




# Atomic-scale understanding of interstitial-strengthened high-entropy alloys

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**Abstract** Interstitial alloying has emerged as a powerful strategy to tune microstructure and microproperties of high-entropy alloys (HEAs) due to the strong interaction of interstitials with constituent elements and crystal defects, which enables the development of advanced alloys with superior mechanical and functional properties. The paper reviews the latest progress in the atomic-scale understanding of the effects of various interstitials, including carbon, boron, nitrogen, oxygen, and hydrogen, on the microstructure, stability, mechanical properties, and deformation behavior of HEAs. Emphases are placed on the in-depth insights on the interaction of interstitials with constituent elements and crystal defects, such as vacancies, stacking faults, and grain boundaries. Key parameters for rapid prediction of intrinsic properties of HEAs are also discussed. Finally, we highlight some unsolved issues and provide perspectives for future research directions.

**Keywords** High-entropy alloy; Interstitial; Atomistic modeling; Mechanical property; Deformation mechanism

## 1 Introduction

High-entropy alloys (HEAs), comprising multiple principal elements in equal or nearly equal concentrations, have

received enormous attention due to their unique microstructure and attractive properties [1, 2]. Originated from the “high entropy” concept, conventional HEAs primarily form substitutional solid solutions with a face-centered cubic (FCC), body-centered cubic (BCC), or hexagonal-closed packed (HCP) structure. The size and modulus mismatch among constituent elements contribute to a pronounced lattice distortion and substantial solid solution strengthening. Such a unique strengthening effect is considered to be the main cause of exceptional mechanical properties of HEAs, and various models and theories have been developed to describe the solid solution strengthening in HEAs. Recently, interstitial alloying is found to be very attractive in providing remarkable solid solution strengthening due to the large distortion induced by small-sized interstitials [3, 4]. Over the past decade, this strategy has been widely used in HEAs and provides tremendous opportunities in improving mechanical properties of HEAs [5].

Generally, interstitials can either segregate to grain boundaries (GBs) or reside in grain interiors in crystalline metallic materials. For the former, carbon, boron, and nitrogen usually form a predominantly covalent bond with the d-band of their neighboring metals, which increases the GB cohesion. By contrast, oxygen and hydrogen usually produce negative charged ions, which decreases the GB cohesion and promotes the intergranular embrittlement [6]. For the latter, carbon and nitrogen are found to provide a strong solid solution strengthening effect and sometimes induce transformation-induced plasticity (TRIP) and/or twinning-induced plasticity (TWIP) effects [7]. By carefully tailoring the metastability, interstitial-containing solid solution alloys can achieve a good combination of high strength and ductility. In addition to the solid solution

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strengthening effect, interstitials in HEAs can induce some unique structural features and novel properties. For example, oxygen and hydrogen are typically considered as harmful elements in conventional metallic materials, which can deteriorate the alloy ductility and toughness. In contrast, hydrogen was reported to suppress the embrittlement and improve mechanical properties of FeCoNiCrMn [8]. Oxygen was found to enhance both strength and ductility of refractory HEAs via forming ordered interstitial complexes (OICs) in chemical short-range order (CSRO) regions [9]. Moreover, some interstitials exhibit high flexibilities in tailoring structures and properties of HEAs. For instance, carbon can trigger either unidirectional [10] or bidirectional [11] FCC-to-HCP phase transformation in Fe<sub>49.5</sub>Mn<sub>30</sub>Co<sub>10</sub>Cr<sub>10</sub>. These interesting observations indicate high potential to design high-performance HEAs through interstitial alloying.

Understanding the underlying mechanisms behind the interstitial alloying and establishing a holistic framework of interstitial effects are crucial for further improving properties of HEAs. However, the mechanisms of the interstitial effects in HEAs are often complicated due to the complex atomic interactions. Computational models and simulations provide powerful tools for interpreting experimental observations from atomic-level insights and guiding materials design with reduced “trial-error” costs. Specifically, through simulating the interactions and arrangements of atoms, these tools provide detailed information on how interstitial alloying influences material behavior at the fundamental level. These atomistic insights are crucial for unraveling how interstitial additions alter the microstructure and properties of materials, thereby providing foundational knowledge and guidelines for the design of advanced materials with desired properties. Motivated by the recent advancement in modeling and simulation of interstitial-strengthened HEAs, this paper aims at providing an overview of the role of interstitials in the microstructure, phase stability, mechanical properties, and deformation behavior of HEAs through computational modeling and simulations. These methods examine the interstitial effects at an atomic level, offering valuable atomic and electronic insights that deepen our understanding of the intrinsic properties of interstitial-strengthened HEAs. The influences of various interstitials, such as carbon, boron, nitrogen, oxygen, and hydrogen, on the intrinsic properties (including solubility, local lattice distortion, and defects energies) of HEAs are systematically analyzed and discussed. Special attention is placed on the in-depth insights on the atomic interactions of interstitials with constituent elements and crystalline defects, such as vacancies, stacking faults (SFs), and GBs (Fig. 1). Finally, we discuss some fundamental and technological areas that should be further explored in future studies.

## 2 Solubility of interstitials in HEAs

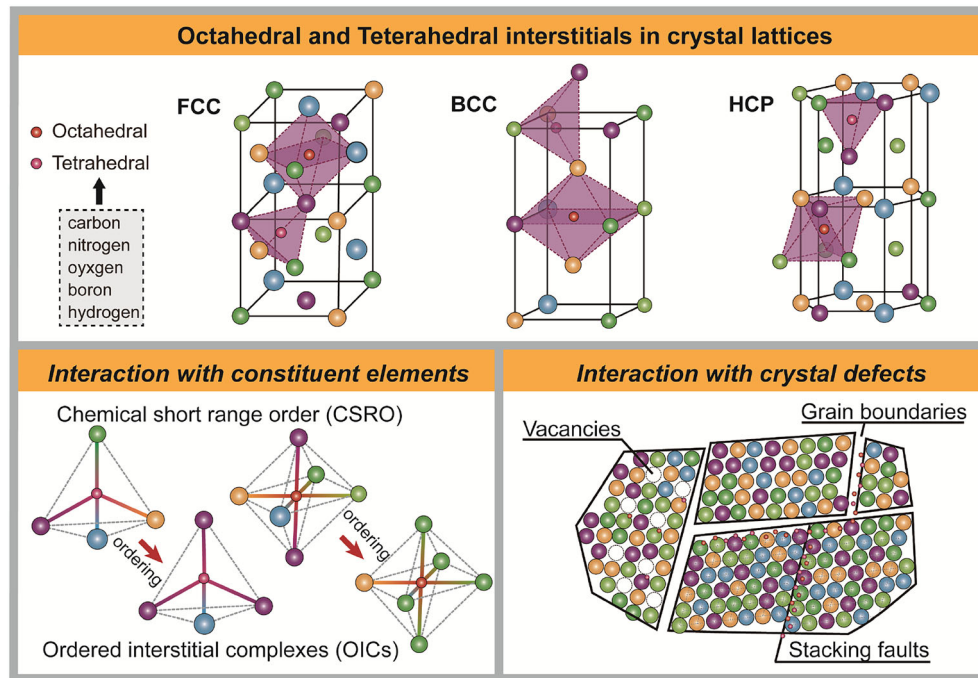
Unlike traditional metals and alloys that generally have a low solubility for interstitial atoms, HEAs can provide a substantially high solubility for them, particularly in refractory BCC HEAs. One of the major reasons for the enhanced solubility in HEAs is ascribed to the highly distorted lattice in these materials, which provides numerous interstitial sites. For FCC, BCC, and HCP crystals, the interstitial sites can be divided into two categories, namely octahedral and tetrahedral ones, as shown in Fig. 1. The solubility of interstitials in HEAs can be estimated from the solution energy,  $E_{\text{solution}}$ , which can be obtained by

