Effect of Alloying Elements on the Stacking Fault Energy and Ductility in Mg$_2$Si Intermetallic Compounds

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ABSTRACT: Alloying elements can pronouncedly change the mechanical properties of intermetallic compounds. However, the effect mechanism of this in Mg$_2$Si alloys is not clear yet. In this paper, systematic first-principles calculations were performed to investigate the effect of alloying elements on the ductility of Mg–Si alloys. It was found that some alloying elements such as In, Cu, Pd, etc. could improve the ductility of Mg$_2$Si alloys. Moreover, the interatomic bonding mechanisms were analyzed through the electron localization functional. Simultaneously, the machine-learning method was employed to help identify the most important features associated with the toughening mechanisms. It shows that the ground state atomic volume ($V_{GS}$) is strongly related to the stacking fault energy ($\gamma_{sf}$) of Mg$_2$Si alloys. Interestingly, the alloying elements with appropriate $V_{GS}$ and higher Allred–Rochow electronegativity (En) would reduce the $\gamma_{sf}$ in the Mg–Si–X system and yield a better ductility. This work demonstrates how a fundamental theoretical understanding at the atomic and electronic levels can rationalize the mechanical properties of Mg$_2$Si alloys at a macroscopic scale.

1. INTRODUCTION

Magnesium (Mg) alloys are one of the development directions of structural and functional materials in the present and will be in the future due to their good damping capacity, low density, high strength-to-weight ratio, and biodegradability. The intermetallic compound of Mg$_2$Si is a promising thermoelectric material because of its high thermoelectric performance. In addition, Mg$_2$Si also has excellent hydrogenation properties, making it one of the candidates for hydrogen storage materials. Due to a narrow band gap of about 0.6 eV, Mg$_2$Si can be used as an infrared detector operating in the 1.2–2.0 μm wavelength range. Meanwhile, nontoxicity, low production cost, and huge thermoelectric conversion efficiency of Mg$_2$Si are required for its successful commercialization. These features make Mg$_2$Si suitable for a variety of applications, including the automotive and aerospace industries. Unfortunately, Mg$_2$Si alloys show very low ductility and strength.

Therefore, designing novel Mg–Si alloys with improved ductility has become critically important. Mg$_2$Si has a face-centered cubic (FCC) CaF$_2$-type crystal structure, which possesses sufficient possible dislocation slip systems. In the previous study, we demonstrated that the strong covalent bond characteristic of the Si atom is the primary reason for its brittleness. There are two main approaches to improve ductility. An effective approach is to refine the grain size, which can enhance the formability of alloys. However, the expensive refining process increases the material cost. Another approach is to introduce alloying elements. The addition of certain alloying elements to a metal is one of the most effective ways of altering the local structure and chemistry of alloys, and it can also be used to improve the ductility and fracture toughness of the alloy. For example, Qu and co-workers improved the room-temperature ductility of TiAl by adding the La element. Alloying can also change the anisotropy of electron distribution around Al atoms that could influence the slip behavior in pure aluminum. For Mg$_2$Si alloys, at present, Al addition plays a critical role in increasing the ductility of Mg$_2$Si. Another research indicates that doping In, Li, Sn, and Bi can efficiently improve the ductility of Mg alloys. However, the understanding of the effect of alloying elements on Mg$_2$Si alloys is still far from sufficient and a great deal of effort is required to find ideal alloying elements.

Actually, it is difficult to directly measure the effect of alloying elements on important features such as the resultant changes in the chemical environment at a boundary. As a result, the calculation approaches based on density functional theory (DFT) have been used to predict the mechanical
properties of Mg alloys. A design map of Mg alloys based on the intrinsic ductility was established by DFT. The stacking fault energy (SFE) is a critical intrinsic material parameter that significantly affects plastic deformation behavior and mechanical properties. Wu et al. found that the dominant factor in the reduction of SFE is the charge redistribution surrounding the alloying elements. Tsuru et al. showed distinctive hybridization between the p band of Mg and the d band of the alloying element, which characterizes the strong fracture toughness of Mg-based binary alloys. Therefore, comprehensive DFT calculations can certainly help us understand the important features and the mechanism in Mg2Si alloys.

