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# Acta Materialia



# Full length article A highly transferable and efficient machine learning interatomic potentials study of $\alpha$ -Fe–C binary system

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ABSTRACT

of defects was conducted.

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# 1. Introduction

Steels remain to be arguably the most common structural materials in the world as human civilization advances from the Iron Age to the ongoing Silicon Age. The overwhelming dominance of steels stems partly from the fact that there is endless variety of microstructures and properties that can be generated by solid-state transformation and processing. Our knowledge of its microstructure evolution and structure performance relationship, nevertheless, is still far from complete. As coal or other fossil fuels are widely used to heat the iron ore in traditional steelmaking processes, carbon (C) is almost always present in steels, either as intentional alloying element or as unwanted impurity. In fact, the addition of mere carbon to iron is sufficient to form a steel. A small concentration of carbon, e.g. 0.1–0.2 weight per cent, has a great strengthening effect on iron [1].

To fundamentally understand the role played by C atoms in steel, it is important to characterize its thermodynamics and kinetics at the atomistic level [2]. Density Functional Theory (DFT) calculations are generally capable of yielding reliable results, yet still encounter challenges in handling large systems or long-time molecular dynamics simulations, despite remarkable progress in computational capabilities achieved over recent decades. Empirical interatomic potentials (EIPs) can significantly reduce computational costs, enabling the simulation of extensive systems over prolonged periods, albeit with a trade-off in precision. Numerous interatomic potentials with diverse formalisms pertaining to the Fe–C system have been developed [2–4], significantly advancing our comprehension of the influence of carbon on iron [5]. These empirical interatomic potentials were constructed through the amalgamation or hybridization of existing disparate interaction terms and refitting part of parameters. Some of the limitations of the base potential were inherited by the new potential. For instance, correctly representing the energy landscape of screw dislocation remains a challenge for all Fe-related empirical potentials [6]. The limitations of EIPs are attributed to their rigid formalisms.

Machine learning interatomic potentials (MLIPs) for  $\alpha$ -iron and carbon binary system have been constructed

aiming for understanding the mechanical behavior of Fe–C steel and carbides. The MLIPs were trained using an extensive reference database produced by spin polarized density functional theory (DFT) calculations. The

MLIPs reach the DFT accuracies in many important properties which are frequently engaged in Fe and Fe-

C studies, including kinetics and thermodynamics of C in  $\alpha$ -Fe with vacancy, grain boundary, and screw

dislocation, and basic properties of cementite and cementite-ferrite interfaces. In conjunction with these MLIPs,

the impact of C atoms on the mobility of screw dislocation at finite temperature, and the C-decorated core

configuration of screw dislocation were investigated, and a uniaxial tensile test on a model with multiple types

Recently, several types of machine learning interatomic potentials (MLIPs) have been proposed for various materials utilizing diverse structure descriptors, such as but not limited to Behler–Parrinello neural network potential (BNNP) [7,8], Gaussian approximation potential (GAP) [9], Spectral neighbor analysis potential (SNAP) [10], Deep potential (DP) [11], and Graph neural network potential (GNNP) [12], etc.. For the performance and computational cost assessment of different MLIPs, please refer to Refs. [13–15]. Given that the primary target

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of this work is on topics related to the plasticity of structural materials, requiring the use of a sizable model, computational speed emerges as the paramount factor for the development of potentials. We developed two MLIPs under frameworks of BNNP and DP for  $\alpha$ -Fe and C binary system based on a comprehensive database produced by spin-polarized DFT calculations. The constructed MLIPs exhibit excellent DFT compatibility on  $\alpha$ -Fe, Fe<sub>2</sub>C, ferrite-cementite interfaces, and interactions of C with pure and defective  $\alpha$ -Fe, such as vacancies, grain boundaries (GBs), and screw dislocations. In conjunction with these MLIPs, the impact of C atoms on the mobility of screw dislocations at finite temperature, and the C-decorated core configuration of screw dislocation, were investigated, and a tensile test for a model with various types of defects, including vacancies, interstitial atoms, GB, cementite, and ferrite/cementite interfaces, was conducted. These demonstrations can be considered as evidence for the good transferability of these MLIPs. For these two MLIPs, the BNNP presents better overall accuracy than the DP, while the DP provides an advantage over BNNP in scenarios requiring the computation of atomic stress.

# 2. Construction of MLIPs

Utilizing the spin-polarized DFT calculations, 32 002 configurations, equivalent to  $2.78 \times 10^6$  atomic environments, were prepared in the database (see Table.S1 in the supplementary materials [16]). These configurations were distributed randomly into a training data set (90%) and a testing data set (10%). Both energy and atomic force were considered in the training processes while the virial tensor was not.

For the BNNP framework [7], to parameterize the local atomic environments (LAEs) of the configurations in reference database, radial and angular atom centered symmetry functions (ACSFs) [17] was adopted. The radial symmetry function is defined as [17]

$$G_i^{\text{rad}} = \sum_{j} e^{-\eta (R_{ij} - R_s)^2} f_c(R_{ij}), \tag{1}$$

where  $R_{ij}$  is the atomic distance between atom *j* and the central atom *i*. The angular symmetry functions is given by [17]

$$G_{i}^{\text{ang-w}} = 2^{1-\xi} \sum_{j} \sum_{k \neq j} (1 + \lambda \cos \theta_{ijk})^{\xi} e^{-\eta (R_{ij} + R_{ik})^{2}} f_{c}(R_{ij}) f_{c}(R_{ik}),$$
(2)

and,

$$G_{i}^{\mathrm{ang-n}} = 2^{1-\xi} \sum_{j} \sum_{k \neq j} (1+\lambda \cos \theta_{ijk})^{\xi} e^{-\eta(R_{ij}^{2}+R_{ik}^{2}+R_{jk}^{2})} f_{c}(R_{ij}) f_{c}(R_{ik}) f_{c}(R_{jk}),$$
(3)

where  $\theta_{ijk}$  is the angle enclosed by the vectors of  $R_{ij}$  and  $R_{ik}$  of two neighboring atoms *j* and *k*, respectively. Both types of ACSF have a common function  $f_c$  called the cutoff function, which is defined as follows:

$$f_{c}(R_{ij}) = \begin{cases} \tanh^{3} \left[ 1 - \frac{R_{ij}}{R_{c}} \right] & \text{if } R_{ij} \le R_{c} \\ 0.0 & \text{if } R_{ij} > R_{c}, \end{cases}$$
(4)

The parameters of  $\eta$ ,  $R_s$ ,  $\xi$ , and  $\lambda$  in equations Eqs. (1)–(3) can be determined by the strategy reported in Ref. [18]. Each element has 16 radial and 60 (48  $G_i^{ang-n}$ +12  $G_i^{ang-n}$ ) angular ACSFs. The cutoff radii for radial and angular ACSFs of Fe atoms are specified as 6.5 and 6.0 Å, respectively. For C atoms, they are set at 5.5 and 5.0 Å, respectively. The hidden layer was set to be 2 layers, each of which has 15 neurons.

For the DP framework [19], a hybrid descriptor with two-body embedding full-information ( $se_e2_a$ ) and three-body embedding DeepPot-SE ( $se_e3$ ) was employed. We used (32, 64, 128) neurons for two-body embedding networks, (8, 16, 32) neurons for three-body embedding networks, and (256, 256, 256, 1) neurons for fitting networks. The hyperbolic tangent function was selected as the activation function. Atoms of Fe and C shared the same cutoff radius ( $r_c$ ) of 6.5 and 5.6 Å for the descriptors of  $se_e2_a$  and  $se_e3$ , respectively, and the same smooth cutoff parameter ( $r_{cs}$ ) of 0.5 Å.

All DFT calculations were performed using Vienna Ab-initio Simulation Package (VASP) [20]. The BNNP was trained using a neural network potential package (n2p2) [21] and the DP was constructed with the help of the DeePMD-kit package (Version 2.2.3) [19]. The potentials validation and application were carried out by Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [22,23]. Simulation visualization and atomistic figures plot are completed with the help of OVITO [24]. PHONOPY code [25] was used to calculate the phonon dispersion curves, the code of Atomsk [26] was employed to build screw dislocation models. Detailed information on database (Table. S1 [16]), the DFT calculations used for preparing the database as well as conducting molecular statics simulations in the validation section is available in the supplementary materials (Table. S2) [16].

# 3. Results: Validation of MLIPs

#### 3.1. Overall accuracy of MLIPs

Overall accuracy of the MLIPs was estimated using root mean squared errors (RMSEs) of energy (*E*) and force (*F*) which definition can be found elsewhere [27,28]. The training and testing data sets have RMSE(*E*)s of 3.58 and 3.57 meV/atom, and RMSE(*F*)s of 83.0 and 83.2 meV/Å, respectively, for BNNP. Those are 4.71 and 5.87 meV/atom and 92.5 and 91.2 meV/Å, respectively, for DP. The values of these RMSEs are in the typical range of machine learning based interatomic potentials [29,30]. Comparison of DFT and BNNP produced energies and forces of the structures in the training and testing data sets are displayed in Fig. 1(a)–(b), those comparison of DP are presented in Fig. 1(c) and Figs. 1(d)–(f), respectively. All points for energy and force of training and testing datasets are distributed along the line with a slope of 1, indicating that the trained data can be well reproduced and no overfitting occurred. Several concerned properties are further checked in the following subsections.

#### 3.2. MLIPs Performance for $\alpha$ -iron

The BNNP and DP performance for basic properties of  $\alpha$ -iron, including lattice constant, elastic constants, defects formation energies, and low index surface energies, are tabulated in Table 1. And thermal expansion curve and  $\gamma$ -surfaces of (110) and (112) planes are presented in supplementary materials (Figs. S1–S2) [16]. All of them show similar performance as our previous Fe–H BNNP [31] and well agree with DFT results [32–42].

Phonon dispersion curves of  $\alpha$ -iron with equilibrium lattice condition are presented in Fig. 2(a). It is clear that both BNNP and DP can well reproduce the DFT [45] and experimental results [47].

