

Discovering short-range ordering in a medium entropy alloy via intelligent big data

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Abstract

Short-range orders (SROs) become more interesting in an intriguing new class of metallic materials – High-Entropy Alloys (HEAs) or Medium-Entropy Alloys (MEAs), that contain multi-principal elements. However, from the atomic level and the compositional viewpoint, observing the SRO in HEAs or MEAs is still very challenging, because it is difficult to distinguish the elements with very closed atomic numbers using electron microscopies. Here we directly observed the SRO in a medium-entropy alloy through deciphering the big data by atomic probe tomography (APT) via a new machine learning method. Using our matching learning method, the nearest neighbors of the detected elements, preferred bonds of each atom, in the APT data can be achieved. Our method offers a new pathway to observe SRO directly not only in those compositionally-complex alloys but also in the traditional dilute alloy systems, which can accelerate the design of advanced materials with favorable SROs for achieving desired properties.

Introduction

Contrary to conventional alloying, High Entropy Alloys (HEAs) do not have a principal component, but they are instead based on near equiatomic mixtures of five or more elements (1–4). The peculiar high strength and toughness inherent in HEA at certain temperature (5, 6) has remained a mystery for material scientists. Conventional perception has that HEAs in their solid solution state are not very strong. Scientists reckoned that one ordered preference between elements in a rare small range, may affect the subtle action of a Marco world. Such phenomena have been validated through various theoretical models, however, until today there is no direct and clear real-time observation on the atomic level. We are endeavor to observe the preference of each element on the atomic level by advancing analytical methodology of APT at the atomic scale via up-to-date machine learning techniques.

Specifically, here we report: short-range preference or nano-range ordering was discovered in typical high- and medium- entropy alloys (CrCoNi) via semantic machine learning technology based on results from APT. The machine learning algorithm namely ‘alien-philic index (API)’ we developed in this study can help to clearly and easily identify the capacity of SRO (7, 8) in multi-component alloys. Later on, we find that high and medium entropy alloys with various capacity of SRO behave ultra-mechanical properties such as yield stress, toughness and hardness. To seek the subtle relationship, furthermore, when we process such alloy in varied high temperature heat treatments (typically like 800°C, 600°C and 1100°C), altered capacity of SRO with different ‘alien-philic index’ are discovered. It seems that 800°C tends to be an ‘SRO favorable temperature’ with a large SRO capacity of 65.8% API. These results provide insight into the role of nano- or short-range ordering and associated ordering strengthening mechanisms in design of metallic materials, so as to other energy related functional materials. Pioneers of high entropy alloys eager to find out the preference of elements for them to make better design of strong and light materials for every application(5, 9, 10). Clearly, there are preference(11, 12). Yet it is hard to directly detect the short-range ordering in HEAs. Thus, they tried hard by using simulation and theoretical methods (11– 13). However, with the development of artificial intelligence and big data method, we find that by, Maximum Likelihood

Estimation (MLE), we can clearly and directly observe every ‘favorable’ atom of each individual element, even at a real-time level.

Result And Discussion

Fig. 1 shows the three-dimensional (3D) Atom Probe tomography dataset of a typical high entropy alloy - CrCoNi. Graph A is 3D atom maps for the distributions of all those three atoms Cr, Co and Ni in three dimensions while B and C are the distributions in the 1 nm-thick x-z slice and x-y slice respectively, in order to present a clearer view of the arrangement. The real time observation shows in D, E and F which illustrate actual position with the nearest four atoms of each atom in the area for Cr, Ni and Co. From the measurement of APT, it seems that three elements are randomly distributed. This is a whole statistic result (composition of each element is presented in the Supplementary Materials) . However, by analyzing each atom individually on its nearest neighbors using intelligent big data analyses (shown in D, E and F), we find certain preference for each atom. From the zoomed in atom-bond visualization *Four-Neighbor Atomic Graph*, as presented in D, E, F, it seems that Cr favor Co and Ni rather than itself. Bonding band of Cr-Co and Cr-Ni are much frequent than Cr-Cr, Co-Co and Ni-Ni. *This observation motivates our algorithms in later discussions.* To *quantify* the preference, or capacity of SRO, we introduce an index called ‘alien-philic index (API)[1]’.

Our concept of ‘alien-philic index’ and real SRO capacity are closely related. Figure 2 to a large extent, supports our aforementioned hypothesis. Figure 2 is the deep Machine learning on APT data analysis in which A, B and C are the “alien neighbor” percentages (ANPs), as the alien-philic index, contrasted with the ANP of an element with no affinity to alien atoms (50%) in the first nearest neighbor shell for Cr, Ni and Co, respectively. The ANPs have the same profile within the same shell, yet have a stepwise pattern across shells, with ANP being highest in the first shell, demonstrating an affinity for alien atoms². Figure 2 D, E and F show the ANP of the four nearest neighbor shells of Cr, Ni, Co, respectively. The three figures are all around 16% higher than their randomly-distributed counterparts. Distribution of neighbor preference in first, second and 3rd shell.