$$E_{\text{solution}} = E_{(\text{HEA} + \text{interstitial})} - (E_{\text{HEA}} + E_{\text{interstitial}}) \quad (1)$$

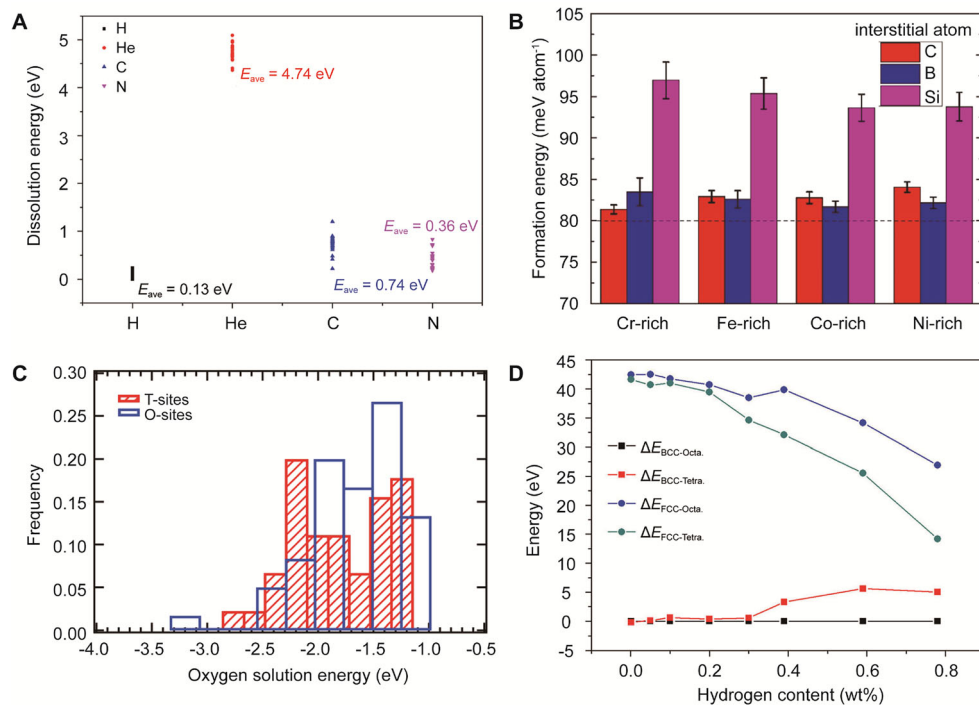
where  $E_{(\text{HEA} + \text{interstitial})}$  and  $E_{\text{HEA}}$  are the energies of the HEA per formula unit with and without one interstitial atom, respectively, and  $E_{\text{interstitial}}$  is the energy of the interstitial atom in its stable state.

Through density functional theory (DFT) calculations, it is found that hydrogen, carbon, nitrogen, and boron all prefer to occupy the octahedral sites in FCC HEAs [12–16]. Specifically, the order of the average formation energies for interstitials in FeCoNiCr is C (0.74 eV) > N (0.36 eV) > H (0.13 eV) [17], as displayed in Fig. 2A. It was found that the behavior of interstitial atoms in a solution is closely related to their local atomic configurations. For example, nitrogen exhibits low dissolution energies at sites with more chromium atoms, whereas carbon shows low dissolution energies at sites with fewer nickel atoms. This suggests a strong attraction between chromium and nitrogen and a significant repulsive interaction between carbon and nickel [16, 17]. Song et al. [16] examined the formation energy of doping carbon, boron, and silicon into FeCoNiCr to assess the phase stability of the materials (Fig. 2B). They found that Si-doped FeCoNiCr HEAs have significantly higher average formation energies compared to C- and B-doped HEAs, indicating that Si-doped FeCoNiCr HEAs are relatively more challenging to synthesize. The instability of Si-doped FeCoNiCr HEAs can be ascribed to the severe lattice distortion induced by the larger atomic radius of Si atoms [16]. Zhou et al. [18] found that HEAs with low hydrogen diffusion coefficients typically have high Co and Mn contents, whereas those with high hydrogen diffusion coefficients are rich in Fe and Ni. For refractory BCC HEAs, the probability of site occupancy for oxygen and nitrogen in the octahedral and tetrahedral sites is comparable (Fig. 2C) [9, 19, 20], whereas carbon prefers to occupy the octahedral sites [21].

It has been proposed that the highly fluctuated local chemical environment and the distorted lattice of HEAs



**Fig. 1** Schematic illustration of the available interstitial sites in crystal lattices and interactions of interstitials with constituent elements and crystal defects in HEAs



**Fig. 2** **A** Dissolution energies of hydrogen, carbon, and nitrogen for FeCoNiCr. Reproduced with permission from Ref. [17]. Copyright 2020, Japan Institute of Metals and Materials. **B** Formation energies of carbon and boron in different local chemical environments for FeCoNiCr. Reproduced with permission from Ref. [16]. Copyright 2022, Elsevier. **C** Statistical distribution of solution energy for oxygen at different interstitial sites in MoNbTaW. Reproduced with permission from Ref. [19]. Copyright 2020, AIP Publishing. **D** Dissolution energy difference between the octahedral and tetrahedral sites of FCC and BCC structures as a function of hydrogen content in TiZrHfNbMo. Reproduced with permission from Ref. [22]. Copyright 2020, American Chemical Society