For an iron-related interatomic potential, the accurate description of the screw dislocation core configuration and its energetics is paramount [45,51]. A supercell with dimensions of  $22[11\bar{2}] \times 38[1\bar{1}0] \times$ 1/2[111] was employed, periodic boundary condition was applied in the [111] direction only. A 1/2[111] screw dislocation was introduced according to the anisotropic elasticity theory of dislocation [26]. Three distinct core configurations, including easy core (EC), hard core (HC), and split core (SC) were created separately, the differential displacement maps (DDmaps) of each core were presented in the supplementary materials (Fig. S3) [16]. To map the 2-dimensional Peierls energy landscape, additional configurations among these core configurations were generated using the interpolation method. Subsequently, the atomic positions of the model were relaxed, excluding the atoms located more than 70 Å from the dislocation core. z coordinates of the atoms in the core were fixed to maintain the core configuration. The 2-Dimensional Peierls energy landscape predicted by BNNP is displayed in Fig. 2(b) (See Fig. S3 in the supplementary materials [16] that predicted by DP). It is clear that there is only one hump between neighboring easy cores, and the whole shape matches the reported DFT results [52]. Taking the energy of the dislocation with EC as reference, the energies of



Fig. 1. Comparisons of BNNP and DFT (a) energies, (b) forces of the configurations in the training and testing datasets. The comparison of DP and DFT (c) energies, (d)–(f) forces. The line with a slope of 1 indicates a perfect training.



**Fig. 2.** MLIPs performance for *a*-iron, (a) Phonon dispersion curve, (b) 2-Dimensional Peierls energy of screw dislocation. (c) Misorientation-energy relationship for the symmetric till grain boundaries with a tilt axis of (110), (d) Twist angle-energy relationship for the twist grain boundaries with termination of (110). Available reported DFT [42,45] and experimental [47] results for phonon dispersion curves and DFT results for GB formation energy [48–50] are also shown.

Properties of  $\alpha$ -Fe produced by BNNP and DP. Results are compared to DFT (from this work and reported literature) and other machine learning interatomic potentials of iron. The subscripts of *i*NN for the bi-vacancy is the *i*<sup>th</sup> nearest neighbor, and those of tri-vacancy are identified by means of Beeler notation [43]. The subscripts of [111] and Tet for quad-vacancy are vacancies arranged along [111] crystal direction or in tetrahedral configuration. For the self interstitial atoms, T and O stand for the interstitial Fe atom following the tetrahedral and octahedral occupation, and [001], [110], and [111] are the dumbbells formed along those crystal directions.

	BNNP	DP	DFT (This work)	DFT (In literature)	Fe–H-BNNP [31]	Fe-H-DP [44]	ANN [45]	GAP [42]
Lattice Constant (Å)								
$a_0$	2.828	2.831	2.830	2.834[ <mark>42</mark> ]	2.830	2.834	2.836	2.834
Elastic constants (GPa)								
<i>C</i> <sub>11</sub>	270	268	297	297[42]	296	280	277	285.9
C <sub>12</sub>	135	134	151	151[42]	147	128	156	154.3
C <sub>44</sub>	92	97	105	109[42]	96	104	104	103.8
Vacancies energy (eV)								
$E_{1V}^{\mathrm{f}}$	2.183	2.216	2.223	2.22[42]	2.203	2.327	2.287	2.26
$E_{2V-1NN}^{f}$	4.217	4.303	4.236	4.24[42]	4.184	4.542	4.071	4.41
$E^{\rm f}_{\rm 2V-2NN}$	4.119	4.214	4.201	4.20[42]	4.183	4.322	3.813	4.30
$E^{\rm f}_{\rm 2V-3NN}$	4.367	4.388	-	4.45[ <mark>42</mark> ]	4.445	4.597	4.581	4.55
$E_{2V-4NN}^{f}$	4.283	4.409	-	-	4.374	4.598	4.385	4.48
$E_{2V-5NN}^{f}$	4.369	4.431	-	-	4.378	4.649	4.307	4.47
$E_{3V-112}^{f}$	5.848	6.053	-	5.82[ <mark>38</mark> ]	5.867	6.234	-	6.19
$E^{\rm f}_{\rm 3V-226}$	6.083	6.203	-	6.13[ <mark>38</mark> ]	6.146	6.318	-	6.38
$E^{\rm f}_{3{ m V}-223}$	6.231	6.442	-	6.70[ <mark>38</mark> ]	6.155	6.249	-	6.35
$E_{3V-115}^{f}$	6.243	6.398	-	6.15[ <mark>38</mark> ]	6.152	6.684	-	6.47
$E_{3V-113}^{f}$	6.211	6.369	-	6.14[ <mark>38</mark> ]	6.185	6.664	-	6.59
$E^{\rm f}_{\rm 3V-333}$	6.560	6.645	-	-	6.665	6.820	-	6.85
$E_{4V-[111]}^{f}$	8.274	8.494	8.182	-	8.114	8.823	-	-
$E_{4V-Tet}^{f}$	7.238	7.557	7.254	7.51[46]	7.278	8.026	-	-
Self-interstitial energy (eV)								
$E_{i-T}^{f}$	4.431	4.617	4.466	4.79[ <mark>42</mark> ]	4.448	4.688	4.871	4.75
E	5.258	5.191	5.279	5.58[42]	5.322	5.166	5.703	5.53
$E_{i-[110]}^{f}$	4.026	4.186	4.023	4.37[42]	4.037	4.200	4.350	4.21
$E_{i-1}^{f}$	4.723	4.556	4.579	5.13[ <mark>42</mark> ]	4.744	4.611	5.023	4.90
$E_{i-[001]}^{f}$	5.146	5.042	5.146	5.48[ <mark>42</mark> ]	5.062	5.038	5.603	5.47
Surface energy (J/m <sup>2</sup> )								
$\gamma_{(001)}$	2.479	2.487	2.488	2.543[ <mark>42</mark> ]	2.479	2.501	2.538	2.547
$\gamma_{(110)}$	2.426	2.433	2.449	2.495[ <mark>42</mark> ]	2.436	2.461	2.474	2.499
$\gamma_{(111)}$	2.667	2.692	2.691	2.752[ <mark>42</mark> ]	2.695	2.686	2.719	2.756
γ <sub>(112)</sub>	2.590	2.592	2.575	2.629[ <mark>42</mark> ]	2.586	2.601	2.623	2.612

dislocation with HC, middle point (MP), and SC are 38.5, 36.7, and 110.9 eV/b (b: Burgers vector length,  $b = \sqrt{3}/2 \times a_0$ ) respectively. Those obtained by DP are 51.0, 35.4, and 82.3 meV/b, respectively. These results show good agreement with DFT results of 57.9, 49.2, and 110.3 meV/b [53], 39.3, 37.9, and 108 meV/b [54], and result of 47.4, 38.3, and 82.3 meV/b obtained by our previously developed Fe–H BNNP [31]. Employing a 40b length screw dislocation model and CI-NEB method [55], kink-pair nucleation enthalpy predicted by BNNP is 0.72 eV and DP is 0.71 eV, which is consistent with the results of the DFT-based line tension model of 0.73 eV [54], 0.86 eV [56], 0.91 eV [57], and the result of 0.7 eV (Fe–H BNNP [31]). Note that only the aforementioned configurations of EC, MP, HC and SC are included in the training data set.

Formation energy versus tilt and twist angles for  $\langle 110 \rangle$  tilt symmetric grain boundaries (GBs) and (110) twist GBs at T = 0 K are displayed in Figs. 2(c)–(d), respectively. Computational scheme of GB energy has been described in detail elsewhere [58,59]. It is not surprising that the BNNP and DP can effectively capture both seen and unseen data regarding tilt GBs [48–50], similar to our prior BNNP for Fe–H binary system [31]. The BNNP and DP also can accurately capture both seen and unseen data regarding twist GBs even in the absence of specific data in the database. Performance of the BNNP and DP for other GBs are presented in supplementary materials (Fig. S4) [16].

### 3.3. MLIPs Performance for carbon in perfect and defective $\alpha$ -iron

#### 3.3.1. Interaction of carbon with perfect $\alpha$ -iron

Solution energy ( $E_s$ ) of C atom in perfect  $\alpha$ -iron at T = 0 K was studied employing a 4×4×4 BCC ( $\alpha$ -iron) supercell, and the calculations

were conducted in the following manner.

$$E_{\rm s} = E_{\rm FeC} - E_{\rm Fe}^{\rm BCC} - \mu_{\rm C},\tag{5}$$

where  $E_{\rm FeC}$  and  $E_{\rm Fe}^{\rm BCC}$  stand for energies of the perfect  $\alpha$ -iron supercell with and without C atom, and  $\mu_{\rm C}$  indicates the chemical potential of C atom in the state of diamond. With the help of BNNP (resp. DP),  $E_{\rm s}$  of C atom at T- and O-site are 0.682 (0.789) and 1.563 (1.567) eV/C-atom, respectively, align well with DFT findings of 0.65 and 1.74 eV/C-atom [59].

The solubility of carbon atoms at T = 0 K in response to volumetric engineering strain is also subjected to testing, which is not included in the database. The BNNP predicted that the C solution energy gradually decreases from 1.627 eV/C-atom at the strain of -5.88% to 0.069 eV/C-atom at +6.12% which revealed that the local carbon concentration could be significantly influenced by local volume expansion or compression. The solution energy is formed by the distortion in the Fe lattice and the bonding of C to surrounding Fe atoms, namely mechanical ( $E_s^{\text{Mech}}$ ) and chemical ( $E_s^{\text{Chem}}$ ) contributions, which can be found following Ref. [60].

$$E_{\rm s}^{\rm Mech} = E_{\rm Fe}^{\rm frz} - E_{\rm Fe}^{\rm BCC},\tag{6}$$

and,

$$E_{\rm s}^{\rm Chem} = E_{\rm FeC} - E_{\rm Fe}^{\rm frz} - \mu_{\rm C},\tag{7}$$

where  $E_{\rm FeC}$  and  $E_{\rm Fe}^{\rm frz}$  indicate energies of relaxed C-soluted Fe and the frozen Fe lattice which is derived from the relaxed C-soluted Fe with the removal of the C atom, respectively.

As tabulated in Table 2, both mechanical and chemical contributions exhibit the same response to volumetric strain as that of the

Solution energy of C atom at the O-site under volumetric strains. Results of BNNP, DP and DFT are listed, and units are all in eV/C-atom.