To rationalize these observations, we used Maximum Likelihood Estimation. Thus, we designed a computer algorithm to gain a better understanding of the relationship among the atoms, especially the comparison of homogeneous neighbors and alien neighbors. This algorithm, for each atom, finds its k closest neighbors, and categorizes these neighbors based on their element. Then the algorithm counted the number of neighbors of the same element (homogeneous neighbors) and the number of neighbors of different element (alien neighbors), and compares these two counts by computing the ratio of alien-neighbor count over the total neighbor count. We call this ratio an “alien neighbor” percentage (ANP) for this atom. This algorithm would perform such procedure on all atoms within the designated boundary, and the average of all ANPs on atoms of the same type is treated as the ANP of this element type. This algorithm repeats the whole process for as little as 1 closest neighbor, and as far as 42 neighbors. Then for each element type, we can get 42 different ANPs to compare with. The ANPs are good indicators of “alien neighbor” preference, and thus good indicators of short-range ordering, because we know, if there is

no short-range ordering, we will not be able to observe differences between short-range and long-range atom relations, and thus the ANP at 1 closest neighbor would be statistically indistinguishable from the ANP at 42 closest neighbors. However, using this algorithm, after we calculated the ANPs for all atoms, we can observe a decreasing trend on ANPs for all three types of elements when we compare the computed trend with the assumed trend under a random distribution. The differences of these two trends give us an idea of the actual existence of a greater preference in heterogeneous atoms when we only count several of the closest neighbors. We further categorize the preferences based on atom shell, where we consider the first 12 neighbor atoms as the first shell, the next 6 neighbor atoms as the second shell, and the further 24 neighbor atoms as the third shell, and others as the fourth shell. From the image above, we can see that the preference is much stronger in the first shell, then becomes more gradual in the second and third shell, and finally dissolves after the fourth shell. From the observations above, we can conclude a decreasing trend on “alien neighbor” preference, and thus a clear short-range ordering, while this ordering vanishes in long range. From the comparison of Figure 2 A, B, and C, we can further conclude that this SRO does not only apply to certain element type, but it applies to all of three element types under analysis of this paper. To further strength our conclusion, we performed the same analysis on several different samples, where some are believed to have higher SROs, and some are believed to have lower SROs. From image A we can see that, although there are differences in actual values of ANPs, the overall decreasing trend is always consistent. After using this algorithm on dozens of datasets, we can conclude that the short-range ordering is so statistically significant that it cannot be accidental to some test set only, but it must have a universal existence on all the materials of such kind. The designed methodology can not only be applied to solid materials but for a border range of functional materials as well. Concrete methodology is presented afterward. In short, Cr-Co, Cr-Ni are strong short-range couple (~23%) based on big-data investigation of all the individual atom based on APT, experimentally. We call it ‘Mutual preference’ element (refer to Fig. 2). Bonding of Cr-Cr, Co-Co and Ni-Ni are relatively weak couples with ~11% chemical bonds. We find certain atoms, Cr-Ni, Cr-Co are mutually attracted. It is hard to break the bonding when you pull them apart(11, 12). That is why CrCoNi is strong and tough. From chemical perspective, Co and Ni are belonging to electron rich metal family. Compared to these two metals, i.e. Co, Ni, Cr seems much more electron deficient. Consequently, Cr prefers to form the intermetallic bonding with Ni or Co to achieve a more stable structure(14, 15). Please refer to extended Figure S4 in Supplementary Materials.

From the tensile curves presented in Fig. 3A, it is found that the annealing treatment does not change the slope of elastic region, which indicates that the present SRO adjustment change little on the original structure of this alloy. With the strain increased to yielding, the 800 °C sample exhibits the highest yield stress and hardness. In the meanwhile, the uniform elongation of 600 °C sample, with an SRO-high state, is compared to the 800 °C sample with a strong SRO-mid state. It means that the yield and tensile strength of HEA can be improved by the increasing the SRO intensity. With annealing temperature increasing to 1100 °C, the 1100 °C sample showing the lowest SRO exhibits the lowest yield stress and hardness, but the highest uniform elongation. The present tensile tests reveal that the intensity of the

SRO state has a strong influence on the yield strength of HEA, that is, higher SRO can result in larger yield strength.

Thus, tuning the capacity of SRO can selectively enhance a desired property. Visualizing from Fig. 2 and 3, there is a clear trend that the HEA with a higher SRO capacity gains superior yield and ultimate stress. Thus, here we desperately to seeking for the reason: varies distribution of alloying elements on atomic scale, and the impact on mechanical properties on micro and macro scales. Understanding of the underlying mechanism is crucial, especially deep from atom scale and link the properties with the local atomic configurations at nano landscape.

One would naturally question if there is a saturation of the dominant SRO type, at what level other types persist (16). From the big data analysis by intelligent machine learning, we find that Cr-Co and Cr-Ni can be regard as the dominant SRO type considering the large capacity (~70% refer to Fig. 2) in kind. Second will be on the extended order, and the organization of SRO motifs, on the length scale of a couple of nanometres (medium- range order, MRO) and beyond. All of such questions and concern of configuration of short chemical ordering can be directedly and clearly observed through output (Fig. 1 D, E, F and movies in the SI) from the big data algorithm and analysis (Fig. 2). That is a big movement in real-time observation of APT big data motif.