result in a wide distribution of H diffusion energy barriers. The site occupancy of hydrogen in HEAs is quite complicated. Some studies suggested that hydrogen favors the tetrahedral sites in alloys such as FCC-TiZrHfMoNb [22], BCC-Ti<sub>23</sub>Cr<sub>25</sub>Ta<sub>23</sub>V<sub>29</sub>, BCC-Ti<sub>27</sub>Cr<sub>27</sub>Ta<sub>18</sub>V<sub>28</sub> [23], and BCC-NbMoTaW [24], whereas some reported that hydrogen prefers to occupy the octahedral sites in BCC-NbTiVZr [25], BCC-TiZrVMoNb [26], BCC-TiZrHfMoNb [22], FCC-TiVNbTa [27], FCC-TiZrVMoNb [26]. The possible reasons for the complicated site occupancy for hydrogen can be threefold: first, the introduction of hydrogen may induce a phase transformation from BCC to FCC structure in refractory HEAs, and hydrogen prefers to occupy the tetrahedral sites in BCC structures but the octahedral sites in FCC structures (Fig. 2D) [22, 26]. Second, hydrogen may occupy both tetrahedral and octahedral sites simultaneously of a structure [25, 27]. Third, the hydrogen occupancy at the octahedral sites is related to the vacancy formation [28]. Although the precise mechanism of this complex site occupancy is still unclear, it is interesting to note that some refractory HEAs show excellent potential for hydrogen storage applications. The hydrogen storage capability can be determined qualitatively from the phonon spectra method by assessing the lattice dynamic stabilities. To date, the reported HEAs with the maximum hydrogen storage capacity is NbTiVZr (2.94 wt%) [25], which is superior to most other BCC HEAs, such as NbTiVZrHf (2.7 wt%), NbMoVZrTi (2.65 wt%) [26], NbMoTiZrHf (1.94 wt%) [22], NbTiZrHfTa (1.68 wt%) [29], TiZrNbTa (1.4 wt%) [30], and MgZrTiFe<sub>0.5</sub>Co<sub>0.5</sub>Ni<sub>0.5</sub> (1.2 wt%) [31]. Moreover, the hydrogen storage capacity of NbTiVZr surpasses that of the well-known hydrogen storage alloy V<sub>20</sub>Ti<sub>28</sub>Cr<sub>52</sub> (2.6 wt%) [32]. These properties position refractory HEAs as promising candidates for future hydrogen storage applications.

In addition to lattice distortion, the solubility and segregation of interstitials are also highly dependent on the inhomogeneous distribution of electrons induced by the chemical effect [24–26]. Numerous studies found that the empirical parameter, valence electron concentration (VEC), combines these two contributions [12, 14, 20, 21], and a high VEC is generally corresponding to a large solubility. Furthermore, the solubility of hydrogen in refractory HEAs was calculated by employing the multi-objective Bayesian optimization-aided DFT calculations [33]; the results suggested that the thermodynamics of the first stage of desorption are determined mostly by the bulk modules, whereas those of the second stage are dominated by the number of states in the d-band. It is worth noting that a high d-band in the second stage that spans more of the Fermi level also exhibits a close dependence on low VECs.

### 3 Local structures and lattice distortion

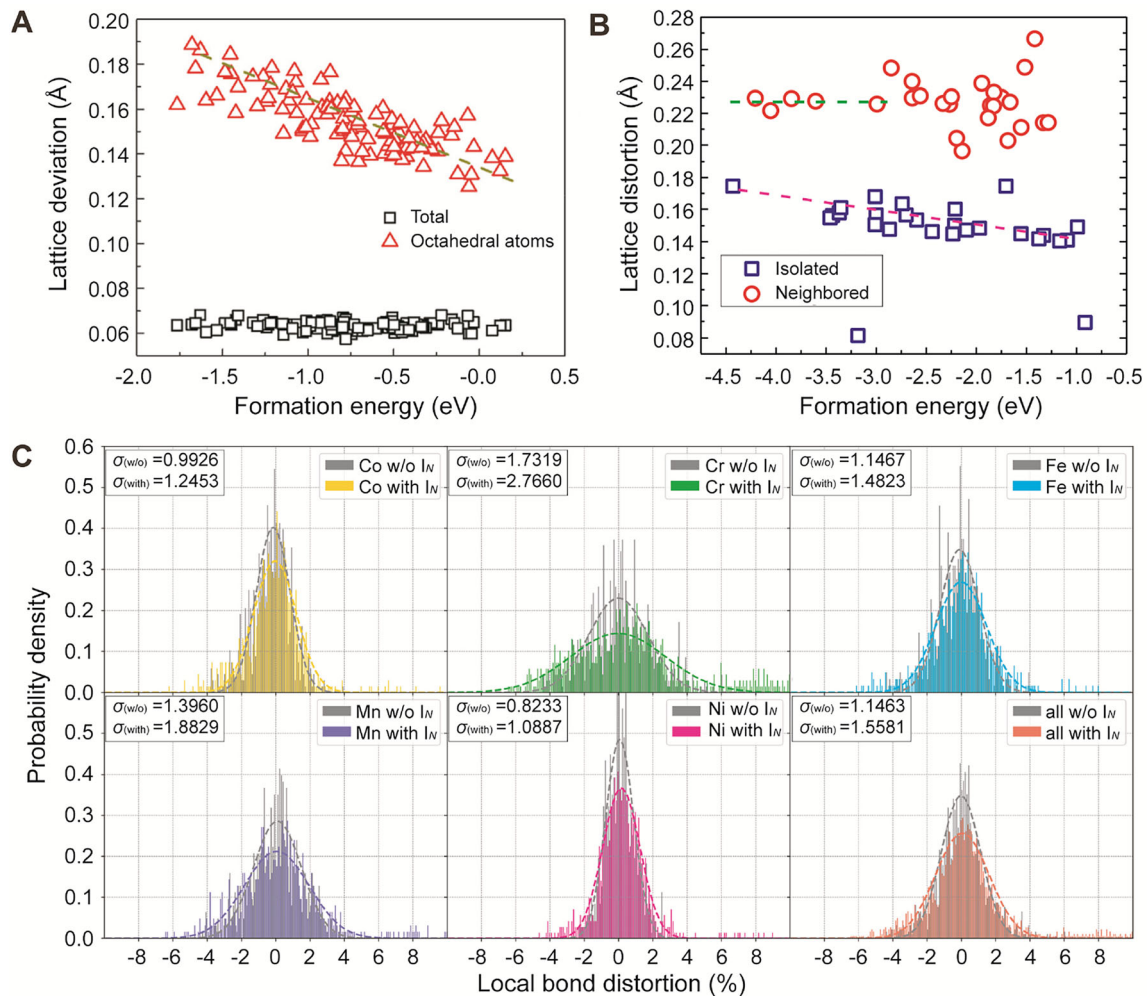
The high strength of HEAs is considered to be closely related to the severe lattice distortion. Experimentally, the lattice distortion can be measured by using X-ray or neutron diffraction techniques. However, such measurements usually ignore the contribution of chemical fluctuations and provide averaged information over all elements. Similar problems also exist for the empirical parameters, which generally do not consider the electronic and magnetic effects. Recently, a parameter  $\Delta d$  [34] based on DFT calculations was proposed to quantitatively describe the degree of local lattice distortion in HEAs, as expressed by