Volumetric strain	n(%)	$E_{\rm s}$			$E_{\rm s}^{\rm Mech}$		$E_{ m s}^{ m Chem}$			
	BNNP	DP	DFT	BNNP	DP	DFT	BNNP	DP	DFT	
-5.88	1.627	1.743	1.577	1.374	1.414	1.394	0.253	0.328	0.183	
-2.97	1.120	1.215	1.143	1.207	1.257	1.229	-0.087	-0.041	-0.086	
0.00	0.682	0.789	0.741	1.053	1.091	1.079	-0.371	-0.303	-0.338	
3.03	0.332	0.456	0.375	0.922	0.942	0.949	-0.590	-0.487	-0.575	
6.12	0.069	0.208	0.029	0.812	0.816	0.814	-0.743	-0.608	-0.785	

#### Table 3

Trapping energy and configuration of C atoms at monovacancy. The black, red, and blue balls indicate the Fe, C, and the monovacancy. The energy of the conf. of  $2 \cdot a$  was used to compute  $E_{\text{trap}}^{3C}$ . Units are all in eV/C-atom.

	1 1			
т	Conf.	BNNP	DP	DFT
1		-0.713	-0.790	-0.676
2- <i>a</i>		-1.050	-1.213	-1.064
2- <i>b</i>		-1.102	-1.156	-1.042
3		-0.385	-0.503	-0.300
4- <i>a</i>	<b>X</b>	0.232	0.218	0.440
4- <i>b</i>		1.566	1.824	2.039

total solution energy. These properties were also examined by DP. All results predicted by BNNP and DP are quantitatively in line with those obtained by DFT calculations (see Table 2).

The diffusion of a C atom in  $\alpha$ -iron takes place between neighboring O-sites, passing through a T-site. The diffusion energy barrier predicted by BNNP (resp.DP) is 0.88 (0.78) eV, which is in accordance with DFT results of 0.86 [61], 0.87 [62] eV. Note that above properties have been included in the training database.

## 3.3.2. Interaction of Carbon with monovacancy in $\alpha$ -iron

The configuration and trapping energy of various number of C atoms in a monovacancy at T = 0 K were tested using a  $4 \times 4 \times 4 \alpha$ -iron supercell with one vacancy (127 Fe atoms). The trapping energy for a C atom in a monovacancy, already trapping (m-1) C atoms from a site with lower trapping energy, can be computed by:

$$E_{\rm trap}^{mC} = (E_{mC} - E_{(m-1)C}) - (E_{\rm FeC}^{\rm O} - E_{\rm Fe}^{\rm BCC}),$$
(8)

where  $E_{mC}$  and  $E_{(m-1)C}$  stand for the energy of the monovacancy model (127 Fe atoms) with *m* and (m-1)C atoms in the vacancy, respectively, and  $E_{FeC}^{O}$  and  $E_{FeC}^{BCC}$  indicate the energy of a perfect  $4 \times 4 \times 4 \alpha$ -iron supercell with and without a C atom occupying an O-site, respectively. A negative value means energetically acceptance at T = 0 K. We conducted additional DFT calculations, and the trapping energies are presented in Table 3, alongside those obtained from MLIPs.

The BNNP and DP predict that a monovacancy can accommodate no more than three C atoms, which quantitatively agrees with DFT calculations. For the configuration point, two C atoms within a monovacancy do not show a preference for either a neighboring configuration or the other one (see Confs. of 2-*a* and 2-*b* in Table 3). Furthermore, when considering four C atoms within the vacancy, the planar configuration (Conf.4-*a*) is energetically favored compared to the tetrahedral configuration (Conf. 4-*b*). These configurations are all consistent with the results obtained from DFT calculations [63–65].

# 3.3.3. Interaction of Carbon with GBs in $\alpha$ -iron

We take  $\Sigma 5(310)[001]$  symmetric tilt GB (short for  $\Sigma 5$  GB) as example to illustrate the BNNP and DP performance. The  $\Sigma 5$  GB model and C solution sites are depicted in Fig. 3. Taking the iron and carbon in BCC and diamond structure, respectively, as the energy reference,



**Fig. 3.** Atomic structure of  $\Sigma$ 5(310)[001] symmetric tilt GB. Big balls, colored in dark yellow and dark gray, represent the Fe atoms located at GB and those with a bcc lattice, respectively. Small red balls stand for C atoms. *a*-*d* denotes the 4 potential C interstitial sites at the GB plane. *a*<sub>0</sub> is the lattice constant of *a*-iron. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the GB formation energy at T = 0 K computed by BNNP reduced from 1.558 J/m<sup>2</sup> for the C-free GB to 0.568 J/m<sup>2</sup> for the 4C-segregated-GB, representing an approximate 60% reduction. The GB was notably stabilized by the presence of C atoms, as evidenced by the rapid reduction in GB formation energy. The formation energy reduction is quantitatively agree with DFT results [58,59] (see Table 4). Following the method mentioned in Section 3.3.1, C solution energy was further split into two components, revealing the solution energy is primarily determined by the chemical contribution (see Table 4). This result is in line with that of available DFT calculations [59]. DP shows the same results as BNNP for the considered GB properties, See Table 4.

#### 3.3.4. Interaction of Carbon with screw dislocation in $\alpha$ -iron

C atom at or in proximity to a screw dislocation can trigger the reconstruction of the core configuration [66,68], presenting a challenge for empirical potentials in accurately capturing this phenomenon [5, 66,69,70]. As shown in Fig. 4(a), a dislocation dipole with easy core configuration was inserted into a 135-atom supercell, whose vectors are defined by the following expressions [6,71]:  $\vec{x} = 5\vec{x}_0$ ,  $\vec{y} = 2.5\vec{x}_0 + 4.5\vec{y}_0$ ,  $\vec{z} = \vec{z}_0$ , where  $\vec{x}_0 = a_0[\sqrt{6}, 0, 0]$  along  $[\bar{1}\bar{1}2]$ ,  $\vec{y}_0 = a_0[0, \sqrt{2}, 0]$  along [110], and  $\vec{z}_0 = a_0[0, 0, \sqrt{3}/2]$  along [111], respectively. We initially placed a carbon atom at an O-site near the screw dislocation core. Subsequently, structure relaxation was conducted, leading to the system undergoing spontaneous reorganization and transitioning towards a hard-core configuration (also called prism configure) with the carbon atom at the center, as illustrated in the subfigures in the upper panel of Fig. 4(b). The BNNP and DP predict that reconstruction can occur when the separation of C atoms is up to 4b in the direction along the dislocation line  $(l_{C-C} = 4b)$ , and the reconstructed hard core is slightly distorted for  $l_{C-C} = 5b$  and 6b, as presented in the lower panel of Fig. 4(b). DDmaps for C separations of  $l_{C-C} = 4b$ , 5b, and 6b are provided in the Supplementary materials (Fig. S5) [16].

The C-dislocation interaction energy per C atom, is defined as the energy difference between the situations where the C atom is in or infinitely separated from the dislocation core [66]. With this definition, negative energies indicate attraction. The carbon-dislocation interaction energy is -0.56 eV for a dipole dislocation model with a 1*b* separation between trapped C atoms in the screw dislocation core ( $l_{C-C} = 1b$ ). Upon increasing the separation between neighboring C

GB energy of C-free and C solution energy of C-decorated  $\Sigma5(310)[001]$  GB, reported DFT results are also listed. The model and C occupied sites are shown in Fig. 3.

	$E_{\rm GB}~({\rm J/m^2})$			$E_s$ (eV/C-atom)			$E_s^{\text{Chem}}$ (eV/C-atom)			$E_s^{\text{Mech}}$ (eV/C-atom)			
	BNNP	DP	DFT [58]	DFT [59]	BNNP	DP	DFT [59]	BNNP	DP	DFT [59]	BNNP	DP	DFT [59]
C-free	1.558	1.497	1.55	1.60	-	-	-	-	-	-	-	-	-
C@site(a)	1.232	1.200	1.25	1.28	-1.032	-0.942	-1.03	-1.081	-0.976	-1.08	0.049	0.034	0.05
C@site(ac)	0.879	0.893	0.90	0.95	-1.114	-0.971	-1.04	-1.148	-0.994	-	0.034	0.024	-
C@site(abc)	0.711	0.719	0.73	0.81	-0.528	-0.548	-0.45	-0.586	-0.584	-	0.058	0.036	-
C@site(abcd)	0.568	0.578	0.58	0.68	-0.453	-0.448	-0.39	-0.515	-0.480	-	0.062	0.032	-



**Fig. 4.** MLIPs performance for the interaction of C with screw dislocation. (a) Unit cell and periodicity vectors within quadrupole arrangement (135 atoms). (b) C induced core configuration reconstruction, (c) Interaction energy of C with screw dislocation. (d) Kink-pair atomic configurations of C separation of 1*b* and 2*b* (upper panel) and atomic configuration variation for kink-pair nucleation for  $l_{C-C} = 2b$  (lower panel). (e) Energy variation during the migration of the positive and negative kinks in cases of  $l_{C-C} = 1b$  and 2*b*. (f) Energy variation for kink-pair nucleation in the case of  $l_{C-C} = 2b$ . The corresponding DFT results in (c), (e) and (f) [66,67] are also presented. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

atoms, the interaction energy first decreases rapidly to -0.91 eV for  $l_{C-C} = 2b$ , and then increase gradually to -0.79 eV for  $l_{C-C} = 6b$ . The interaction energies predicted by BNNP, DP and calculated by DFT [66]

are depicted in Fig. 4(c). A notable agreement can be observed, suggesting that the BNNP and DP can accurately depict the interaction of C atoms with dislocation.

Carbon diffusion is of great importance for steels and for the actual C distribution in the alloy. While migration barriers for C can be as high as 0.9 eV in bcc lattice (see Section 3.3.1), it can be much lower at defects, such as the barrier could be 0.2 eV around dislocation core from an EAM estimation [72]. As well known, it is difficult to correctly capture the core reconstruction induced by C using EAM potential [5,72], thus, it is meaningful to revisit the solution energy of C around a reconstructed core induced by C and diffusion energy barriers for several interesting paths using high accuracy MLIPs. The dislocation model used in Section 3.2 with a dislocation length of 2b was adopted. A 3-fold low solution energy region around the core can be found, which meets the experimental observed Cottrell atmosphere around dislocation [73] (please also refer Section 4.2). Diffusion barriers, obtained by NEB method, could be low to ~0.3 eV, confirms the existence of the socalled "high-mobility zone" around the core of dislocation [72] in the model used. These results are presented in the supplementary materials (Fig. S6) [16]. It would be very valuable to study the C diffusivity in the Cottrell atmosphere based on the C distribution obtained in Section 4.2.