Deep and critical experiments of structures should be carried out to review the aforementioned qualitative analysis. To reveal the underlying mechanism of how the local atomic configurations (i.e. SRO) affect the whole mechanical performance, we conducted microstructure analysis on every sample involved in this study. It has been well established that the improvement of degree of SRO can lead to a transition of glide mode from wavy slip to planar slip (17– 19). The reason can be attributed to the local interactions between dislocations and SRO. When the first dislocation faces a higher SRO, it needs to overcome a higher resistance to move through the lattice, meanwhile, destroying the SRO that cannot be restored later. Once this dislocation source is activated, other dislocations can follow the first one push against it thus making it easier to overcome the high resistance, leading to the deformation localized to a single slip plane, i.e., planar slip. Due to the high SRO, dislocations have more chances to dissociate and react, which can bring out unique deformation features. For example, in the 800 °C state with the highest SRO (Fig. 4E), Lomer Cottrell (LC) locks are formed through the reactions of two partials $(a/6)\langle 112 \rangle$ from two dissociated dislocations. Due to the immobile feature, LC locks act as both dislocation barriers and multiplication sources, which are responsible for the significant work hardening and high tensile strength. However, stress can also concentrate near the LC lock, resulting in the initiation of cracks and bringing the decrease of ductility as observed in the 800 °C state (Fig. 3). In contrast to the 800 °C state, only some LC locks are found in the 600 °C state with the higher SRO, and few LC locks are seen in 1100 °C state with lowest SRO. In addition to the effect of SRO on slip mode transition, the correlation between SRO and SFE (stacking fault energy) has been put forward. Generally, high SRO can lead to low SFE. Recent work

reported by Ding et al (12). has revealed the capability of SRO in tuning SFE in CrCoNi alloy through first-principles calculations. Here we find that high SRO (600 °C and 800 °C states) shows the prevalent SFs, but not in the low SRO of 1100 °C state. Moreover, nanotwin-HCP lamellae are observed as well (Fig. 4D and 4E), similar to the reported work (20, 21). These observed features in the high SRO should be responsible for the high yield strength.

We have also found that Cr's neighbors tend to be Ni ($R=1.15 \text{ \AA}$) and Co ($R=1.16 \text{ \AA}$), and their radius are slightly smaller than Cr ($R=1.18 \text{ \AA}$). For the SRO-high sample, the ANPs of the 1st shell and 2nd shell are both greater than 50%. To match the scale, we simply defined Cr-SRO as a spherical shrinkage zone with a diameter of 0.8 nm, which is consistent with the findings of Ritchie et al (22). Correspondingly, there are equal expansion zones in the entire system. We discussed the SRO strengthening mechanisms revealed by MD simulations. Recently, Wang et al. have proposed a scaling meta-atom molecular dynamic (MAMD) method. They introduce a scale factor in a meta-atoms system with a single interatomic potential, which can introduce abundant tunable local lattice distortions, thereby preserving the effect of lattice mismatch. This method has successfully revealed how lattice distortion at atomic scale impacts dislocation behavior in HEAs (23, 24).

The fully relaxed configurations for $s = 0.0, 0.1, 0.2$ and 0.3 represent SRO-none, SRO-low, SRO-mid and SRO-high, respectively. The dislocation motion is strongly affected by the SRO. The dislocation trajectories under the constant shear stress range from 400 to 1000 MPa for different scaling factors. For $s = 0.0$, the dislocations move at constant speeds after a small period of acceleration. However, the dislocation motion is characterized by local pinning and depinning processes for $s = 0.2$ and $s = 0.3$, with completely different trends in dislocation position under different stresses, as is shown in Fig. 5.

Therefore, a significant difference of dislocation behavior has been generated although the atomic positions only deviate about $0.01 \text{ \AA}$ in average from the perfect FCC lattice sites, which may contribute to the distinct mechanical behavior of HEAs. The foregoing discussions confirm that SRO is indeed a strong barrier to the transmission of dislocations.

[1] 'alien-philic atoms' are atoms which, in their k-nearest atoms vicinity, have more than $[2k/3]$ neighbor atoms of a different type; Here we name this proportion as alien-philic score

Conclusion

Great materials are designed to save energies. In our pursuit of concrete structure-property relations thus far, we find that not only for strong and tough structural material, by knowing elements' configuration (such as ordering and preference), it is possible to optimize the alloy design strategy of a high entropy alloy for special usage in every condition, extreme environment especially. Traditional APT can only provide statistical distribution of components, but not for a quantitative independent analysis deep to a short-range configuration combined with environment around each atom. Thus, it is difficult or almost

impossible to analyze one of the most concerned questions in the field of structural materials: the impact of short-range ordered phenomena in high entropy alloys on the overall properties of alloys. By applying the most advanced Intelligent machine learning technology and big data analyses, we enable independent, personalized analysis of each of the tens of millions of atoms collected by APT, which includes analyses on the closest neighbor, capacity (ANP Index) of SRO bonds for each individual atom. Furthermore, we have successfully achieved the real-time observation of SRO through experimental methods for the first time. Further by improving the analytical methods with AI, we have identified the action and effect of SRO on the overall mechanical property of a certain material through experiments and theoretical analyses. In addition, we discovered the contribution and impact of how short chemical ordering on the overall mechanical properties of high entropy alloys through experimental and big data analyses. Based on different SROs, the effective regulation of the mechanical properties of high entropy alloys can be realized, and the SRO capacity and the overall properties of the materials will be organically linked. Using the algorithm of machine learning, it is realized for the first time, the SRO is observed in real time by apt and the SRO capacity and ratio given. We anticipate in future that we can process on the intelligent material design to have optimization of material properties via machine learning on high entropy alloys and other materials, such as energy-friendly material, vis our machine learning algorithm. We expect intelligent and smart design of future materials based on our finding of elements' inherent preference at the atomic level. It can be achieved in the future for Machine learning optimization of high entropy alloy design, so as for the other materials. And as a whole, we dedicate to make contributions on designing energy-saving, light but strong materials for low-cost industry applications.