$$\Delta d = \frac{1}{N} \sum_i^N \sqrt{(x_0 - x_i)^2 + (y_0 - y_i)^2 + (z_0 - z_i)^2} \quad (2)$$

where  $N$  is the total number of interstitial atoms, and  $(x_0, y_0, z_0)$  and  $(x_i, y_i, z_i)$  are the Cartesian coordinate positions of atoms before and after the structural relaxation, respectively. It is found that the local lattice distortion in HEAs increases significantly with increased concentration of interstitials. Taking FeCoNiCr as an example, the fluctuation and magnitude of  $\Delta d$  induced by oxygen interstitials are much larger than those of global lattice distortion [15], as illustrated in Fig. 3A. Similar trends were also observed for carbon [35], nitrogen [36], and hydrogen [24] in FeCoNiCrMn, and carbon [21] and oxygen [20] in refractory HEAs. For conventional HEAs without interstitials, the origin of the local lattice distortion is disputed. Some studies suggested that the local lattice distortion derives mainly from the CSRO induced by the preferable bonding between certain atomic pairs [37], while others argued that the underlying mechanism is related to the charge transfer among constituent elements [38]. For interstitial-containing HEAs, the interaction of interstitial atoms with other host elements can induce the formation of complex CSRO. For example, carbon [12], nitrogen [14], oxygen [39], and hydrogen [40] were found to favor the Cr-rich regions while avoiding Ni-rich regions in FeCoNiCrMn. This additional CSRO was found to directly affect the local lattice distortion. As shown in Fig. 3C, the local bond distortions from of the Cr-rich environment in FeCoNiCrMn become wider for all elements with the addition of nitrogen. Such an effect can be enhanced by further doping carbon [41]. The competition between carbon and nitrogen compels carbon to diffuse from the (Cr,Mn)-enriched regions to the (Co,Fe)-enriched regions.

In addition to CSRO, the formation of OICs is also found to contribute to the local lattice distortion. As displayed in Fig. 3B, the ordered structures show larger lattice distortions compared to the disordered structures, and the degree of lattice distortions keeps nearly unchanged





**Fig. 3** **A** Local (triangles in red) and global (squares in black) lattice distortions with oxygen interstitials in FeCoNiCr. Reproduced with permission from Ref. [15]. Copyright 2020, Elsevier. **B** Local lattice distortions of ordered ('neighbored', circles in red) and disordered ('isolated', squares in blue) structures in FeCoNiCr with quadruple oxygen interstitials. Reproduced with permission from Ref. [15]. Copyright 2020, Elsevier. **C** Local lattice distortions with and without nitrogen interstitials for Cr-rich environment in FeCoNiCrMn. Reproduced with permission from Ref. [36]. Copyright 2021, Elsevier

responding to the variation of formation energies. It suggests that the OICs can be stable in oxygen-containing FeCoNiCr, which arouses severe lattice distortions [15, 41]. Since these local distortions break the high symmetry of interstitial positions and show strongly anisotropy strains, the resulted strengthening effect is more pronounced than that from substitutional solid solutions that typically have spherically symmetric stress fields. Zhou et al. [42] found that carbon-ordered structures can effectively inhibit dislocation motion, significantly enhancing the strength and hardness of alloys, while the stress field of dislocations can substantially increase the degree of ordering of within structures. Notably, OICs are frequently reported in refractory HEAs via adjusting the composition of CSRO regions [9, 43, 44]. Zhou et al. [45] found that the presence of OICs can significantly increase the critical shear stress needed for continuous dislocation

movement, with a pinning-cutting behavior observed when an edge dislocation encounters an OIC, and a cross-slip behavior occurring when a screw dislocation encounters an OIC. Unlike FeCoNiCrMn systems, refractory HEAs often have multiple types of CSRO regions. For instance, in HfNbTiZr, some CSRO regions are enriched in Ti and Zr and some are enriched in Hf and Ta. While the energy distributions for different regions are similar, both carbon and oxygen are found to favor Ti-enriched regions and dislike Nb-enriched regions [20, 21]. This result was further confirmed by the mean square displacement (MSD) based on molecular dynamics calculations. It was found that the MSD slope corresponding to Nb is much larger than that to other elements, indicating a large diffusion rate and unstable binding of Nb–O pairs. Therefore, decreasing the Nb content and increasing the concentration of

interstitials may be an efficient approach to introduce OICs in refractory HEAs.

The intrinsic origin for CSRO and OICs associated with local lattice distortion is attributed to the large difference between the interstitials and surrounding metallic elements in charge transfer [35]. The addition of oxygen and nitrogen in NbHfTiZr lowers the Bader charges of metallic atoms, because they can attract electrons from local environments [46]. This allows the original metal–metal bond to transform into ionic-like metal–interstitial bonds, thus enhancing the atomic cohesion. Moreover, some studies suggested that the charge transfer between atomic pairs is highly related to the magnetic moments of atoms by changing their unpaired spins [17]. In FeCoNiCrMn HEAs, it is found that the magnetic moments of Cr are increased near the interstitials, while those of other elements are decreased. The order of change is related to the effects of interstitials on charge transfer and bond lengths. The charge transfer potency of interstitials is found to follow the order of  $N > C > B > H$  in FeCoNiCr [16, 17].