Recently, Ventelon et.al. [67] investigated the effect of C decoration on the mobility of screw dislocation in  $\alpha$ -iron using extensive DFT calculations. The atomic structures of C induced configuration reconstructed screw dislocation with a kink-pair was firstly reported for  $\alpha$ -iron [67]. We utilized a dislocation dipole model, as shown in Fig. 4(a), featuring a dislocation length of 8b with  $l_{C-C} = 1b$  and 2b, to assess the transferability of the MLIPs in the description of the C-decorated kinks. In contrast to the kinks with mirror symmetry observed in pure  $\alpha$ -iron (see Fig. S7 in the supplementary materials [16]), two nonequivalent kinks were formed on the screw dislocation for the two C separations, named as K<sup>+</sup> and K<sup>-</sup> respectively, as shown in the upper panel of Fig. 4(d). In the case of  $l_{C-C} = 1b$  (1b-K<sup>±</sup>), the atomic configuration of 1b-K<sup>-</sup> reveals two C atoms occupying distorted octahedral sites, slightly differing from the DFT calculations that only one C atom has an octahedral-like coordination [67], and other C atoms all occupy the center of reconstructed hard core configuration. The kinks configuration for  $l_{C-C} = 2b$  (2b-K<sup>±</sup>, see the upper panel of Fig. 4(d)) predicted by BNNP and DP are all C atoms at the center of the prism, which matches those obtained by DFT calculations [67].

We utilized a dipole model (Fig. 4(a)) with a dislocation length of 40*b* and kinks distance of 20*b* to determine the migration energies for the four types of kink (1b-K<sup> $\pm$ </sup>, 2b-K<sup> $\pm$ </sup>). The migration energy barrier of 1b-K<sup>+</sup> and 1b-K<sup>-</sup> kinks predicted by BNNP are 0.43 and 1.45 eV, and those of 2b-K<sup>+</sup> and 2b-K<sup>-</sup> are 0.95 and 1.10 eV, respectively. All four of these barriers exhibit good agreement with DFT results of 0.58, 1.52, 0.92, and 1.01 eV [67]. The similar results can be produced by DP, as plotted in Fig. 4(e). The migration of the kink involves the diffusion of C atoms near the kink.

The energy barrier for the nucleation of a kink-pair was also tested in the case of a C distance of 2b ( $l_{C-C} = 2b$ ) in a dipole model with a dislocation length of 4b and a kink separation of 2b. The atomic configurations and energy evaluation are presented in the lower panel of Fig. 4(d) and Fig. 4(f), respectively. The nucleation energy predicted by BNNP is 1.34 eV, in line with the DFT result of 1.19 eV. Additionally, the kink nucleation process also includes the diffusion of the C atom in the vicinity of the kink. Both atomic configuration of C-decorated kinks and energy obtained by DP are similar as those predicted by BNNP, as presented in Figs. 4(d)–(f).

#### 3.4. MLIPs Performance for cementite and cementite-ferrite interface

Considering that cementite (Fe<sub>3</sub>C) is the primary form of iron carbide in steel, it is essential to verify the performance of the MLIPs for the Fe<sub>3</sub>C- $\alpha$ -Fe system. There are 16 atoms in a Fe<sub>3</sub>C unit cell, including 4 C atoms and 12 Fe atoms. In accordance with Fe<sub>3</sub>C space group of *Pnma*, C atoms occupy the same Wykoff position of 4*c* while Fe atoms could occupy the Wyckoff positions of 4c or 8d. We named Fe atoms as Fe<sub>1</sub> and Fe<sub>2</sub> for their occupation of 4*c* or 8*d*, respectively (See the inner

panel of Fig. 5(b)). The lattice and elastic constants of Fe<sub>3</sub>C produced by MLIPs show good agreement with those of DFT calculations and reported experimental results [74–76], See data for  $\theta$ -Fe<sub>3</sub>C in Table 5. Using a Fe<sub>3</sub>C unit cell with the equilibrium lattice constant, the phonon dispersion curves along high symmetry path produced by BNNP and DP are shown in Fig. 5(a), which aligns with our DFT result and the DFT result reported in the literature [75].

The formation energy of various point defects in Fe<sub>3</sub>C at T = 0 K has been anticipated through the utilization of a 2 × 2 × 2 supercell. The formation energy of each defect was computed as follows:

$$E_{f} = E_{\text{Fe}_{m}\text{C}_{n}} - E_{\text{Fe}_{n}\text{C}_{q}} - (m-p)\mu_{\text{Fe}} - (n-q)\mu_{\text{C}},$$
(9)

where, the  $E_{\text{Fe}_{m}\text{C}_{n}}$  and  $E_{\text{Fe}_{p}\text{C}_{q}}$  indicate the energy of supercell with and without defects (p : q = 3 : 1).  $\mu_{\text{Fe}}$  is the chemical potential of Fe in BCC lattice. The introduction of a vacancy was accomplished by deleting a designated atom, three such kinds of defect were tested, namely Fe<sub>1</sub>-vac, Fe<sub>2</sub>-vac, and C-vac. The defect of antisite was realized by switching the type of the atom at the site, for instance, C–Fe<sub>1</sub>-anti stands for a Fe atom at a site of Fe<sub>1</sub> switched to C, two other cases of this type of defect were tested and they are C–Fe<sub>2</sub>-anti and Fe–C-anti. The results produced by BNNP, DP and reported DFT [77] are plotted in Fig. 5(b), a good agreement is distinctly evident.

To facilitate a direct comparison with DFT results, two (110) surfaces with different terminals that adhered to the stoichiometry of cementite were chosen and shown in Fig. 5(c). The surface energy produced by BNNP (resp. DP) is 2.251 (2.273) and 2.529 (2.506)  $J/m^2$  for Sur-I and Sur-II, respectively, agree with those obtained by DFT calculations of 2.214 and 2.513  $J/m^2$ . The interface of these two surfaces with ferrite was also examined, and the atomic structures are illustrated in Fig. 5(d). The interface energy of Int-I and Int-II produced by BNNP (resp. DP) are 0.608 (0.619) and 1.755 (1.763)  $J/m^2$ , again, well aligns with our DFT results of 0.571 and 1.609  $J/m^2$ , and reported DFT results of 0.449 and 1.765  $J/m^2$ [78]. The significant distortion observed in the ferrite region in Int-II is indicative of its high interface energy.

Regarding the low energy cementite-ferrite interface (Int-I), we conducted additional examinations to assess the energy response to the relative sliding of the two phases. The result predicted by BNNP is displayed in Fig. 5(e). We observed a low energy barrier of  $0.022 \text{ eV}/\text{\AA}^2$ when the two halves relatively glide along Fe[111]/Fe<sub>3</sub>C[010] direction, which is lower than that in pure  $\alpha$ -iron gliding along the same direction in the same iron orientation, measured a value of 0.074 eV/Å<sup>2</sup> (see Fig. S2 in the supplementary materials [16]). This suggests that the motion of the ferrite part along a trench situated between two rows of carbon atoms in cementite part is approximately three times more facile compared to its occurrence in pure iron. Additional DFT calculations as what we did using BNNP for the Int-I have been carried out and the point by point comparison with the results obtained by BNNP are illustrated in Fig. 5(f), providing quantitative confirmation of the conclusions derived from the BNNP predictions. Almost the same results can be obtained using the DP, as plotted in the point by point comparison figure (Fig. 5(f)).

### 3.5. MLIPs Performance for Carbides and the importance of the similarity

Beyond the examined cementite mentioned above, there are also other metastable iron carbide phases such as  $Fe_4C$ ,  $Fe_5C_2$ ,  $Fe_2C$ , FeC, and  $Fe_7C_3$  etc., with various space groups. Performance of two MLIPs in estimating the lattice constant and elastic constants of 14 carbides has been investigated, and the results are summarized in the Table 5, which is provided alongside the DFT results.

One can observe remarkable accuracy in the description of  $\omega$ -Fe<sub>3</sub>C, as even the small elastic constants ( $C_{15}$ ,  $C_{25}$ ,  $C_{35}$  and  $C_{46}$ ) are precisely captured. The MLIPs also exhibit the superb capability to accurately describe the carbide of o-Fe<sub>7</sub>C<sub>3</sub> (*Pbca*), not only under pressure free condition but also in systems subjected to an external pressure of 150



**Fig. 5.** MLIPs performance for cementite and ferrite-cementite interface. (a) phonon dispersion curves of cementite, (b) formation energy of point defects in cementite. (c) Two (110) surface configurations of cementite, (d) atomic configuration of two ferrite-cementite interfaces, (e) Energy responds to the relative gliding of the two phase in the interface of int-I in (d) obtained by BNNP. A comparison of the results obtained by DFT, DP, and BNNP is presented in (f). The subscript of f and c in (c)–(d) represent the coordinate system for ferrite and cementite, respectively. The big gray balls and small red balls in (b)–(d) stand for Fe and C atoms, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

GPa, although none data regarding these carbides are prepared. These 14 carbides can be classified into four types based on the local atomic environment associated with Fe–C bonding, which is summarized in Table 6. Plenty of atomic structures regarding the strained and unstrained cementite were included in the database, therefore, the BNNP and DP presented high transferability for the carbides in type-I. As addressed in Section 3.3.1, the BNNP and DP can correctly describe a single C atom occupying an O-site in iron with and without deformation, this can explain the good transferability of BNNP and DP for the carbides

in type-II. A few structures of carbides in type-III and type-IV were included in the database, as tested in Sections 3.3.1 and 3.3.2, and those structures are in strain-free state, leading to challenges in achieving good transferability of MLIPs for these carbides. Note that, for the unseen structures of FeC with ZnS, CsCl and NaCl lattice, BNNP exhibits better performance than that of DP in this work. Considering the BNNP and DP performance across these 14 carbides, it can be inferred that the structural similarity between the seen and unseen structures is a significant determinant of the transferability of MLIPs.