Experimental And Computational Details

Material. The processing for the 3-component CoCrNi high-entropy alloy involved essentially identical procedures, namely high-purity elements (> 99.9% pure) which were arc melted under argon atmosphere and drop cast into rectangular cross-section copper molds. The ingots were then homogenized at 1200°C for 24 h in vacuum, cut in half lengthwise and then cold forged and cross rolled at room temperature along the side that is 25.4 mm to a final thickness of ~ 10 mm (total reduction in thickness of ~ 60%). Each piece was subsequently annealed at 800°C for 1 h in air leading to a fully recrystallized microstructure. They were drop cast, cold forged/cross rolled and recrystallized (at 800°C).

Atom probe tomography. Needle-shaped specimens required for APT were fabricated by lift-outs and annular milled in a FEI Scios focused ion beam/scanning electron microscope (FIB/SEM). The APT characterizations were performed in a local electrode atom probe (CAMEACA LEAP 5000 XR). The specimens were analyzed at 50 K in voltage mode, a pulse repetition rate of 200 kHz, a pulse fraction of 20%, and an evaporation detection rate of 0.5% atom per pulse. Imago Visualization and Analysis Software (IVAS) version 3.8 was used for creating the 3D reconstructions and data analysis.

Machine Learning algorithm. To find evidence to disprove this null hypothesis, we consider the "alien-philic"¹ Cr , Co , Ni atoms respectively. It naturally follows that, due to the randomness in null hypothesis, the proportion of "alien-philic" atoms of element e ,

$p[alien]_{-e} = \frac{totalnumberofalienphiliceatoms}{totalnumberofatoms} \leq 0.5$ were the null hypothesis true. Therefore, we calculate for all atoms the "alien-philic scores" for each k – size neighborhood, $p[alien]_{Cr}^k$, $p[alien]_{Co}^k$, $p[alien]_{Ni}^k$, respectively. Due to the ceiling operation of the threshold $\lceil \frac{2k}{3} \rceil$, there is a difference between the actual threshold and theoretical threshold of $\frac{2k}{3}$. This threshold is calculated as:

$\delta_{thres} = t_{actual} - t_{theory} = \lceil \frac{2k}{3} \rceil - \frac{2k}{3}$ is minimized with $k = 3a + 1$, achieving a $\delta_{thres(min)} = \frac{1}{3}$. Therefore, we only keep the "alien-philic scores" of three elements with $k = 3a + 1 \forall a \in \{1 \dots 16\}$.

The results of the calculations, the "alien-philic" scores display a stair-like decaying pattern as k increases, with $p[alien]_{Cr}^k \approx 0.6$, $p[alien]_{Co}^k \approx 0.6$, $p[alien]_{Ni}^k \approx 0.6$, suggesting an "alienphilic" distribution in the first layer (where $k \leq 12$).

MD simulation. Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS)[25] is employed for simulations. The interaction between SRO and $1/2[110]$ edge dislocation in CrCoNi-MEAs is computed by using a 500 nm (length)*300 nm (width)*20 nm (height) cuboid, which contains 200 expansion zones and 200 shrinkage zones. Free boundary conditions are imposed in z - $[11\bar{2}]$ direction, while the x - $[\bar{1}10]$ and y - $[111]$ directions are periodic. The forces on atoms in the out-most layer are zeroed to maintain the stability of the system. One $1/2[110](111)$ edge dislocation is created in the center of the simulation box. All simulations are performed with a fixed time step of 1 fs. The positions and velocities of other atoms are integrated on the Nosé-Hoover style non-Hamiltonian equations from isothermal-isobaric (npt) ensembles at 0.5 K.

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Declarations

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Author Contributions S.C, and C.T.L conceived the study. C.T.L supervised the study. S.C, H.Y. and R.F and processed the materials and the mechanical tests, and conducted the microstructure characterization. S.C, A.Z. and W.L. designed the algorithm. S.C, L.L. and C.T.L. discussed the data and interpreted the results. S.C.C, R.F. and C.T.L. wrote the main draft of the paper. A. X. G completed the MD simulation and the corresponding part of the writing. All authors contributed to the discussion of the results, and commented on the manuscript.

Competing Interests: The authors declare no competing interests.

Data

Additional information Extended data is available for this paper at <https://doi.org/xxx>.

Supplementary information is available for this paper at <https://doi.org/xxx>.