In the present work, we introduced the SFE to understand the effect of alloying elements on the ductility of Mg−Si alloys. We chose the {111}(110) habit slip system because it is the habit slip system for FCC metals. Then, extensive DFT calculations were performed to reveal the general trends of the energetic properties of a large number of alloying elements across the periodic table doping to the slip plane in the FCC Mg2Si matrix. For possible alloying elements, we considered 33 elements including five elements from group I (i.e., Li, Na, K, Rb, and Cs), four elements from group II (i.e., Be, Ca, Sr, and Ba), four elements from groups III and IV (i.e., Al, Ga, In and Sn), and all 3d and 4d transition metals (TMs). Following that, we also unraveled the intrinsic microscopic electronic mechanism of Mg2Si alloys doped with alloying elements.

Finally, the effects of possible factors on the variations of SFE in Mg2Si were investigated by machine learning (ML) due to its advantage in effectively finding features related to performance. The results of this study can provide a theoretical reference for alloying design of multicomponent Mg−Si alloys.

2. RESULTS AND DISCUSSION
2.1. Solution Energy. The solution energy ($E_{\text{sol}}$) can be described by

$$E_{\text{sol}} = E_{\text{tot}}^{\text{Mg-Si-X}} - \sum_i n_i E_{\text{bulk}}^i$$

(1)

where $E_{\text{tot}}^{\text{Mg-Si-X}}$ is the total energy of the alloying-element-doped Mg2Si supercells, $n_i$ is the atom number of each element in the supercell, and $E_{\text{bulk}}^i$ denotes the energy per atom of each element in the stable ground state. The alloying element can be alloyed into the Mg2Si system spontaneously if the solution energy $E_{\text{sol}}$ is negative. The more negative the $E_{\text{sol}}$ is, the more likely it will appear.

The calculated equilibrium lattice parameter of undoped FCC Mg2Si was 0.636 nm. This result is in good agreement with the experimental value of 0.635 nm. The solution energies of alloying elements in Mg2Si were calculated by considering the magnetic moment of these alloying elements during the electronic self-consistent calculation. Since the
lattice distortion will cause the system energy change when the alloy element atomic size difference is large, we consider the influence of the ground state atomic volume ($V_{GS}$) on the $E_{sol}$. As shown in Figure 1 and Table 1, the solution energies have a strong correlation with the $V_{GS}$. The alloying elements of groups I, II, III, and IV have the same trend that as $V_{GS}$ increases, the solution energy increases. On the contrary, for all 3d- and 4d-TM alloying elements, it is observed that the

