m}^{-2}\) from fully annealed Nb [30] was adopted.

\(^b\)\(k_{gb} = 240 \text{MPa}\mu\text{m}^{1/2}\) from fully annealed NbTiZrHf [21] was adopted for calculations, since \(k_{gb}\) is not available for the majority of the concentrated alloys. For pure Nb, \(k_{gb} = 340 \text{MPa}\mu\text{m}^{1/2}\) [31]; \(d\): average grain size.

NA: not applied.

### Table 3. Normalized friction stresses \(\sigma_f/E\) of the traditional bcc metals, bcc Nb-based, and fcc Ni-based equiatomic alloys (number of components \(\geq 3\)).

| Material | Ni-based [14] | Nb | Ta [34] | Mo [34] | W [34] | Nb-based |
|----------|---------------|----|---------|---------|--------|----------|
| \(\sigma_f/E(10^{-5})\) | 0.5–1.0 | 1.3 | 0.6     | 0.4     | 0.7    | 0.6–10.0 |

\(M_{\text{HEA}}\): Taylor factor, \(M_{\text{HEA}} = 2.75\); \(\alpha\): an empirical constant, \(\alpha = 0.4\); \(G\): shear modulus, \(G = 0.367E\); \(b\): Burgers vector, \(b = (\sqrt{3}/2)\alpha\); \(\rho\): dislocation density \(\rho \approx 4 \times 10^{15} \text{m}^{-2}\); \(d\): average grain size.
the internal stress, and \( \sigma_f, \text{HEA} \) and \( \sigma_f, \text{Nb} \) are the friction stresses of HEA andNb, respectively.

According to Equation (1), this internal stress is dependent linearly upon the bulk modulus. Since bulk modulus \( (B) \) and Young’s modulus \( (E) \) are also linearly proportional [35], the internal stress is finally scaled with lattice distortion as \( \Delta \sigma_f \propto \sigma_0 \propto E \cdot \Delta \delta, \) or \( \Delta \sigma_f / E = c \delta, \) where \( c \) is the scaling constant. The functional relationship between the normalized internal stress \( (\Delta \sigma_f / E) \) and lattice distortion \( (\delta) \) for the Nb-based alloys is plotted in Figure 3, in which the error bars were calculated from estimated uncertainties of dislocation densities \( (\rho) \) and the Hall-Petch coefficients \( (k_{HP}) \) in these alloys. It is particularly noted that, in the figure, we use a positive strain for NbTi since it is perceived that the interaction between dislocation and distorted lattice is similar to the solute-dislocation interaction based on solid-solution theories [39,40]. According to these theories, when lattice strain is dominant, dilation and contraction can cause identical strengthening effect. It is evident in Figure 3 that, despite some data scatter, a linear relationship between the two variables is apparent, satisfactorily validating our model. As mentioned before, the linear correlation has also been observed in the fcc Ni-based equiatomic alloys [23,24,36–38]. For direct comparison, data from the fcc Ni-based equiatomic alloys are also included in Figure 3. Readily noted in the figure is the fact that bcc Nb-based alloys are more sensitive to lattice distortion \( (c = 0.219) \) than fcc-Ni-based equiatomic alloys \( (c = 0.035) \). Furthermore, the larger data variation in bcc Nb-based alloys may be caused by the inconsideration of magnetic and electric energies, and a non-Schmid effect in the model. For the non-Schmid effect and in the case of fcc-based HEAs, there are several reports indicating that single crystalline HEAs follow Schmid’s Law [41,42]. To our best knowledge, similar result has not yet been reported in single-crystal bcc HEAs. However, since the yielding of bcc metals often does not obey Schmid’s Law [43–45], it is conceivable that dislocation slip in bcc-HEAs may also occur in a non-Schmid fashion. At the present moment, the evaluation of these non-Schmid factors in equiatomic alloy systems is challenging and obviously beyond the scope of our simplified model. Nevertheless, to improve the model’s predictability it will be considered and included in the future.

Peierls stress is usually regarded as the lattice friction stress and it is given as [46]

\[
\tau_f = \frac{2G}{1 - \nu} \exp \left[ -\frac{2\pi w}{b} \right]
\]

where \( \tau_f \) is the friction shear stress, \( G \) is the shear modulus, \( \nu \) is the Poisson’s ratio (Note: \( \nu \) is not available for the majority of the Nb-based alloys, thus we borrow it from the Poisson’s ratio of Nb), \( w \) is the width of dislocation core, and \( b \) is the Burgers vector. The dislocation core width normalized by the Burgers vector \( (w/b) \) is, then,

\[
\frac{w}{b} = \frac{1}{2\pi} \ln \left[ \frac{2MG}{\sigma_f (1 - \nu)} \right]
\]

where \( \sigma_f = M \tau_f \), and \( M \) is the Taylor factor. Values of \( w/b \) in Nb-based equiatomic alloys are calculated and listed in Table 1. A graph comparing the normalized dislocation core width of the bcc Nb-based and fcc Ni-based alloys is shown in Figure 4. It is apparent in the figure that pure Nb has a higher \( w/b \) value \( (w/b = 1.25) \) than the Nb-based equiatomic alloys \( (w/b < 1.10) \), indicating that dislocation glide is more difficult in the equiatomic alloys. Also noted is that \( w/b \) values in these Nb-based equiatomic alloys (except for the pure Nb and binary NbTi) fall within the range of 0.92–0.99 and are almost constant \( (w/b = 0.95 \pm 0.03) \), similar to that observed in the Ni-based equiatomic alloys, suggesting that entropy effect becomes appreciable and effective only when the number of principal elements is higher than three, that is, ternary systems (so-called medium entropy alloys). It is consistent with the notion that NiCoCr [47] and NiCoV [48] medium entropy alloys often exhibit some unusual mechanical properties, such as ultra-strength \( (> 1 \text{ GPa}) \) with decent ductility \( (> 20\%) \). Furthermore, the \( w/b \) values for bcc Nb-based alloys are noted to be much lower than those for fcc Ni-based equiatomic alloys \( (w/b = 1.33 \pm 0.04) \), which is consistent with the notion that lattice friction stress of bcc Nb-based alloys is much higher than that of fcc Ni-based equiatomic alloys.
Figure 4. The normalized dislocation core width \((w/b)\) in bcc Nb-based and fcc Ni-based equiatomic alloys.

In summary, we have quantitatively evaluated the lattice distortion in a series of single-phase, bcc-structured, Nb-based equiatomic alloys. The magnitude of lattice distortion in these bcc alloys does not necessarily increase with increasing number of constituent components and is larger than that in fcc Ni-based equiatomic alloys. After subtracting the strength contributions from all possible mechanisms, the lattice friction stress of each of the current Nb-based alloys is extracted and we find that it scales linearly with the lattice distortion in these alloys. In addition, lattice distortion in bcc Nb-based alloys can apparently produce a much higher hardening effect than that in fcc Ni-based equiatomic alloys. The much higher friction stress of bcc-HEAs compared to that of fcc-HEAs may be attributed to a smaller dislocation core. The current results suggest that remarkable strength enhancement in HEAs is probably resulted from a high friction stress, which is, in turn, caused by lattice distortion.

Disclosure statement
No potential conflict of interest was reported by the authors.

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