Figures

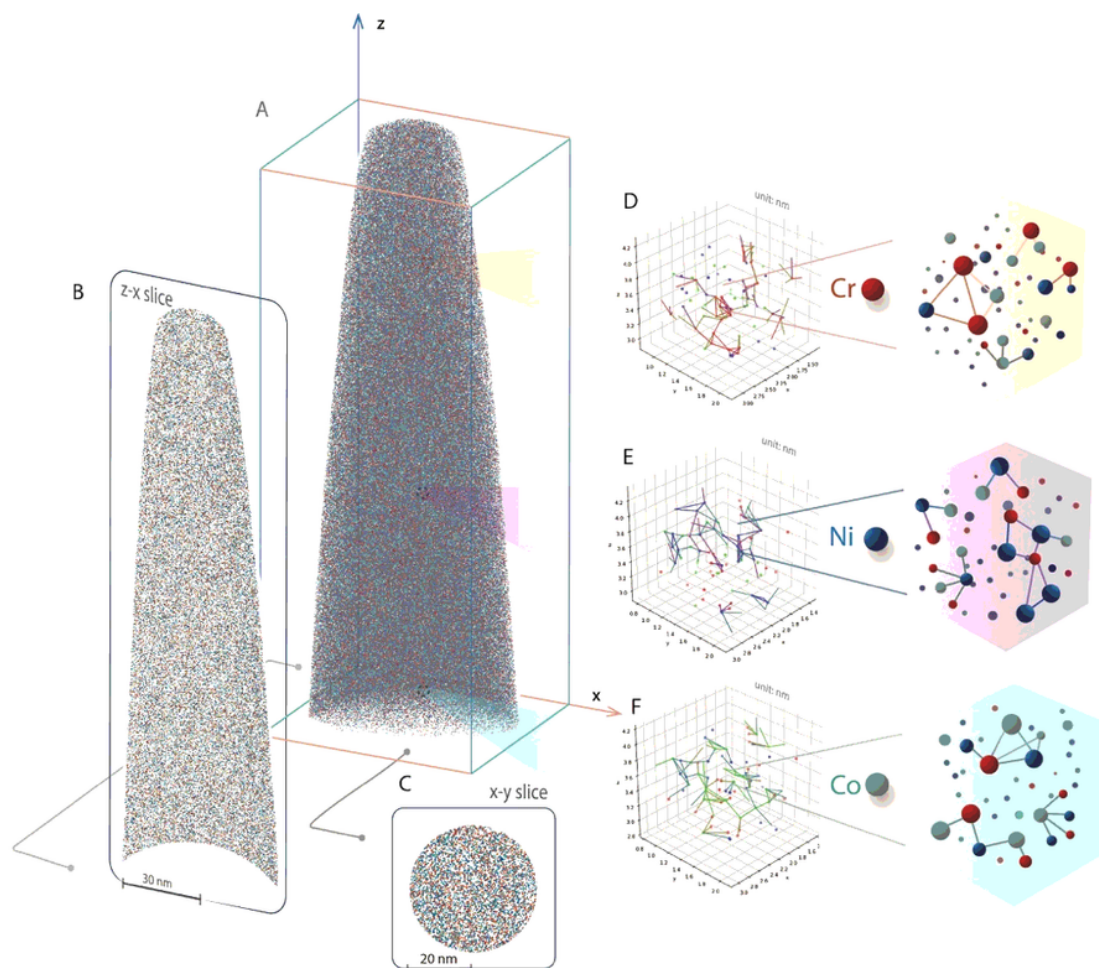


Figure 1

Three-dimensional Atom Probe tomography (APT). **A** is 3D atom maps for the distributions of all those three atoms Cr, Co and Ni in three dimensions. **B** and **C** show the atom distributions in the 1 nm-thick x-z slice and x-y slice respectively, in order to present a clearer view of the arrangement. **D**, **E** and **F** illustrate actual position with the nearest four atoms of each atom in the area for Cr, Ni and Co, respectively. From a macroscopic view, it seems that three elements are randomly distributed. However, by analyzing each atom individually on its nearest neighbors using intelligent big data analysis (shown in **D**, **E** and **F**), we find certain preference for each atom. From the zoomed in atom-bond visualization *Four-Neighbor Atomic Graph*, as presented in D, E, F, it seems that Cr favor Co and Ni rather than itself. Bonding band of Cr-Co and Cr-Ni are more frequent than Cr-Cr, Co-Co and Ni-Ni. *This observation motivates our algorithms in later discussions.* To quantify the preference, or capacity of SRO, we introduce an index called 'alien-philic index (API)[1]'. Concrete methodology is presented afterward.

[1] Due to imperfection of integer rounding, percentage values in 3k-1 and 3k (for all k) nearest atom ranges show oscillations in value, yet still comply with the general trend of the “ANP decay”.