| alloying elements | $E_{sol}$ (with M) (eV) | $E_{sol}$ (without M) (eV) | $\gamma_u$ (J/m²) | $\gamma_s$ (J/m²) | $D$ | $V_{GS}$ (Å³) | En |
|-------------------|------------------------|---------------------------|-----------------|-----------------|-----|---------------|----|
| Li                | 0.078                  | 0.078                     | 0.997           | 1.297           | 0.390 | 16.59         | 0.97|
| Be                | 1.499                  | 1.500                     | 0.942           | 1.425           | 0.454 | 7.89          | 1.47|
| Na                | 0.742                  | 0.742                     | 0.924           | 1.215           | 0.395 | 29.24         | 1.01|
| Mg                | 0.000                  | 0.000                     | 0.925           | 1.336           | 0.433 | 22.89         | 1.23|
| Al                | 0.729                  | 0.729                     | 0.998           | 1.415           | 0.425 | 16.48         | 1.47|
| K                 | 2.089                  | 2.089                     | 0.595           | 1.015           | 0.511 | 73.11         | 0.91|
| Ca                | –0.215                 | –0.215                    | 0.872           | 1.225           | 0.422 | 37.77         | 1.04|
| Sc                | –0.519                 | –0.519                    | 1.084           | 1.502           | 0.416 | 22.24         | 1.20|
| Ti                | 0.430                  | 0.474                     | 1.118           | 1.531           | 0.411 | 16.69         | 1.32|
| V                 | 1.665                  | 1.665                     | 1.121           | 1.637           | 0.438 | 13.01         | 1.45|
| Cr                | 1.253                  | 2.518                     | 1.042           | 1.436           | 0.413 | 11.19         | 1.56|
| Mn                | 2.452                  | 2.451                     | 1.021           | 1.616           | 0.475 | 10.49         | 1.60|
| Fe                | 1.403                  | 2.285                     | 0.968           | 1.463           | 0.454 | 10.73         | 1.64|
| Co                | 1.608                  | 1.951                     | 0.916           | 1.470           | 0.481 | 10.25         | 1.70|
| Ni                | 1.089                  | 1.089                     | 0.882           | 1.479           | 0.503 | 10.32         | 1.75|
| Cu                | 0.590                  | 0.590                     | 0.890           | 1.371           | 0.462 | 11.07         | 1.75|
| Zn                | 0.562                  | 0.562                     | 0.951           | 1.337           | 0.422 | 13.96         | 1.66|
| Ga                | 0.844                  | 0.844                     | 0.938           | 1.327           | 0.424 | 18.86         | 1.82|
| Rh                | 2.966                  | 2.966                     | 0.398           | 0.884           | 0.667 | 90.72         | 0.89|
| Sr                | 0.540                  | 0.540                     | 0.659           | 1.095           | 0.499 | 54.23         | 0.99|
| Y                 | –0.321                 | –0.321                    | 0.937           | 1.429           | 0.458 | 32.37         | 1.11|
| Zr                | 0.279                  | 0.279                     | 1.128           | 1.614           | 0.429 | 23.20         | 1.22|
| Nb                | 1.360                  | 1.401                     | 1.154           | 1.612           | 0.419 | 18.18         | 1.23|
| Mo                | 2.105                  | 2.117                     | 1.101           | 1.564           | 0.426 | 15.69         | 1.30|
| Tc                | 1.861                  | 1.861                     | 1.055           | 1.686           | 0.479 | 14.29         | 1.36|
| Ru                | 1.417                  | 1.417                     | 0.972           | 1.628           | 0.503 | 13.51         | 1.42|
| Rh                | 0.597                  | 0.597                     | 0.894           | 1.541           | 0.517 | 13.64         | 1.45|
| Pd                | 0.210                  | 0.210                     | 0.875           | 1.418           | 0.486 | 14.41         | 1.35|
| Ag                | 0.602                  | 0.602                     | 0.931           | 1.345           | 0.433 | 16.33         | 1.42|
| Cd                | 0.447                  | 0.447                     | 0.971           | 1.306           | 0.403 | 19.50         | 1.46|
| In                | 0.899                  | 0.899                     | 0.918           | 1.314           | 0.429 | 24.26         | 1.49|
| Sn                | 1.244                  | 1.243                     | 0.806           | 1.210           | 0.451 | 33.29         | 1.72|
| Cs                | 3.971                  | 3.971                     | 0.159           | 0.742           | 1.397 | 115.77        | 0.86|
| Ba                | 1.454                  | 1.454                     | 0.408           | 0.907           | 0.667 | 63.59         | 0.97|

Properties of Mg–Si–X systems are the solution energy ($E_{sol}$) with the initial magnetic character being considered or not. The SFE, cleavage free surface energy, and ductility parameter are noted as $\gamma_u$, $\gamma_s$, and $D$, respectively. Two features of alloying elements of ground state atomic volume ($V_{GS}$) and Allred–Rochow electronegativity (En) are also listed.

Figure 2. First-principles calculated GSFE curves of pure Mg2Si along the (a) ⟨110⟩ and (b) ⟨112⟩ directions.
solution energies and $V_{GS}$ have a negative correlation. Alloying elements are easier to insert on the Mg site in Mg$_2$Si if their $V_{GS}$ values are similar to those of Mg. After the electronic self-consistent calculations, it is found that the magnetic character of the initially set magnetic alloying elements Ti, Cr, Fe, Co, Nb, and Mo still exists, while other systems become nonmagnetic. Therefore, in the following calculations, only the magnetism of the alloying elements Ti, Cr, Fe, Co, Nb, and Mo is considered.