## 4 Effect of interstitials on defect structures and energetics

### 4.1 Interstitial effects on vacancies

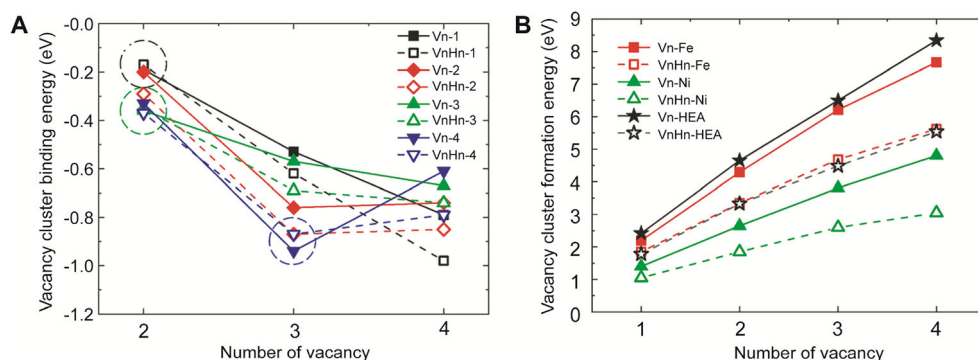
Vacancies are inevitable defects for metallic materials upon preparation or irradiation. They usually play multiple roles in affecting mechanical behaviors through facilitating atom diffusion, activating dislocation nucleation, hindering dislocation motion, and/or inducing crack initiation. It is found that the formation of monovacancies in FeCoNiCrMn is more difficult than the constituent elemental metals [47]. The addition of hydrogen significantly decreases the formation energy of vacancies and stabilizes the hydrogen–vacancy complex. The reason is ascribed to the charge transfer from metallic elements to their neighboring hydrogen atoms, which reduces the mean free path of electrons and energy dissipation rates. Upon deformation, the neighboring monovacancies can aggregate and grow into multi-vacancy clusters. Using the hydrogen enhanced vacancy stabilization model, Wang et al. [47] found that the binding energies of vacancies and hydrogen–vacancy complex in different local chemical environments are all negative for FeCoNiCrMn (Fig. 4A), indicating that the growth and coalescence of vacancies are spontaneous. Nevertheless, their growth behavior can be hindered in some cases when hydrogen is trapped (the dashed circles), which avoids the formation of large clusters. Besides, the growth rate of vacancies in FeCoNiCrMn is found to be lower than that in pure Fe and Ni, as shown in Fig. 4B.

These results suggest that multicomponent HEAs may yield exceptional resistance to hydrogen embrittlement.

Regarding boron interstitials, Wang et al. [48] calculated the vacancy formation energy of B-doped AlCrMoNbZr HEAs while considering different local chemical environments. They found no clear pattern in the vacancy formation energy relative to the variety and number of atoms in the local chemical environment, which can be attributed to the significant lattice distortion caused by boron doping. Consequently, the increase in the average vacancy formation energy in B-doped HEAs facilitates vacancy formation, which is not conducive to enhancing the initial irradiation resistance performance. As for carbon interstitials, the vacancy migration energy is significantly increased in Fe<sub>49.5</sub>Mn<sub>30</sub>Co<sub>10</sub>Cr<sub>10</sub>C<sub>0.5</sub> HEAs [35]. The values are weak negative related to the average metallic radii of the local surrounding environment [28, 49] but associated with the magnetic states and chemical compositions. In Fe<sub>50</sub>Mn<sub>30</sub>Co<sub>10</sub>Cr<sub>10</sub>, the vacancy migration energy for carbon interstitials follows the order of  $Co > Fe > Mn > Cr$  [35], leading to nanoscale localized elemental fluctuations and often implying stronger local lattice distortion. As a result, the carbon interstitials hinder the vacancy diffusion and increase the local lattice distortion, thereby enhancing the recombination of radiation-induced point defects and inhibiting irradiation hardening [35].

### 4.2 Interstitial effects on generalized SFs

The plastic deformation behavior of HEAs is highly dependent upon their stacking fault energies (SFEs). Generally, a low SFE ( $< 20 \text{ mJ m}^{-2}$ ) leads to martensitic transformation, while a high SFE ( $> 40 \text{ mJ m}^{-2}$ ) results in dislocation gliding and shear banding, and SFEs in the range of  $20\text{--}40 \text{ mJ m}^{-2}$  can induce deformation twinning [50]. The calculated SFEs of equimolar FeCoNiCrMn HEAs are in the range of  $-93$  to  $-5 \text{ mJ m}^{-2}$  at 0 K [51], suggesting that the dominant deformation mechanism is the martensite transformation at this temperature. It is found that doping hydrogen to HEAs [8, 13] decreases the SFEs, while adding carbon [12] and nitrogen [14, 52] to HEAs increases the SFEs. In conventional FCC alloys, it is generally recognized that carbon increases the SFEs, whereas the effect of nitrogen on SFEs can vary, either increasing, decreasing, or having minimal effects, depending on the specific alloy compositions. Many studies suggested that the excellent resistance to hydrogen embrittlement for FeCoNiCrMn is related to the reduced SFEs by hydrogen, which induces the formation of nanotwins and SFs [53]. Zhu et al. [8] found that the addition of hydrogen reduces the energy barrier for twin nucleation by using first-principles calculations. Their experimental observations confirmed that a higher density of deformation twins was



**Fig. 4** **A** Binding energies of vacancy clusters and hydrogen-vacancy clusters at different positions in FeCoNiCrMn; **B** formation energies of vacancy clusters and hydrogen-vacancy clusters in pure Fe and Ni and FeCoNiCrMn. Reproduced with permission from Ref. [47]. Copyright 2022, Elsevier

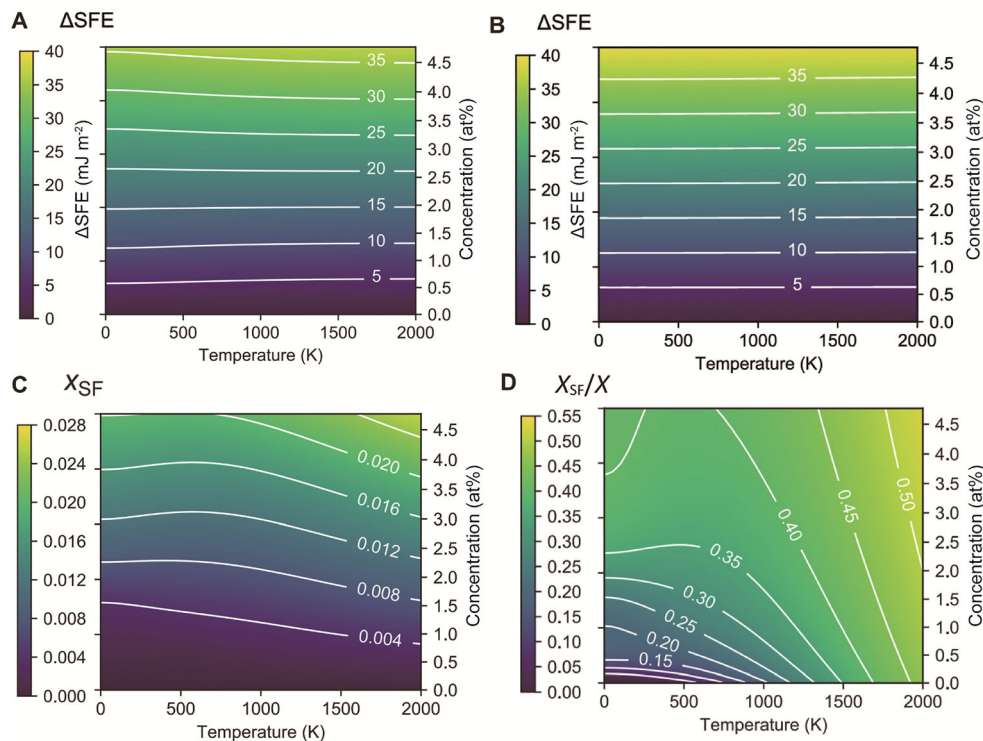
generated in the FeCoNiCrMn HEAs under a lower hydrogen concentration environment, resulting in a larger strength and a higher ductility after hydrogen charging. Luo et al. [54] experimentally found that while hydrogen alloying does not increase the yield strength of FeCoNiCrMn HEAs, it does enhance the strain-hardening rate. This indicates that the beneficial effect of nanotwins on strain hardening primarily improves both strength and ductility.