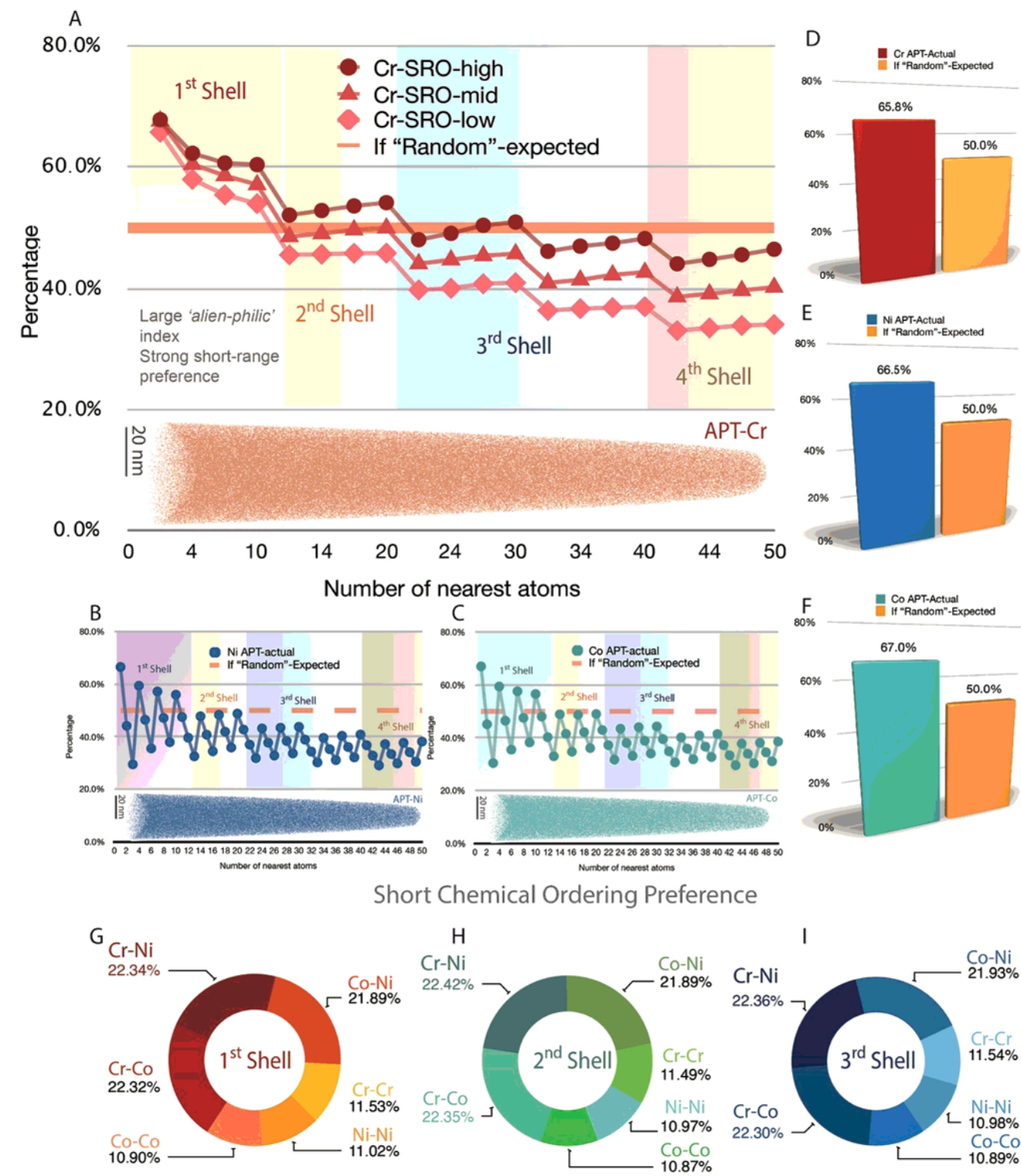


Figure 2

Machine learning APT data analysis. **A**, **B** and **C** are the “alien neighbor” percentages (ANPs), as the alien-philic index, contrasted with the ANP of an element with no affinity to alien atoms (50%) in the first nearest neighbor shell for Cr, Ni and Co, respectively. The ANPs have the same profile within the same shell, yet have a stepwise pattern across shells, with ANP being highest in the first shell, demonstrating an affinity for alien atoms.* [Note: Due to imperfection of integer rounding, percentage values in 3k-1 and 3k (for all k) nearest atom ranges show oscillations in value, yet still comply with the general trend of the “ANP decay”.] **D**, **E** and **F** show the ANP of the four nearest neighbor shells of Cr, Ni, Co, respectively. The three figures are all around 16% higher than their randomly-distributed counterparts. Distribution of neighbor preference in first, second and 3rd shell.