### 2.2. Generalized SFE of Mg$_2$Si

The generalized SFE (GSFE) is expressed as

$$\gamma_{GSF} = \frac{E_f - E_p}{S}$$

where $E_f$ is the total energy of the stacked configuration with the fault vector $u$. $E_p$ is the total energy of the stacked configuration without slipping. $S$ is the faulted area of the supercell.

In the Mg–Si system, there are two types of slip planes, the Mg–Mg plane and Mg–Si plane. Each slip plane has two possible slip systems, the $\{111\}\{112\}$ slip system and $\{111\}\{110\}$ slip system. Figure 2 presents the GSFE curves of all slip systems for the undoped Mg$_2$Si. The maximum energy on the GSFE curve is the unstable SFE ($\gamma_{us}$), which determines the energy barrier for the nucleation of trailing dislocations. It occurs around $0.8b^{(112)}$ and $0.5b^{(110)}$ for the Mg–Mg plane and at $b^{(112)}$ and $0.5b^{(110)}$ for the Mg–Mg plane. The Burgers vectors are $b^{(112)} = (1/6)a_0$ and $b^{(110)} = (1/6)a_0$. The GSFE curves indicate that the $\gamma_{us}$ of the Mg–Mg plane along the $(110)$ direction is lower than that along the $(112)$ direction, while the $\gamma_{GSF}$ of the Mg–Si plane is oppositely. The Mg–Mg plane exhibits a lower $\gamma_{us}$ compared to the Mg–Si plane in both slip directions. So, the $(110)$ direction on the Mg–Mg plane is the slip system in the Mg$_2$Si alloy.

### 2.3. Alloying Effect of $X$ on the Ductility of Mg$_2$Si

The ductility can be predicted by the ductility parameter $D$. It is the ratio of the cleavage free surface energies $\gamma_s$ to the unstable SFEs $\gamma_{us}$. An increased value of $D$ represents an enhancement of ductile tendency. The ductility parameter $D$ is defined as

$$D = \frac{0.3\gamma_s}{\gamma_{us}}$$

The cleavage free surface energy $\gamma_s$ is given by

$$\gamma_s = \frac{E^{FS1}_{FS} + E^{FS2}_{FS} - E_p}{S}$$

where $E^{FS1}_{FS}$ and $E^{FS2}_{FS}$ represent the total energies of two half slabs, respectively.

The effects of alloying elements on the SFE and ductility of Mg$_2$Si are shown in Figure 3 and also listed in Table 1.
alloying elements of groups I, II, III, and IV have the same trend that as $\gamma_{us}$ decreases, the $D$ increases, indicating that there is an increasing tendency of ductility. This trend can be seen clearly in the $\gamma_{us}$ for a given alloying element.

The variation of $\gamma_{us}$ with respect to the 3d- and 4d-TMs alloying elements seems to be more complicated and follows a concave-up parabolic-like dependency, which is distorted at the right end. Moving to the right of the periodic table, $\gamma_{us}$ tends to increase slightly and peaks at V and Nb where it begins to decrease to its minimum at Ni and Pd. On the whole, the ductility seems to be worse for late TMs, in particular, those of columns 9 and 10. Finally, it is worth mentioning that our ductility seems to be worse for late TMs, in particular, those of group II, In from group III and IV, Cu from 3d-TMs, and Pd from 4d-TMs. They do not change the value of $\gamma_{us}$ in Mg$_2$Si. The ELF vertical slip planes of the five slip systems are depicted in Figure 4. The range of the alloying elements deplete the electrons between Si1 and Si2 atoms. Moreover, due to the different electron attraction ability of alloying elements, the influence of the strength of neighboring Si1–Si2 bonds on the SFE presents a linear trend. The smaller the strength of neighboring Si1–Si2 bonds is, the lower the SFE is.