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Lattice constant and elastic constants of carbides obtained by BNNP, DP, and DFT calculations.

Carbides	Space Group	Method	La	ttice constant	(Å)	Elastic constants (GPa)								
			a	b	с	C <sub>11</sub>	C <sub>22</sub>	C <sub>33</sub>	C <sub>12</sub>	C <sub>13</sub>	C <sub>23</sub>	$C_{44}$	C <sub>55</sub>	C <sub>66</sub>
θ-Fe <sub>2</sub> C	Pnma	BNNP	5.020	6.737	4.469	365	341	310	172	153	177	14	122	115
5		DP	5.028	6.688	4.498	354	357	315	187	145	186	24	139	134
		DFT [75]	5.04	6.72	4.48	388	345	322	156	164	162	15	134	134
		EXP [76]	5.08	6.73	4.51	-	-	-	-	-	-	-	-	-
$\epsilon$ -Fe <sub>3</sub> C	P6322	BNNP	4.659	4.659	4.331	335	335	367	191	193	193	86	86	72
		DP	4.641	4.641	4.295	309	309	276	147	136	136	76	76	81
		DFT [79]	4.630	4.630	4.304	325	325	339	144	134	134	121	121	91
$\omega$ -Fe <sub>3</sub> C	P2/M	BNNP	4.263	4.267	2.756	343	370	336	197	142	127	40	11	94
		DP	4.242	4.201	2.785	231	290	229	138	64	83	71	-9	66
		DFT	4.227	4.221	2.739	317	364	347	161	126	119	37	-1	105
		DF1 [80]	4.227	4.227	2.738		-	-	_	-	-	-	-	-
						$C_{16}$	$C_{26}$	$C_{36}$	C <sub>45</sub>					
						6	-27	11	-22					
						-9	-28	-9	-62					
						-8	-32	8	-35					
n-Fe C	Pnnm	BNND	2.815	4 328	4 666	-	- 301	-	- 173	170	230	125	92	94
$\eta$ -1 $c_2c$	1 mm	DP	2.013	4 225	4 621	491	509	496	307	293	382	139	92	84
		DFT [81]	2.835	4.271	4.722	378	340	323	158	136	189	136	97	110
Fe <sub>2</sub> C	Pbcm	BNNP	4.455	4.943	4.881	293	575	382	256	241	235	106	29	140
2 -		DP	4.471	4.977	4.910	428	543	469	258	281	256	106	35	129
		DFT [82]	4.515	4.928	4.915	359	537	401	159	149	103	112	12	140
$\chi$ -Fe <sub>5</sub> C <sub>2</sub>	C2/2	BNNP	11.552	4.505	4.947	364	344	404	210	189	194	126	110	38
N 52	,	DP	11.550	4.542	4.975	417	399	431	232	214	226	137	114	48
		DFT [82]	11.570	4.573	5.061	349	341	410	288	151	164	139	132	35
						C <sub>15</sub>	C <sub>25</sub>	C <sub>35</sub>	$C_{46}$					
						-3	23	-2	-8					
						6	16	2	-6					
						-11	22	-1	-12					
o-Fe <sub>7</sub> C <sub>3</sub>	Pnma	BNNP	4.475	6.855	11.795	374	446	440	211	238	276	101	77	31
		DP	4.489	6.927	11.828	389	441	437	222	252	257	93	85	40
		DFT [79]	4.501	6.833	11.697	397	419	432	162	157	171	138	116	86
h-Fe <sub>7</sub> C <sub>3</sub>	$P6_3mc$	BNNP	6.852	6.852	4.421	450	450	377	252	223	223	83	83	98
		DP	6.886	6.886	4.444	395	395	364	220	209	209	82	82	87
		DFT [79]	6.808	6.808	4.470	458	458	370	175	180	180	126	126	141
o-Fe <sub>7</sub> C <sub>3</sub>	Pbca	BNNP	4.475	11.855	13.639	371	454	454	215	232	265	90	69	44
		DP	4.491	11.954	13.701	386	440	443	232	246	254	74	79	48
		DFT	4.495	11.811	13.547	378	417	425	181	190	190	124	95	75
		EXP [83]	4.5202	11.9747	13.7572	-	-	-	-	-	-	-	-	-
$o-Fe_7C_3$	Pbca	BNNP	3.945	10.676	12.242	1170	1171	1154	707	698	658	271	213	181
(150 GPa)		DP	4.042	10.998	12.041	1546	1042	15/4	1350	1355	1310	138	88	/2
		DFI DFT [94]	3.947	10.817	12.408	9/4	1072	1139	628	643 E00	020 E0E	204	169	128
		EXP [83]	- 3 9526	- 10.742	-	-	-	-	-	-	-	200	-	155
FeC-	Fm3m	BNNP	10 470	-	-	287	_	_	169	_	_	92	_	_
102306	1	DP	10.398	_	_	281	281	281	131	131	131	96	96	100
		DFT [79]	10.443	_	_	309	_	_	122	_	_	104	-	_
FeC	P43m	BNNP	4.276	_	-	388	-	-	385	-	-	2	-	-
	(ZnS)	DP	4.344	-	-	614	669	739	326	232	229	143	146	149
		DFT	4.257	-	-	354	_	_	210	-	_	67	_	_
FeC	P43m	BNNP	2.509	-	-	550	-	-	230	-	-	54	-	-
	(CsCl)	DP	2.486	-	-	1630	1620	1649	1040	1011	1020	75	71	78
		DFT	2.454	-	-	825	-	-	148	-	-	-30	-	-
FeC	$Fm\bar{3}m$	BNNP	4.052	-	-	771	-	-	320	-	-	100	-	-
	(NaCl)	DP	3.976	-	-	1172	-	-	937	-	-	233	-	-
	_	DFT	3.992	-	-	609	-	-	212	-	-	84	-	-
Fe <sub>4</sub> C	P43m	BNNP	3.849	-	-	122	-	-	192	-	-	-121	-	-
		DP	3.839	-	-	80	-	-	96	-	-	-245	-	-
		DFT [85]	3.89	-	-	146	-	-	189	-	-	80	-	-

# 3.6. Computational cost assessment of potentials of BNNP, DP, EAM and $\ensuremath{\textit{MEAM}}$

Fig. 6 illustrates the comparison of computation costs among the BNNP, DP, EAM [5], and MEAM [86] potentials, tested on Cray CS-Storm 500GT of the MASAMUNE-IMR super computer at Tohoku University [87]. Each node used in the study was equipped with two Intel Xeon Gold 6150 CPUs, for a total of 36 cores, and 10 NVIDIA Tesla V100 GPUs serving as accelerators.

For a BCC iron model with size of  $27 \times 27 \times 27$ , and a C atom was placed at an O-site in each  $3 \times 3 \times 3$  BCC unit. Totally, the model

contains 39,366 Fe atoms and 729 C atoms. It is 27.38  $\mu$ s/atom/step for the BNNP using 2CPUs (36 cores), while it is 12.71  $\mu$ s/atom/step for the DP using 1CPUs and 5GPUs in a node. The computational speed can be further enhanced if more GPUs are available, the speed is 6.77  $\mu$ s/atom/step for the DP using 2CPUs and 10GPUs in a node. It is worth noting that DP exhibits computational speeds only 1 order slower compared to MEAM potential and 2 orders slower compared to EAM potential.

Carbides	are	class	ified	acco	rding	to	local	aton	nic	environ	ment	associ	ated	with	Fe-0
bonding,	big	gray	and	small	red 1	balls	stand	1 for	the	e Fe and	l C at	oms, r	espec	tively	

Type	Bonding manner	Carbide	typical unit	Remark
I	Cementite	$\varepsilon$ -Fe <sub>3</sub> C, $\chi$ -Fe <sub>5</sub> C <sub>2</sub> , h-Fe <sub>7</sub> C <sub>3</sub> , Fe <sub>2</sub> C, o-Fe <sub>7</sub> C <sub>3</sub> ( <i>Pnma</i> ), a-Fe <sub>7</sub> C <sub>2</sub> ( <i>Pbca</i> )	X	
П	Octahedral bonding	ω-Fe <sub>3</sub> C, $η$ -Fe <sub>2</sub> C, NaCl-FeC, Fe <sub>23</sub> C <sub>6</sub>		
III	Tetrahedral bonding	ZnS-FeC, Fe <sub>4</sub> C	<b>*</b>	102306
IV	C replace a Fe atom in BCC-Fe unit	CsCl-FeC	I <u>N</u>	



Fig. 6. Computational cost assessment of potentials of BNNP, DP, EAM [5] and MEAM [86].

#### 4. Atomistic simulations on $\alpha$ -Fe–C systems

### 4.1. Carbon effects on the motion of screw dislocation

Ensuring the constructed MLIPs precisely capture the motion of screw dislocations and accurately depict the impact of carbon on the mobility of these dislocations at finite temperatures is vital for understanding the carbon-induced strengthening of  $\alpha$ -iron through MLIPs. To tackle this essential aspect, we carried out molecular dynamics shear deformation tests on  $\alpha$ -iron crystal having a screw dislocation, examining the behavior of the screw dislocations in environments both with and without carbon introduction.

Fig. 7(a) shows the dislocation model used in simulations, which is 83.47, 48.19, and 59.01 Å (= 24*b*) in the *x* = [121], *y* = [ $\overline{1}$ 01], and *z* = [1 $\overline{1}$ 1] directions, respectively. Periodic boundaries are taken along [121] and [1 $\overline{1}$ 1] directions. A 1/2<111> screw dislocation is introduced at the center of the simulation cell, utilizing the displacement field derived from the anisotropic elasticity theory of dislocation, and the model contained a total of 20,736 Fe atoms. A shift of *b*/2 is added in the *z*-direction to the periodic boundary conditions across the *x* surfaces to stabilize the single screw dislocation [88,89]. Two C concentrations of 0.5 at% and 1.0 at% were considered for the C-introduced case, and those C atoms were introduced through random deposition method.