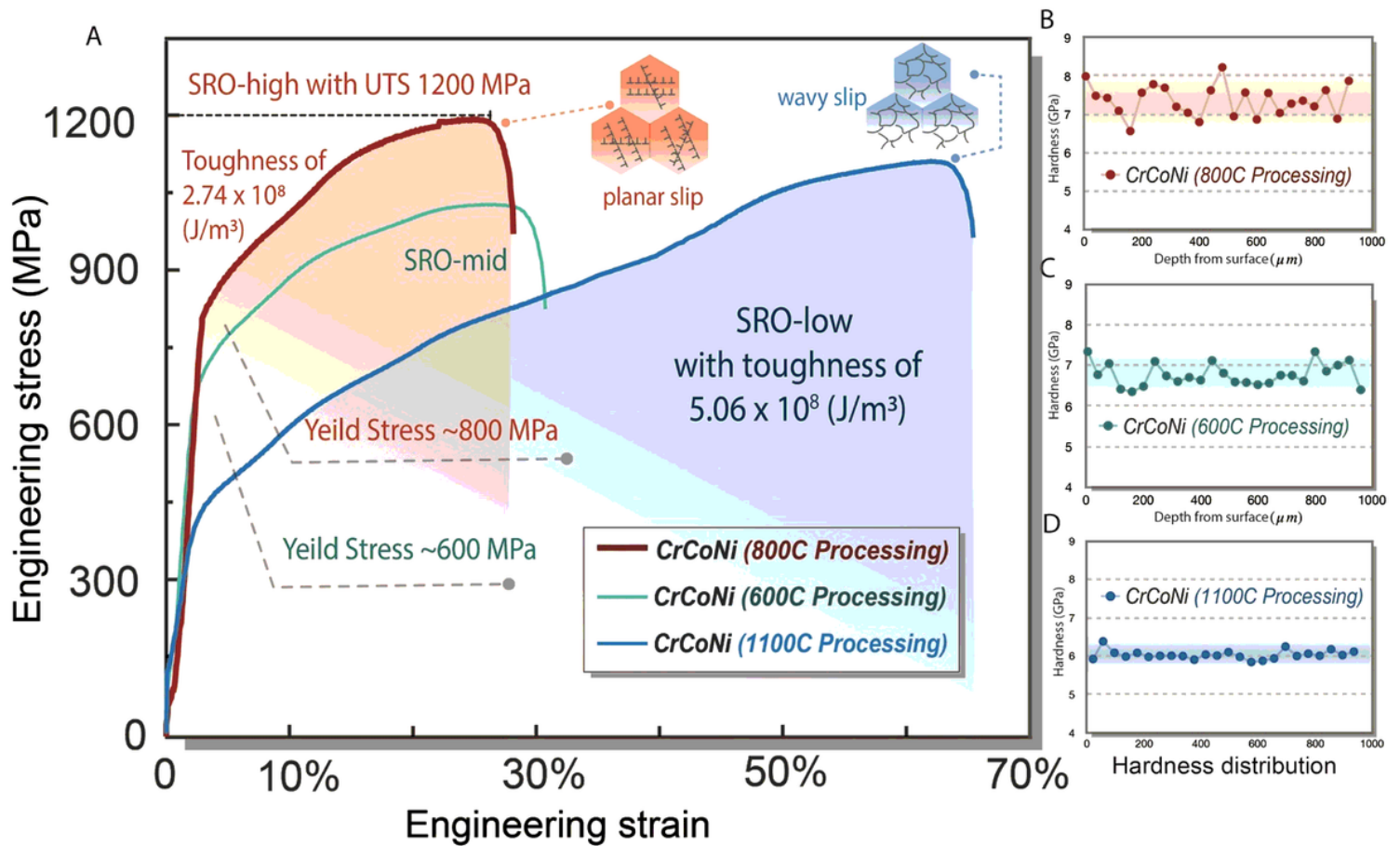


Figure 3

Mesoscopic behavior controls macroscopic performance: Tunable SRO capacity governs mechanical properties of medium-entropy alloy (CrCoNi). Uniaxial tensile tests show engineering stress–strain curves of CrCoNi in different processing temperature with various SRO capacities (600 °C in blue, 800 °C in red and 1100 °C in green scatterings, here the 600 °C, 800 °C and 1100 °C represent the annealing and quenching treatment with temperature of 600 °C, 800 °C and 1000 °C under xx min); sideward graphs **B**, **C** and **D** show the corresponding results from a nanoindentation hardness distribution of CrCoNi processed under high temperatures in 800 °C, 600 °C and 1100 °C , in red, green and navy respectively.

Clearly, from experimental results, there is an obvious trend and relationship between SRO with stress, hardness and toughness. *A trend of the relationship between SRO and stress curves can be observed; tunable SRO capacity makes adjustable stress and/or toughness; SRO-high contributes stress and hardness; SRO- low enhance toughness.*