Carbon and nitrogen are found to increase the SFE of HEAs, which turns the TRIP into TWIP, sometimes even dislocation slips, depending on the content of soluble interstitials. Specifically, as shown in Fig. 5A, B, the increase of SFE is qualitatively evaluated as  $8 \text{ mJ m}^{-2}$  for 1 at% of nitrogen [14], which is close to that of carbon ( $7 \text{ mJ m}^{-2}$ ) [12]. However, they exhibit distinct effects on the deformation behavior of FeCoNiCrMn HEAs. Nitrogen is found to provide solid solution strengthening effect but has little impact on the deformation mode [55], whereas carbon is reported to trigger such deformation mechanisms as phase transformation, deformation twinning, and dislocation slip [10, 56]. Li et al. constructed various configurations for nitrogen-containing FeCoNiCrMn HEAs and found that there is an average increased SFE of approximately  $147 \text{ mJ m}^{-2}$  [52] for 1 at% nitrogen. Notably, Chen et al. recently found that the SFE of  $\text{Fe}_{50}\text{Mn}_{30}\text{Co}_{10}\text{Cr}_{10}$  presents a sophisticated dependence on the concentration of nitrogen, as the structure of  $(\text{Fe}_{50}\text{Mn}_{30}\text{Co}_{10}\text{Cr}_{10})_{100-x}\text{N}_x$  evolves from a single FCC-to-FCC + HCP dual phase, and ultimately reverts to a predominantly FCC phase with increasing nitrogen [57]. Additionally, carbon shows an obvious anti-Suzuki behavior, which tends to diffuse away from the SF planes in FeCoNiCrMn [12], similar to that in  $\gamma$ -Fe and TWIP steels [58]. As displayed in Fig. 5D, the ratios of local carbon in the vicinity of SFs and in the bulk region are all less than 1 within the temperature range of 0–2,000 K. Typically, the addition of carbon increases the

SFEs of FCC alloys, such as TWIP steels, Fe–Mn–C–Al steels, and Cantor alloy and its derivatives, resulting in a reduced tendency for twinning. Li et al. [56] experimentally observed that the density of nanotwins decreases as the carbon content increases from 0 to 0.8 at% in CoCr–FeMnNi HEAs at room temperature. Meanwhile, the interstitial HEA with 0.8 at% carbon exhibits more than five times the yield strength of the C-free reference alloy. The significant increase in strength can be attributed to the substantial interstitial solid solution strengthening, which is related to the strong interactions between interstitial carbon and dislocations. Importantly, tuning the SFE of alloys through interstitials provides an effective method for precisely designing their deformation mode. For example, adding approximately 0.5 at% carbon to a non-equiatomic TRIP-assisted dual-phase HEA results in the combined activation of TWIP and TRIP effects [59].

#### 4.3 Interstitial effects on GBs

GBs also play an important role in controlling mechanical properties and deformation behavior of HEAs. Based on DFT calculations, Guan et al. investigated the segregation behavior of hydrogen at  $\Sigma 3(111)$  GBs of CoNiCr and FeCoNiCr alloys [60]. It is found that the solution energies of hydrogen at GBs of CoNiCr are higher than those in the bulk, suggesting that hydrogen is hard to segregate to GBs. The reason is ascribed mainly to the existence of Cr, which shrinks the charge density isosurface and limits the movement of neighboring hydrogen atoms. Moreover, the dipole moment interaction between Cr and hydrogen compels hydrogen atoms at interstitial sites away from each other, which improves the resistance to hydrogen embrittlement. Nevertheless, the introduction of Fe weakens this suppression effect, improves the solubility of hydrogen at GBs, and alters the diffusion process of hydrogen. The negative solution energies and low binding



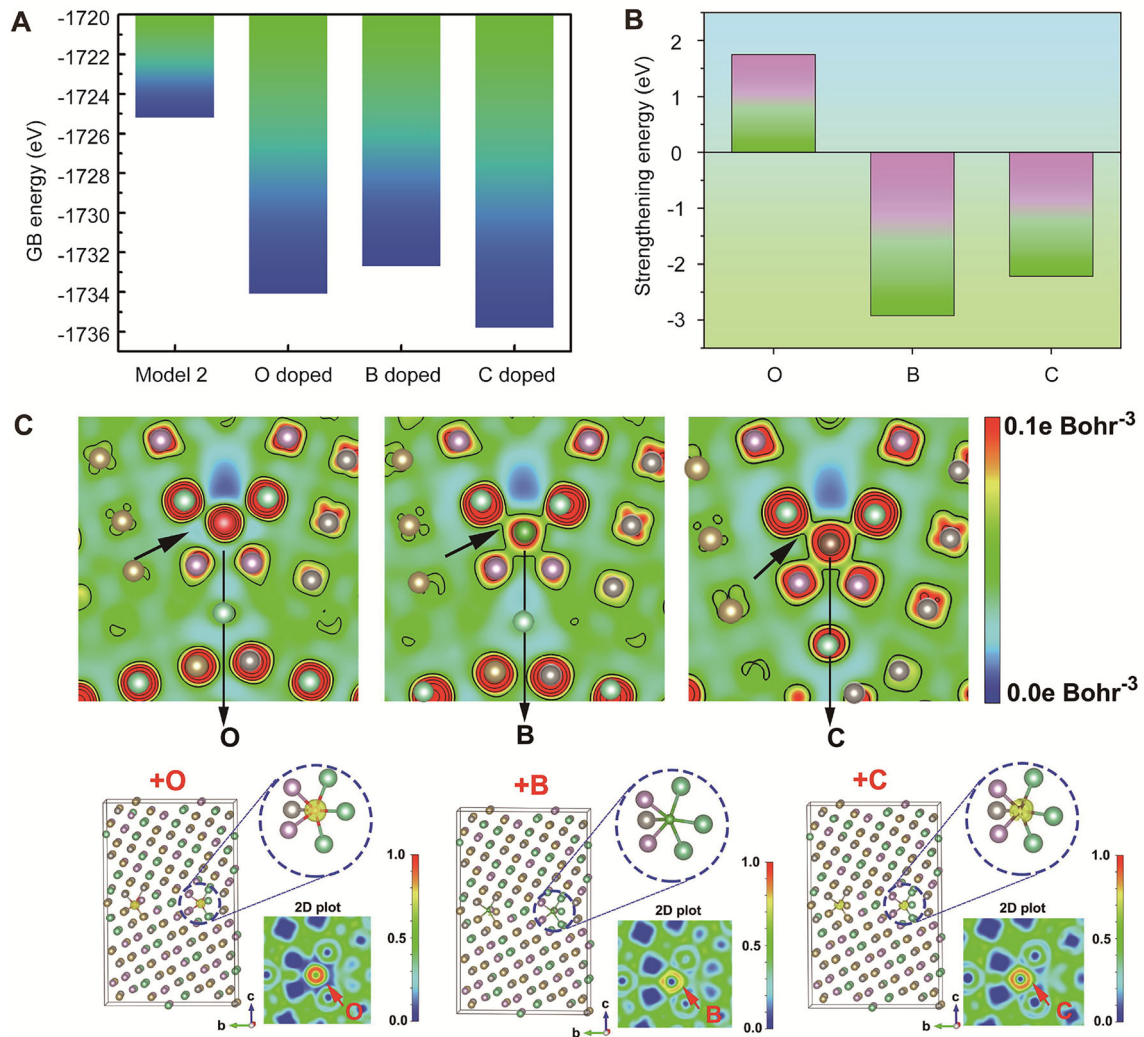
**Fig. 5** Impacts of **A** carbon and **B** nitrogen on the SFEs of FeCoNiCrMn as a function of temperature and concentration. Reproduced with permission from Ref. [12]. Copyright 2019, American Physical Society and Ref. [14]. Copyright 2021, Springer. **C** Ratios of the number of carbon atoms to that of available interstitial sites at the stacking faults ( $X_{SF}$ ) as a function of temperature and carbon ratio in the bulk region ( $X$ ), and **D** ratios of  $X_{SF}$  to  $X$  for FeCoNiCrMn HEAs. Reproduced with permission from Ref. [12]. Copyright 2019, American Physical Society