To drive the screw dislocation gliding on the ( $\overline{1}01$ ) plane along the [121] direction, dynamical simulations were performed under constant strain rate realized by exerting a constant velocity in +z and -z directions on the top and bottom slabs with 7 Å thickness as shown in Fig. 7(a), respectively. Two reflection walls [91] were respectively

set below the top slab and above the bottom slab to keep the C atoms staying in the region. All simulations were conducted on constant temperature of 300 K by Nosé Hoover thermostat with a time step of 0.5 fs. The applied engineering shear strain rate is  $1 \times 10^8$  s<sup>-1</sup>. The results obtained using BNNP were presented in Figs. 7 and 8, and those obtained by DP are provided in the supplementary materials (Figs. S8–S9) [16].

In the C-free screw dislocation model, as shown in Fig. 7(b), the dislocation possesses an easy core configuration at the strain-free state, and with the increase in shear strain, a kink-pair is nucleated at a strain level of 0.999%, then, the kinks propagate along the dislocation line in the {110} plane until the entire dislocation reaches the neighboring Peierls valley at the strain of 1.012%. The migration of the dislocations follows the theory of the kink mechanism [92]. The stress increases almost linearly with the rise in shear strain, reaching a maximum of approximately 0.8 GPa at the strain of 1%. Subsequently, the shear stress fluctuates around 0.75 GPa (see the upper panel of Fig. 8(a)).

For the C-introduced models, the nucleation of the kink-pair occurs in a segment either with C atoms positioned in front of the dislocation (arrows of A, B in Fig. 7(c) and D in Fig. 7(d)) or devoid of C atoms (arrow C in Fig. 7(c)), then, the kink-pair expands until it meets C atoms which act as obstacles resulting in the pinning effects on the migration of kinks (see the dotted line encircled part in snapshots with the strain of 1.386%-1.526% in Fig. 7(c) and 0.356%-2.391% in Fig. 7(d)). The maximum of the stress is higher than that of the C-free model. It is about 1.1 GPa at the strain of 1.5% for the model with 0.5 at.% C, and it is 1.75 GPa at 2.4% for the 1.0 at.% C model, respectively (see the upper panel of Fig. 8(a)), which demonstrates the locking effects of C on dislocation motion. The carbon locking effect is remarkably strong, and the dislocation becomes unlocked only after the remaining segments have migrated a distance equivalent to about two Peierls valleys, which can be observed by the position of dislocation, marked by double-headed arrows in Fig. 7(b)-(d), during the simulation. As the results, a serration of stress is observed, which is more significant in the case of higher C concentration. The C atoms in a cylindrical region centered at the dislocation with radius of  $\sqrt{3}a_0 \approx 4.0$  Å and length of 24*b* was collected and the linear C concentration ( $\lambda_{\rm C}$ ) was computed following  $\lambda_{\rm C} = \frac{n_{\rm C}}{L_{\rm Dis}}$ , where  $n_{\rm C}$  and  $L_{\rm Dis}$  is the number of C atoms in the region and the length of the dislocation, respectively. The C concentration evolution over strain was plotted in the lower panel of Fig. 8(a), which is synchronized with the stress serration demonstrating the carbon locking effect.

The carbon locking effect can be further elucidated by examining the displacement of the dislocation along its gliding direction *x* against the shear strain, which is plotted in the upper panel of Fig. 8(b). The jump distance after unlocking is longer than the C-free case, and the incubation time required for the jump is roughly proportional to the jump distance, thus dislocation exhibits jerky motion [93]. The distance between neighboring Peierls valleys in {110} planes is  $\Delta = \frac{\sqrt{6}}{3}a_0 \approx 2.3$  Å. In the strain rate and temperature, the longest observed jump is 5 $\Delta$  in the model with 0.5 at.% C and 10 $\Delta$  in the model with 1.0 at.% C.

One can observe the core configuration transition from an easy core to a hard core resulting from the interaction between the dislocation and the C atoms nearby. Such transition is commonly observed at both strain-free (atoms encircled by rectangles with solid green line in Fig. 7(c)-0.00% and (d)-0.00%) and strained states ((c)-1.526% and (d)-1.842% in Fig. 7). Importantly, following the passage of the dislocation, the C atoms revert to an octahedral coordination with Fe atoms, and no cementite trace is found after passing dislocation. The evolution of local atomic configurations around a C atom, transitioning from octahedral to prism and back to octahedral, is shown in the magnified subfigures enclosed by solid red lines in Figs. 7(c) and (d). The strong interaction between the dislocation and C atoms also results in the occurrence of local cross slip, as shown in the snapshots with strains of 0.085% to 0.356% in Fig. 7(d). The local cross slip occurrences during the simulation can be directly recognized through the displacement of the



**Fig. 7.** Model used for simulations and the simulation results obtained using BNNP. (a) Screw dislocation model employed in simulations. The directions of x, y, and z please see the main text. Yellow and cyan balls indicates the Fe atoms in the slabs of top and bottom. Green line in z direction stands for dislocation which is analyzed by the DXA tool implemented in the OVITO code [90]. (b)–(d) Local atoms around the dislocation for the first dislocation migration for the models with C concentrations of 0.0 at.%, 0.5 at.%, and 1.0 at.%, respectively. Small red balls stand for C atoms, and big colored balls stand for Fe atoms in different atomic planes. Several specific atomic configurations are encircled and magnified in (c) and (d) (see the main text). (b)–(d) share the same coordinate system and their shear strain (%) are also printed. The solid double-headed arrow in (b)–(d) denotes the position of dislocation in the corresponding snapshot, while dotted double-headed arrow signifies the location where the dislocation has swept. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

screw dislocation in the *y* direction, as plotted in the lower panel of Fig. 8(b). A higher carbon concentration results in a more frequent occurrence of dislocation cross-slip. The entire structural evolution in these three simulations is depicted in movies provided in the Supplementary materials (Movies.S1–S3, those obtained by DP please see Movies. S4–S6) [16].

# 4.2. Core configuration of a screw dislocation with C decoration

To accurately delineate the locking effects of carbon atoms on screw dislocations, the core configuration of a screw dislocation in the presence of carbon atoms at finite temperatures is a particularly significant factor in this scenario, a phenomenon that remains unexplored. Rather than randomly assigning the location of C atoms in the model, as demonstrated in Section 4.1, it can also be determined based on the carbon chemical potential at a given temperature and concentration. Using the screw dislocation model employed in Section 4.1, and the grand canonical Monte Carlo (GCMC) combined with MD simulations (the MD/GCMC hybrid method), C atoms can be inserted in and extracted from the supercell, and then the Fe-C system may approach

an equilibrium state. The C-decorated core configuration of the screw dislocation and also the Cottrell atmosphere around were studied.

To estimate the C concentration in  $\alpha$ -iron, a series of MD/GCMC simulations on NVT (MD) and  $\mu$ VT (GCMC) ensembles at 900 K were conducted, and the relationship between the chemical potential ( $\mu_{\rm C}$ ) and C concentration in  $\alpha$ -iron ( $C_{\rm bulk}$ ) at the temperature based on BNNP was obtained and plotted in Fig. 9. In the simulation for screw dislocation, the  $\mu_{\rm C}$  was set to be -9.15 eV to match the low carbon steel with the  $C_{\rm bulk}$  of ~0.3 at.% at the temperature (900K). 25 GCMC trials were conducted in each 5 MD steps with a time step of 0.5 fs on NVT+ $\mu$ VT ensembles.



Fig. 8. Simulation results obtained using BNNP. (a) The shear stress  $(\tau_{y_2})$  responses to the shear strain of models with different C concentrations(upper panel) and the linear C concentration ( $\lambda_C$ ) nearby the screw dislocation(lower panel). (b) Displacement of the dislocation in x- and y-direction.



**Fig. 9.** The relationship between C chemical potential and concentration in  $\alpha$ -iron at the temperature of 900 K, obtained by BNNP. The results of DP please refer to the supplementary materials (Fig. S10) [16].

that obtained by a thermodynamical model parameterized on DFT calculations (0.1 at.%C) [94]. The extracted C-decorated dislocation core configuration is illustrated in the middle panel of Fig. 10(b), in which the positions of Fe atoms were averaged over  $2.0 \times 10^5$  MD steps, while the locations of C atoms were recorded every 1000 MD steps throughout the final  $2.0 \times 10^6$  MD steps. It is clear that the dislocation core is completely transferred to the hard core configuration, and the trapped C atoms stay inside the prism units. To quantitatively describe the probability of occurrence of C atoms in the reconstructed dislocation core, the residential density ( $\rho_r$ ) of C atoms within bins, each with the same cross-sectional area as the prism of the hard core configuration and a length of 0.5 Å along the dislocation line, was computed following  $\rho_{\rm r} = \frac{n_{\rm b}}{n}$ , where  $n_{\rm b}$  and  $n_{\rm s}$  represent the count of the occurrences of C atoms in each bin and the number of snapshots, respectively. The result is shown in the bottom panel of Fig. 10(b). C atoms can be trapped by any prism unit and they exhibit a preference for separating 2b along the dislocation line, which meets the energetic predication at 0 K (See Fig. 4(c)).

A snapshot of the model at a MD step of  $2.6 \times 10^6$  is displayed in the upper panel of Fig. 10(b). As guided by the dotted blue lines, one can observe a three-fold distribution pattern of the C Cottrell atmosphere around the screw dislocation. To confirm this anisotropic radial distribution, the residential density of C atoms in the model was computed following the same manner as that for C atoms in the dislocation core. The C residential density projected onto XY plane is displayed in Fig. 10(d). The C three-fold distribution matches the lobeshaped regions of the Cottrell atmosphere, spaced 120 degrees apart around screw dislocations, as observed in atom probe experiments on lath martensite steel (0.85 at%C) [73] and other theoretical predictions [95,96], also agree with the distribution of low solution energy of C around the dislocation core at 0 K (see Fig. S6 in the supplementary materials [16]).