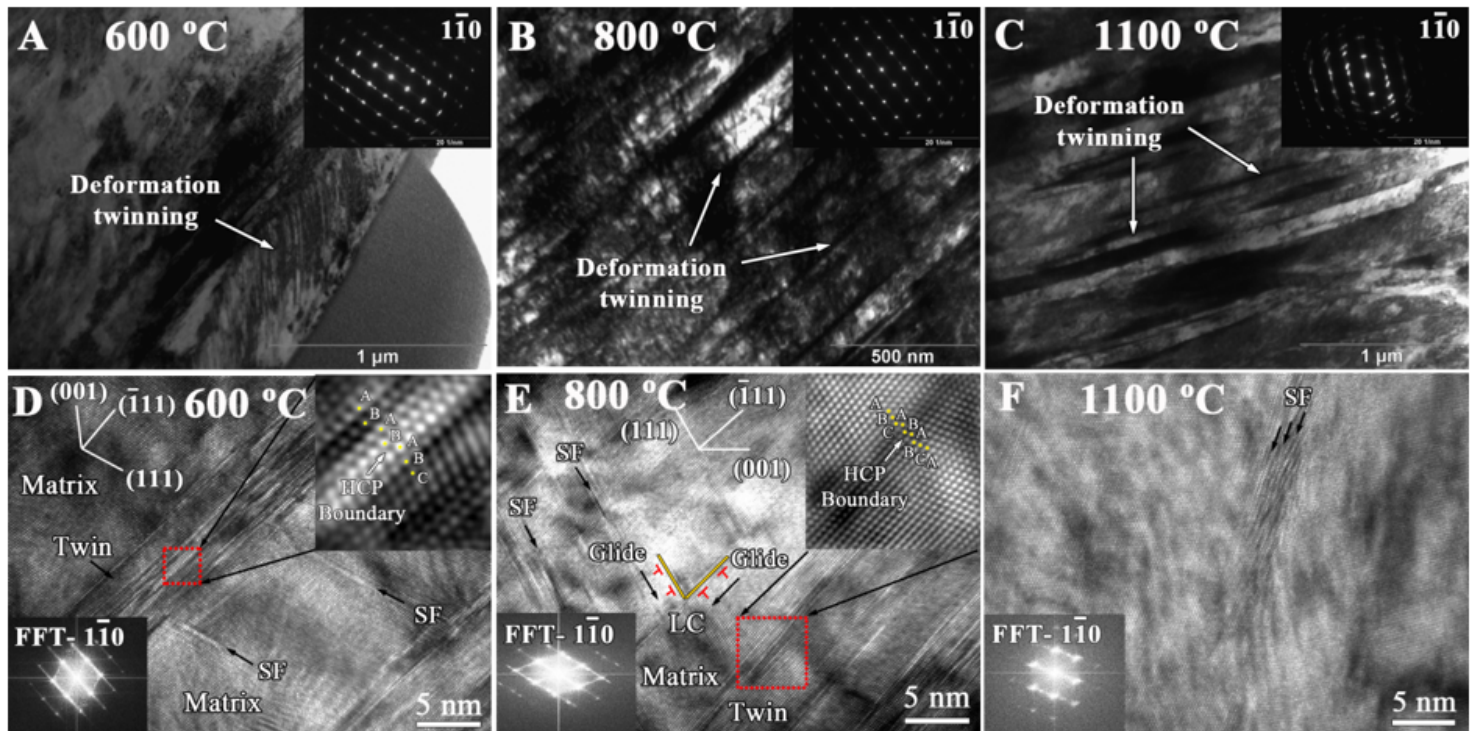


Figure 4

Intrinsic synergy and mechanism—TEM observations of the deformation mechanisms. **A, B, and C** are the bright-field (BF) images of the 600 °C, 800 °C, and 1100 °C annealed states, respectively, showing the presence of deformation twinning. **D, E, and F** are the high-resolution TEM (HRTEM) along zone axis of 600 °C, 800 °C, and 1100 °C, annealed states, respectively. The upper insets in **Figs. D and F** are the FFT-filtered HRTEM of the marked red-dot boxes, highlighting the formation of HCP structure at the nanotwin boundary of the 600 °C and 800 °C annealed states. Many stacking faults (SFs) are observed in the 600 °C and 800 °C states, while a few SFs are found in the 1100 °C annealed state. Moreover, many Lomer-Cottrell (LC) locks are formed in the 800 °C annealed state. In contrast, only some LC locks are found in the 600 °C state, and few LC locks are seen in 1100 °C state.

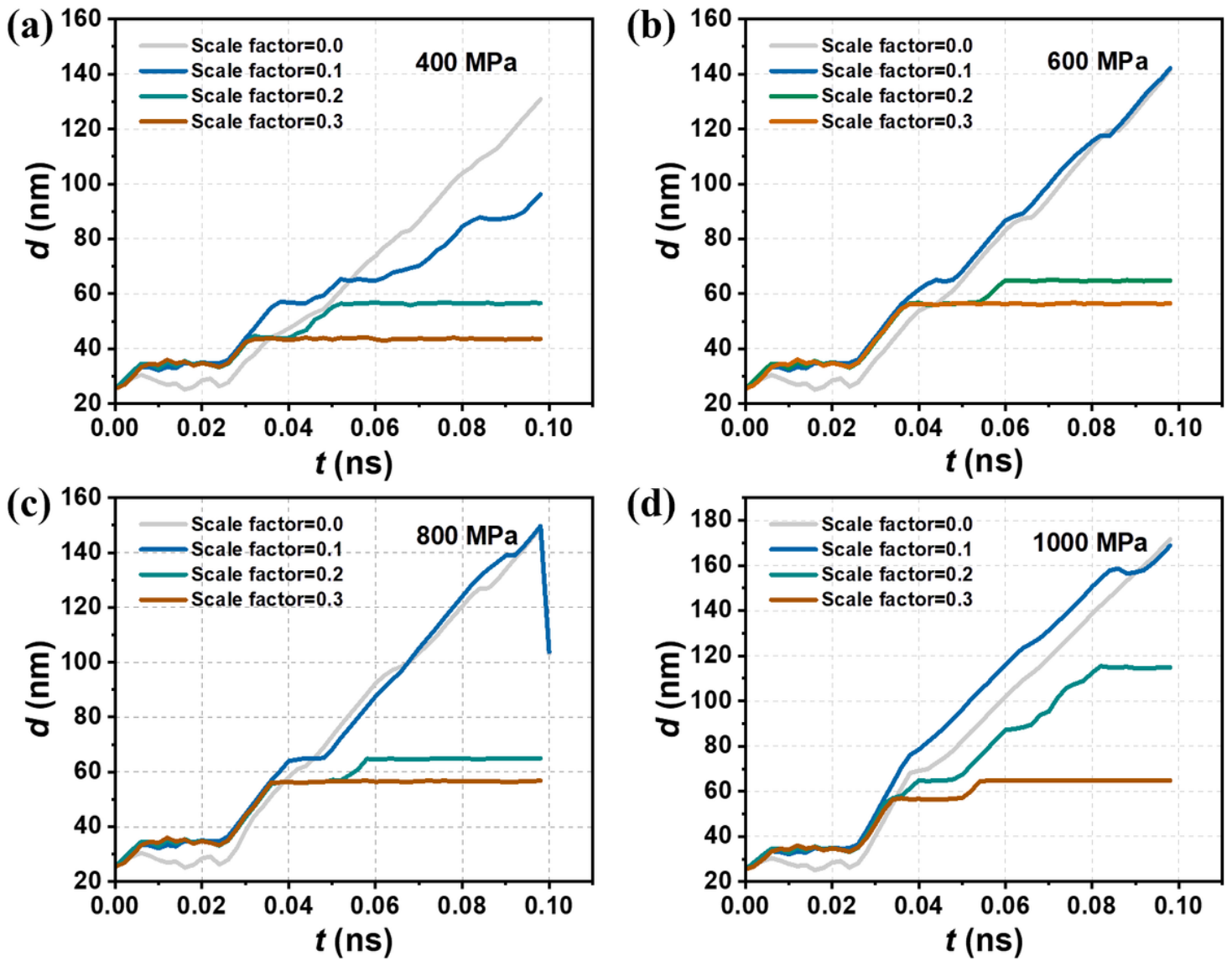


Figure 5

Comparison studies the $1/2\langle 110 \rangle\{111\}$ dislocation motion under external shear stress for $s = 0.0, 0.1, 0.2$ and 0.3 . (a) 400 MPa, (b) 600 MPa, (c) 800 MPa, (d) 1000 MPa.

Supplementary Files

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