energies of hydrogen in FeCoNiCr suggest that the dissolution of hydrogen at GBs is spontaneous but the interaction among hydrogen atoms is weak. In addition, Fe is found to block the migration channels of hydrogen at GBs while promoting the diffusion of hydrogen from the bulk to GBs. Despite those, the high migration barriers and the large lattice distortion make the diffusion of hydrogen at GBs in FeCoNiCr difficult. By analyzing the density of states (DOS) before and after hydrogen charging, it is found that hydrogen makes little effects on the d electron density of Ni and Co while providing additional flexibility of electron deformation for Cr or Fe.

By using three-dimensional atom probe tomography (3D-APT), Yuan et al. reported that the GBs are concentrated with more than one type of interstitials in TiZrHfNb alloys [61]. The DFT calculations of GB energies for three different  $\Sigma 3(112)[001]$  configurations suggest that the atomic distribution of GBs is not random or uniform, and the structure with Ti and Zr segregation is very stable. The segregation of boron, carbon, and oxygen to GBs can reduce the GB energy and stabilize the GBs, following the order of  $C > O > B$  (Fig. 6A) [61]. Along with the co-segregation of Zr and Ti at GBs, these interstitials cause severer embrittlement. Moreover, oxygen at GBs is

suggested to be the main cause of intergranular cracking in NbMoTaW HEAs [62]. In contrast to the positive strengthening energy of oxygen shown in Fig. 6B, carbon and boron are found to yield negative values at the typical  $\Sigma 5(310)[001]$  GBs, which manifests the enhanced GBs cohesive. In addition, the more negative segregation energies and higher diffusion rate suggest that carbon and boron are more inclined to segregate to GBs. Thus, the introduction of carbon and boron can effectively eliminate the segregation of oxygen at GBs, thereby enhancing the cohesion of GBs. The underlying mechanisms were explained by the results of charge density distribution and the electron localization function (ELF), where the additions of boron and carbon lead to an abundant charge accumulation, strong interaction with adjacent metallic elements, and the transformation from ionic to metallic bond (Fig. 6C). The enhanced GB bonding in the B/C-doped HEAs significantly inhibits crack initiation at GBs. Consequently, crack propagation becomes more challenging due to the higher energy barrier and the complex interactions with the distorted lattice. As a result, the deformability of the B/C-doped HEAs is dramatically enhanced [61].