The C concentration in neighborhood of dislocation core was further studied. A cylindrical region along the dislocation line and centered at the geometric center of the hard core with radius of 3.6 Å (about one more atomic layer out of the core) and length of 24b was considered, as indicated by a red circle in Fig. 10(b). The numbers of enclosed C and Fe atoms were  $nC_{nb}$  and  $nFe_{nb}$  (= 288 for the model used in this work), which were collected through out the simulation. The C concentration  $(C_{\rm nb} = \frac{nC_{\rm nb}}{nFe_{\rm nb}})$  versus the MD steps is presented in Fig. 10(c). During the final  $7.5 \times 10^6$  GCMC samplings,  $C_{\rm nb}$  reaches  $6.0 \pm 0.6$  at.%, close to the theoretical prediction of 6–7 at.% [95], higher than another theoretical prediction based on EAM potential of  $10 \pm 1$  at.% [96], and lower than the experimental finding of  $8 \pm 2$  at.% obtained by energy-compensated position-sensitive atom probe analysis. We attribute this difference to the high temperature and low background C concentration (0.3 at.% in simulation and 0.85 at.% in experiment). We also suspect the sampling times might be not enough, as shown in Fig. 10(a) and (c), although the C equilibrium state has been reached for the dislocation core and the whole model, it has not yet been reached for the region surrounding the core. A simulation with large model is required for the estimation of the size of the Cottrell atmosphere around dislocation (extend to  $70 \pm 10$  Å from the dislocation reported in experiment [73]). The same simulation based on DP was also conducted and similar results are presented in the supplemental materials (Figs. S11-S12) [16].

The 900 K chosen in simulation is rather high, approaching the Curie temperature for Fe and hence in the BCC paramagnetic region, and probably far from interest for applications. The computational cost is high for the MD/GCMC hybrid method study of Fe-C system employing MLIPs at the room temperature which is currently unaffordable.



**Fig. 10.** (a) C concentration in the model( $C_{model}$ ) and C linear concentration in the core of the dislocation  $\lambda_{C_{core}}$  versus the MD steps throughout the simulation. (b) A snapshot of the model at the MD step of  $2.6 \times 10^6$  (upper), the C-decorated core configuration (middle) by showing the averaged Fe atoms position and C atoms history position with an interval of 1000 MD steps over the final 2.0 million MD steps in each prism unit. The residential density ( $\rho_i$ ) of C atom along the dislocation line is displayed in the bottom panel. Dark gray and red balls stand for Fe and C atoms, respectively. C atoms inside the core of screw dislocation are defined as  $C_{core}$ . (c) Concentration of C atoms at the dislocation nearby ( $C_{mb}$ ). (d) The residential density of C in model projected onto XY plane, a 3-fold C distribution can be observed. The results obtained by DP are presented in the supplemental materials (Figs. S11–S12) [16]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

# 4.3. Crack propagation along a grain boundary with Cementite in $\alpha$ -iron containing various types of defects

Experimental observations reveal that cementite precipitation at martensite grain boundaries exhibits a disk-shaped morphology, primarily extending along the martensite grain boundary plane [97]. To highlight the robustness of the MLIPs, a tensile test has been conducted on a model with notch at both sides along  $\Sigma 5(310)[001]$  symmetric tilt GB, and with cementite in  $\alpha$ -iron crystal containing multiple types of defects, as illustrated in Fig. 11(a). The model dimensions in the x, y, and z directions are 19.84, 197.15 and 108.54 Å, respectively. The periodic boundary condition was employed in the x and z directions. The surfaces were created by enlarging the size of the model in the ydirection, and the cracks were introduced by removing 9 atom planes along the GB. The initial crack length was set to ~32 Å. Fe atoms at the center of the model were replaced by cementite (Fe<sub>3</sub>C) to build the arbitrary Fe/Fe<sub>3</sub>C interface. Point defects of vacancies and interstitial were generated by randomly deleting and inserting atoms during a MD NVT simulation at T = 300 K. The densities of Fe vacancy and interstitial Fe atom were both set to 0.05 at.%, and the density of interstitial C atom was set to 0.25 at.%. The model for tensile test, which comprises a total of 36,355 atoms, is depicted in Fig. 11(b).

Temperature and time step were set to 300 K and 0.5 fs, respectively. The model was firstly thermalized at 300 K for 10 ps on the NPT ensemble with a null pressure condition in the *x* and *z* directions and with fixed length condition in the *y* direction. The uniaxial tensile test was conducted in *z* direction with a constant engineering strain rate of  $1.0 \times 10^9$  s<sup>-1</sup> by controlling model dimension in the *z* direction. Note that the zero pressure condition in the *x* direction was maintained while that in the *z* direction was canceled during the tensile test. The typical results of the tensile test using BNNP are shown in Fig. 12.

The tensile test even for such complicated model with defects of crack, GB, interstitial C and Fe atoms, and carbide, was successfully concluded, snapshots during the simulation were displayed in Fig. 12(a). We observed that the model showed elastic behavior in the low strain state (< 7.5%). With the progressive increase in strain, crack started to open (7.5%–10.0%), and propagating along the GB<sub>2</sub> until reaching the cementite core (17.5%–20.0%). Despite clear crack opening can be seen at the strain of 10%, the cementite seems to be stable and no void and cracking around the cementite were seen even a little shape change of the cementite was found.

In Fig. 12(a), we also illustrated the diffusion of a Fe-IA (Interstitial Atom), represented by an enlarged subfigure adjacent to each snapshot. In Fig. 12(b), the dislocation emission and motion were depicted. At



Fig. 11. (a) The model with multiple defects employed in the tensile test using BNNP. The subscripts u and l adopted in the axes represent the upper grain and lower grain, respectively, while c denotes cementite. The large dark gray and small red balls indicate Fe and C atoms. (b) The Fe atoms with bcc lattice in model are set to be invisible using OVITO package [90]. Consequently, the defects in the model are presented and categorized. Vac, IA, IA-Cluster indicate the defects of vacancy, interstitial atom, and cluster of IA, respectively. CT1 and CT2 indicate the crack tips on the right and left sides, respectively, and y- atom for their propagation directions.

the strain of 9.76%, a dislocation emitted from the crack tip in GB<sub>1</sub>, with the Burgers vector of  $\frac{1}{2}\langle 111\rangle$ , moved on a  $\{110\}$  plane in the  $\langle 111\rangle$  direction, and eventually impinge on the GB<sub>2</sub> in the model. At the strain of 11.05%, a dislocation emitted from the distorted GB<sub>2</sub>, and also conducted a motion on a  $\{110\}$  plane in  $\langle 111\rangle$  direction, and finally impinge on the surface in the model. The dislocation motion direction projected to the  $\{001\}$  plane is the  $\langle 110\rangle$  direction.

Traction–separation (T–S) curve is associated with the peak stress required to overcome the cohesive strength and the maximum interaction range for traction and is often used to quantitatively describe brittle fracture [99]. With the help of BNNP, DP, and DFT, the separation-stress curves of C-free and C-segregated  $\Sigma$ 5(310)[001] symmetric tilt GBs were calculated by taking the derivative of the separation-energy curve, which is obtained by rigidly separating a GB model following the designed paths. The results indicated that MLIPs can properly describe this kind of simulations. The detailed information can be found in the supplementary materials (see Fig. S13) [16].

To demonstrate the structure–property relationships, we performed stress intensity factor analysis for the two crack tips in the simulation from DP. For a two-dimensional model, such as the model used in this simulation, the model-I stress intensity factor of  $(K_I)$  can be obtained based on the Irwin's method [100,101]:

$$K_{\rm I} = \lim_{r \to 0} \sigma \sqrt{2\pi r},\tag{10}$$

where *r* is the distance measured from the crack tip in the crack propagation direction, and  $\sigma$  is the opening stress normal to the crack surface which can be determined by the atomic stress [102,103] in tensile test (r = y,  $\sigma = \sigma_{zz}$  in this simulation). The average values of *y* and  $\sigma_{zz}$  were used to fit  $K_1$ , considering the GB and carbon atom influence, as shown in the upper panel of Fig. 13, the simulation

results are also included in this figure. For detailed information on the averaging method, please refer to our previous work [44].

The values of the stress intensity factor  $K_{\rm I}$  for both crack tips, CT1 and CT2, increase with rising tensile strain. The observed difference in  $K_{\rm I}$  between CT1 and CT2 is attributed to variations in the crack tip shapes at the atomic scale. Another potential factor might be the small size of the model, which results in a scattered distribution of data points used for  $K_{\rm I}$  fitting. The  $K_{\rm IC}$  is the value of  $K_{\rm I}$  before the first event occurring. It is 0.784 MPa m<sup>1/2</sup> for CT1 where Fe bonds break occurring at the strain of 0.059. The CT1 propagated along the  $\langle 110 \rangle$  direction in a {110} plane from the strain of 0.085, and  $K_{\rm I}$  was decreased at strain of 0.093 due to crack propagation induced stress release. The bonding breaking of CT2 took place at the strain of 0.069 with  $K_{\rm IC}$ of 0.721 MPa  $m^{1/2}$ . After that, CT2 propagated along the original GB plane, the  $K_{\rm I}$  was increased and maintained at a high value of 0.85 MPa  $m^{1/2}$  due to the presence of carbon atoms near the crack front. The mechanism of this kind of crack asymmetrical propagation could be found elsewhere [104,105]. Local atomic configurations before and after events occurring are shown in Fig. 13. Conducting fracture simulation and analysis using linear elastic fracture mechanics (LEFM) [106] could offer a more direct and comprehensive understanding of carbon's effects on the toughness of various systems, such as crack propagation along planes [107] or along GBs [108]. This may be an avenue for future research

The simulation encompasses a range of phenomena, such as crack propagation, diffusion of point defects, dislocation nucleation from crack tips and at the cementite– $\alpha$ -iron interface, interactions between dislocations and point defects, grain gliding, GB migration, dislocation impingement on cementite and GB, as well as interactions between the crack tip and cementite (see Fig. 12, and Movie. S7 in the supplementary materials [16]). The capability of the MLIPs is effectively



**Fig. 12.** Simulation results obtained using BNNP. (a) Snapshots during the tensile test using BNNP. A case of Fe-SIA diffusion is depicted. The black dots in each subfigure are Fe atoms and played as the position reference. (b) Dislocation emission from crack tip (upper panel) and GB (lower panel) during the tensile test. The atoms are colored according to the Voronoi volume ( $\mathring{A}^3$ /atom) obtained using the Voronoi analysis implemented in the OVITO package [98]. Big and small balls stand for Fe and C atoms, respectively. The engineering tensile strain ( $\gamma$ ) of each snapshot is also printed. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

demonstrated through the simulation of such a complex system. These interatomic potentials can serve as reliable tools for understanding of diverse topics related to the C and  $\alpha$ -Fe binary system.