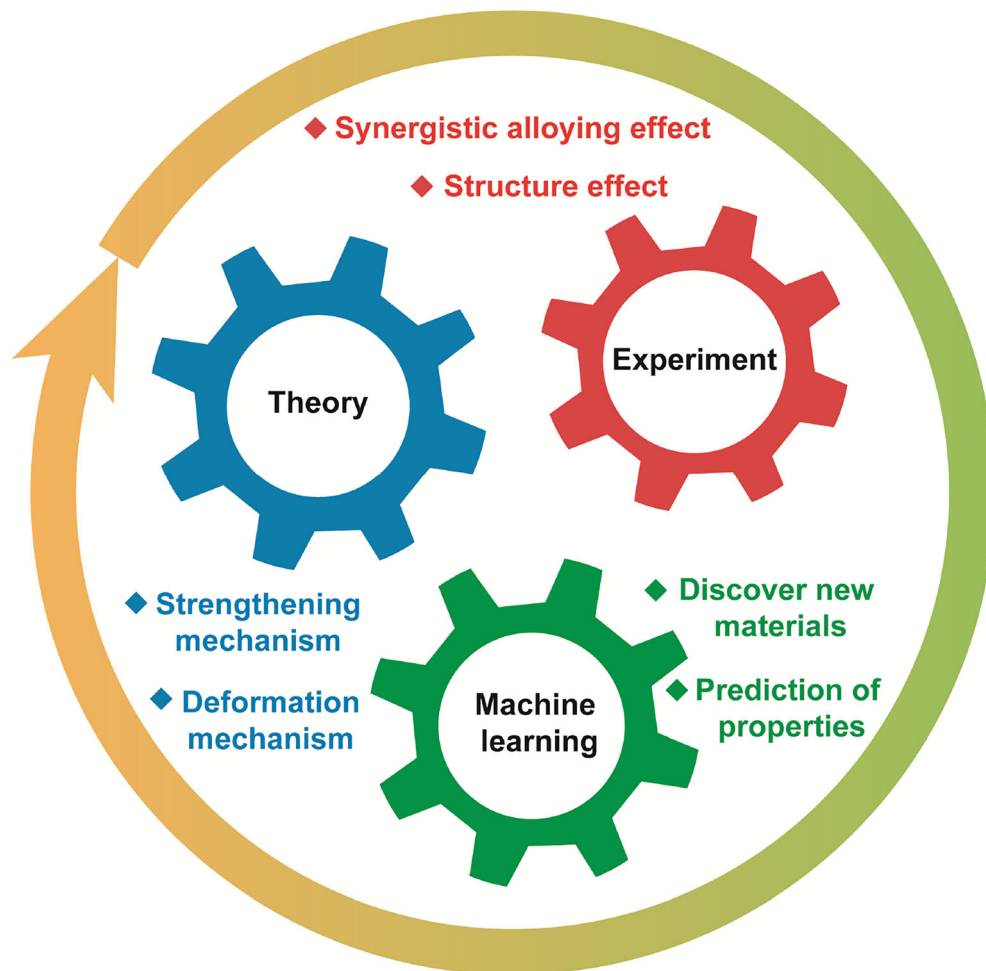


**Fig. 6** **A** GB energies of  $\Sigma 3(112)[011]$ GBs for TiZrHfNb with and without oxygen, boron, and carbon. Reproduced with permission from Ref. [61]. Copyright 2023, Elsevier. **B** Strengthening energies and **C** charge density distribution, ELF of  $\Sigma 5(310)[001]$ GBs for NbMoTaW with oxygen, boron, and carbon. Reproduced with permission from Ref. [62]. Copyright 2022, Elsevier

## 5 Concluding remarks and future outlook

The atomic understanding of interstitial effects in HEAs has been greatly improved by computational modeling and simulations. The interactions of interstitials with constituent elements and defect structures show great potential in tuning mechanical properties of HEAs. The effect of interstitials on the lattice structure of HEAs are analyzed from the solubilities, site preference, bonding, and charge transfer. It is found that the strengthening effects of interstitials are highly depending on local chemical environments. Particularly, the interstitial-induced local lattice distortions and OICs can contribute substantially to the strengthening of HEAs. Moreover, interstitials also have an important impact on the defect structures and associated defect energetics, which can remarkably affect the plastic deformation and fracture behavior of HEAs. Therefore, the

control of interstitials and their interactions with constituent elements and defect structures provides new opportunities for optimizing mechanical properties and deformation behavior of HEAs. Further investigations are required for better understanding and control over properties of interstitial-strengthened HEAs from the theory, experiment, and machine learning aspects (Fig. 7). From a theoretical perspective, it is crucial to gain a deep understanding of the mechanisms behind strengthening and deformation. On the experimental front, it is particularly intriguing to explore how multiple interstitials and ordered structures influence the mechanical properties of interstitial-strengthened HEAs. In parallel, machine learning offers a powerful tool for predicting material properties and discovering new materials.



**Fig. 7** Outlook on future research of interstitial-strengthened HEAs

### 5.1 Theoretical analysis and modeling of strengthening and deformation

The interaction of interstitial atoms with dislocations, twins, and SFs is complex and multifaceted. These interactions can significantly influence the deformation behavior of materials, affecting their mechanical properties. As a result, there is a need to develop new theoretical frameworks and models to better understand and predict dislocation activities. Such advancements could lead to improved material design and performance by providing deeper insights into the fundamental mechanisms governing material deformation. Moreover, an accurate evaluation of interstitial solid solution strengthening is crucial for the fundamental understanding of mechanical properties of HEAs. However, classical models, such as Fleischer and Labusch models, are found not accurate enough to describe the relationship between the concentration of interstitials and increased yield strength of HEAs [63]. These models often rely on parameters like lattice mismatch and shear modulus mismatch, which are not well-defined or sufficient

for complex HEAs. Moreover, additional factors, such as severe lattice distortion, complex phase interactions, and non-linear effects, might also influence the strengthening effect. Thus, it is vital to establish a physical model suitable for interstitial-strengthened HEAs for a better understanding of and eventually control over the interstitial solid solution strengthening in HEAs.

### 5.2 Mechanistic understanding of alloying and structure effects

Due to the severe lattice distortion, HEAs can usually absorb more than one type of interstitials. Recent findings suggest that the mechanical properties of HEAs can be efficiently tuned by co-doping of multiple interstitials. For example, the synergy of interstitial carbon and nitrogen in FeCoNiCr is found to significantly enhance the maximum shear stress [64]. Additionally, the competition between carbon and hydrogen is found to suppress the hydrogen trapping at GBs, thereby alleviating the hydrogen embrittlement [65]. However, the synergistic effects of multi-interstitials and their

interactions on the mechanical properties of HEAs are quite complicated and remain to be elucidated an in-depth understanding of which would facilitate the design of novel alloys with superior properties.

In addition to disordered HEAs, interstitials have also been utilized in some ordered multicomponent alloys. A typical example is an FCC/L1<sub>2</sub> Ni<sub>43.9</sub>Co<sub>22.4</sub>-Fe<sub>8.8</sub>Al<sub>10.7</sub>Ti<sub>11.7</sub>B<sub>2.5</sub> alloy [66], in which a disorder layer enriched in Fe, Co, and boron is formed at GBs. This layer allows for the dislocation transmission across GBs, thus resulting in excellent combination of high strength and ductility. Inspired by these exciting results, a fundamental understanding of the interstitial effects on the structure and properties of ordered multicomponent alloys, such as L1<sub>2</sub>, B2, L1<sub>0</sub>, and L2<sub>1</sub> alloys, may provide tremendous opportunities for developing strong-yet-ductile ordered intermetallic alloys.

### 5.3 Machine learning of interstitial-strengthened HEAs

Available data suggest that a change in interstitial type and concentration may bring a significant variation in the phase stability and deformation behavior of HEAs. Thus, the exploration of unexplored composition space in interstitial-strengthened HEAs is urgently needed. Recently, the combination of machine learning and DFT calculations has attracted increasing interest from researchers, which has been successfully applied to accelerate the discovery of some novel interstitial-strengthened HEAs [18, 33], demonstrating high capability in predicting mechanical properties and guiding the design of new high-performance HEAs. In addition, machine learning can also be utilized to develop the interatomic potentials for the Monte Carlo-Molecular Dynamics (MC-MD) method with a large atomic scale. Specifically, deep learning potentials, trained on extensive datasets, can capture the nuanced behaviors of atoms in these alloys with high precision. This advanced approach allows researchers to integrate MD simulations with deep learning potentials, enabling more accurate and efficient predictions of various material properties. By leveraging the computational power and adaptability of deep learning, scientists can overcome previous limitations and unlock the full potential of HEAs in various applications.

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**Data availability** The data that support the findings of this study are available from the corresponding author upon reasonable request.

### Declarations

**Conflict of interests** The authors declare that they have no conflict of interest.

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