# 5. Discussion

# 5.1. Limitations of MLIPs and improvement

Although the reported MLIPs show high accuracy and good transferability in several important scenarios of Fe–C system, limitations are also accompanying. It is challenging to describe configurations with unseen LAEs. Encountering extrapolation in simulations using MLIPs is dangerous, the simulation trajectory might deviate to somewhere, and results in unphysical phenomena. This is a general challenge for all types of MILPs, which can be fixed by accordingly enriching LEAs of database. In certain cases, combining the Ziegler–Biersack–Littmark screened nuclear repulsion term [109] is helpful, as it can push atoms that are too close to separate, helping the system return to the correct trajectory.

In comparison to the efficiencies of BNNP and DP for the Fe–H binary system [31,44], DP for Fe–C does not exhibit a 40-fold increase in speed than BNNP. We could explain this issue from two aspects: first, the current BNNP model employed a short cutoff radius of the angular ACSFs for Fe, which significantly increased its computational efficiency [Fe-(FeC)<sub>BNNP</sub>: 6.0 Å v.s. Fe-(FeH)<sub>BNNP</sub>: 6.5 Å]. Second, the size of the neural network for three-body embedding (*se\_e3*) in the current DP model is relatively large [Fe-(FeC)<sub>DP</sub>: (8, 16, 32) v.s. Fe-(FeH)<sub>DP</sub>: (10, 20)], resulting in a substantial increase in computational



**Fig. 13.** Stress field near crack tips and the intensity factor response to the crack propagation from the simulation using DP. The atoms are colored according the atomic stress (in GPa) in tensile direction.  $\gamma$  is the engineering tensile strain. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

cost of DP. As the result, the reported DP seems not efficient enough. The motivation behind training the DP model lies in the scenario of atomic stress calculation, which is challenging for the BNNP currently. With the assistance of the shared database, we anticipate that utilizing a smaller embedding neural network will improve the efficiency of the DP model while maintaining accuracy.

The main task of the training of MLIPs is to find the relationship between the atomic structure and its energy. The database of Fe–C binary system we shared could be used to train new MLIPs under various machine learning frameworks, such as GAP, SNAP, Atomic cluster expansion (ACE) [110], Moment tensor potentials (MTP) [111], etc. Actually, the database of Fe–H binary system we shared in our previous work [31] has been adopted by other scientists to train new models [15]. Thus, based on this shared database, it is possible to obtain an MLIP that is more efficient and accurate than the reported MLIPs, especially for the scenarios considering fewer properties.

#### 5.2. Influence of high strain rate on conclusion obtained from simulations

The strain rates of  $10^8 \sim 10^9 \text{ s}^{-1}$  used in simulations are significantly high comparing to those in experiment of  $10^{-3} \text{ s}^{-1}$  [112] or even lower [113]. This disparity primarily arises from the inherent differences between the timescales in simulations and in laboratory experiments. The high strain rate is anticipated to influence the outcomes derived from the simulations. The simulation in Sections 4.1 and 4.3 did not fully capture carbon diffusion due to the limitations of the MD timescale and low diffusivity of C at room temperature (approximately 15 nm over 24 h at 293 K [73]). According to Orowan's equation, the average dislocation velocity ( $v_{\rm dis}$ ) is proportional to the mobile dislocation density ( $\rho_{\rm dis}$ ) and the applied deformation rate ( $\dot{\gamma}$ ), assuming a constant dislocation density over the observation timescale. The relationship is expressed as follows:

$$v_{\rm dis} = \frac{\gamma}{\rho_{\rm dis}b},\tag{11}$$

where b is the magnitude of the Burger's vector. If the strain rate were sufficiently low, the gliding speed of dislocations would decrease.

Consequently, more carbon atoms could catch up with and be trapped by the moving dislocations, resulting in stronger strengthening effects than those observed in this simulation. Therefore, while the current simulation settings may underestimate the strengthening effect, it does not affect the overall conclusions drawn from the simulation.

#### 5.3. Dislocation core reconstruction

Combining Sections 4.1 and 4.2, the C and screw dislocation interaction could be described as follows: the solid solution C atoms and the carbon-free dislocations are in mutual chasing. The motion speed of dislocations is likely faster than that of C atoms considering their respective energy barriers (kink pair nucleation energy: ~0.7 eV, carbon diffusion in a bcc lattice: ~0.9 eV). Upon entering their interaction region, the dislocation and C atoms attract and combine, leading to the reconstruction of the dislocation core. This event was observed at the onset of the MD+GCMC simulation in Section 4.2 (not presented). As a result, the dislocation is anchored by C atom at the location of their encounter. Subsequently, more C atoms will be captured by dislocation and the Cottrell atmosphere was formed around the dislocation, and dislocations become immobile. The C-anchored dislocation could adjust its configuration to a low energy state of straight shape by the diffusion of C atoms. The straight screw dislocation is considered as the signature of C and is frequently observed in experiments [114].

Under shear strain driving, kink pair nucleation starts from a site in the dislocation where nearby carbon atoms are present in its gliding direction. In other words, if carbon atoms diffuse to a site near the carbon-decorated dislocation, this might also trigger kink pair nucleation. The presence of the Cottrell atmosphere around the dislocation significantly increases this likelihood. New simulations were conducted for a dislocation model with Cottrell atmosphere to demonstrate the atomic evolution during the dislocation unpinning process. The final snapshot of the simulation in Section 4.2 was used as the initial state [115]. The unpinning stress was 3.04 GPa (3.74%) at 300 K, much higher than that obtained in Section 4.1 due to more C atoms in the core of the dislocation resulting in a stronger pinning effect. Unfortunately, the C diffusion was not observed during the dislocation unpinning, even at the elevated simulation temperature of 673 K (the corresponding unpinning stress is 2.17 GPa at 3.31%). This result could be attributed to the high shear strain rate and the short physical time of the simulation. Despite this, the gliding of the carbon-decorated dislocation still presents the Peierls mechanism as predicted by experiments [113]. The simulation vividly presents the evolution of the atomic structure during dislocation unpinning from the Cottrell atmosphere.

In contrast to the carbon-free dislocation, the energy barrier for kink migration is no longer negligible, because kink can be impeded by pre-existing carbon atoms (see Fig. 7(d)). Simulations in this work support the strengthening mechanism proposed by Ventelon et al. [67] that the glide of the dislocation is governed by the formation and migration of kinks, both related to the diffusion of C atoms. As the carbon-anchored segment is always the final part of the dislocation to glide away under shear driving, highlighting the primary contribution of the reconstructed core to the strengthening effects [67]. Lower unpinning stress could be expected if C diffusion occurs, accelerated MD methods [116] might be helpful in this scenario, which is out of the scope of this work.

# 6. Conclusion

In this work, we constructed machine learning interatomic potentials (MLIPs), including Behler–Parrinello neural network potential (BNNP) and deep potential (DP), for the  $\alpha$ -Fe–C binary system based on a comprehensive database obtained by extensive spin-polarized DFT calculations. In the validation part, we systematically tested the MLIPs performance for the systems of pure  $\alpha$ -iron, interaction of C atoms with perfect and defective  $\alpha$ -iron, properties of cementite and ferrite– cementite interfaces, and various carbides. The test results provide us with evidence of the high accuracy and good transferability of our MLIPs, such as the prediction of the screw dislocation core configuration reconstruction induced by C atoms, which is difficult for EIPs.

The impact of C atoms on the mobility of screw dislocation at 300K was investigated through MD simulations in conjunction with MLIPs. The reconstruction of the hard-core configuration introduced by the adjacent C atoms was presented in both the strained and unstrained models. The reconstructed hard core unit cannot be maintained and the C atom will revert to the octahedral bonding manner after the dislocation has passed. The critical stress of the screw dislocation gliding in the C-introduced model is higher than that in the C-free model, revealing the C locking effect on the mobility of screw dislocation. The reconstructed hard-core shows primary contribution to the locking effect. The dislocation motion behavior is changed from the steady motion in the C-free model to the jerky motion in the C-introduced models.

With the help of the MLIPs, the C-decorated core configuration of screw dislocation was studied using MD/GCMC hybrid method at 900 K. With the C decoration, the dislocation core undergoes a transformation into a hard-core configuration. C atoms are located inside the core and prefer a separation of 2b along the dislocation. The distribution of C atoms around the dislocation exhibits a three-fold pattern, aligning with the experimental observations of the Cottrell atmosphere. The nucleation of kinks can occur spontaneously at the temperature, influenced by the location of C atoms in the vicinity of the dislocation.

Finally, we showcase the capabilities of the MLIPs by conducting a tensile test on a model containing multiple types of defects, including vacancies, interstitial atoms, surface, edges, GB, cementite, ferrite/cementite interfaces, cracks, and dislocations. During the tensile test, the crack tips showed a asymmetry propagation behavior, accompanying phenomena of diffusion of point defects, dislocation nucleation from crack tips and at the ferrite/cementite interface, interactions between dislocations and point defects, grain gliding, GB migration, dislocation impingement on cementite and GB, as well as interactions between the crack tip and cementite.

The BNNP and DP developed for the  $\alpha$ -Fe–C binary system possess individual advantages. Specifically, the BNNP demonstrates better accuracy than the DP, while the DP provides an advantage over BNNP in scenarios requiring the computation of atomic stress. Our validation and demonstration suggest that these potentials can be utilized in simulations for the C and  $\alpha$ -Fe systems involving various types of defects. These MLIPs can also generate the parameters required for multiscale simulations, such as kinetic Monte Carlo technic [72,117], or for analytical models [67]. We hope that the new MLIPs will soon provide fundamental insights into the atomic-scale deformation and fracture mechanisms of carbon steel.

# CRediT authorship contribution statement

Fan-Shun Meng: Writing – original draft, Methodology, Investigation, Conceptualization. Shuhei Shinzato: Methodology, Validation. Shihao Zhang: Investigation. Kazuki Matsubara: Investigation. Jun-Ping Du: Methodology, Investigation. Peijun Yu: Validation, Investigation. Wen-Tong Geng: Writing – original draft, Conceptualization. Shigenobu Ogata: Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

# Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

# Data availability

The database used for training the machine learning interatomic potentials and the two MLIPs are available online: https://github.com/mengfsou/MLPs-FeC.

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# Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.actamat.2024.120408.

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