2.4. Electron Localization Functional Analyses. The effect of alloying elements on the interatomic bonding characteristics of slip planes can be studied in detail by the electron localization functional (ELF). The ELF is very important for an analysis of interatomic bonding mechanisms. We select five typical alloying elements, K from group I, Sr from group II, In from group III and IV, Cu from 3d-TMs, and Pd from 4d-TMs. They do not change the value of $\gamma_{us}$ and decrease the value of $\gamma_{us}$ in Mg$_2$Si. The ELF vertical slip planes of the five slip systems are depicted in Figure 4. The range of ELF is between 0 and 1, where 0 represents no bonding between the adjacent atoms, 0.5 represents a perfect metallic bond, and a higher ELF value indicates that the electrons are more localized (ELF = 1 can be interpreted as perfect localization). All ELF contour plots reveal similar chemical bonding characteristics and indicate that K, Sr, In, Cu, and Pd can weaken the nearest Si–Si bond (Si1–Si2 bond in Figure 4) in Mg$_2$Si alloys.

The ELF value on a line segment with equal steps can be calculated by linear integration of volumetric ELF data. The minimum ELF value in the middle of the Si1–Si2 bond is seen clearly in the ELF images of (a) pristine and (b) K-, (c) Sr-, (d) In-, (e) Cu-, and (f) Pd-alloyed-Mg$_2$Si systems in the (101) plane. The color scale is given at the right.

Figure 4. ELF images of (a) pristine and (b) K-, (c) Sr-, (d) In-, (e) Cu-, and (f) Pd-alloyed-Mg$_2$Si systems in the (101) plane. The color scale is given at the right.

Figure 5. SFE of the different alloying elements as a function of the minimum ELF value in the middle of the Si1–Si2 bond.

The alloying elements decrease the strength of neighboring Si1–Si2 bonds. In experimental studies, compared to Al and Ti, solution of Cu can effectively increase the ductility of Mg$_2$Si.
factor affecting the SFE of Mg2Si. The importance of the electronic effect is relatively low, indicating that its influence on SFE is not as strong as that of the size effect.

The Pearson correlation coefficient is also chosen for evaluating the linear relationship between variables $X$ and $Y$

$$
corr(X, Y) = \frac{cov(X, Y)}{\sigma_X \sigma_Y}$$

where $cov(X, Y)$ represents the covariance between $X$ and $Y$ and $\sigma_X$ and $\sigma_Y$ represent the standard deviations for $X$ and $Y$, respectively. The Pearson correlation coefficient varies from $-1$ to $1$. The value of the coefficient is $1/\sqrt{2}$, which indicates a perfect linear positive/negative correlation between $X$ and $Y$. The value of the coefficient is $0$, which means that there is no linear relationship between the two variables.

Multiple correlation matrices between $\gamma_{\text{us}}$ and 13 features of Mg2Si were computed, as shown in Figure 7. The correlation coefficient $\text{cov}(\gamma_{\text{us}}, V_{\text{GS}})$ was used. It should be noted that the absolute correlation coefficient value of $0.90$ indicates a strong relationship between $\gamma_{\text{us}}$ and $V_{\text{GS}}$. As a consequence, the most possible feature that contributes to SFE is $V_{\text{GS}}$. Other features of the electronic effect have a relatively low correlation with $\gamma_{\text{us}}$ compared to $V_{\text{GS}}$. This is consistent with the above feature importance results. It is worth mentioning that both solution energy and SFE have a strong correlation with $V_{\text{GS}}$. Similarly, $V_{\text{GS}}$ may also affect other properties of the alloy. Therefore, the size effect should be one of the most important features in alloy design.

### 2.6. Statistical Analysis

A visual representation of the relationship between $\gamma_{\text{us}}$ and the most contributing factors $V_{\text{GS}}$.
is shown in Figure 8a. In general, alloying elements with a larger $V_{GS}$ would be favorable as alloying element addition reduces SFE in Mg$_2$Si alloys. It is worth noting that for transitional elements, the opposite trend is shown. As the $V_{GS}$ increases, $\gamma_{us}$ also increases. This may be due to the fact that for alloying elements with $V_{GS}$ greater than Mg, as the $V_{GS}$ increases, the distance between the alloying element and Si1 increases (as shown in Figure 9), and the interlayer spacing of the slip plane increases, which, in general, weakens the Si1−Si2 bond, resulting in a decrease in $\gamma_{us}$. For alloying elements with $V_{GS}$ less than Mg, the distance between the alloying element and Si1 is always smaller than the distance between Mg and Si1 in the pure Mg$_2$Si system. Such trends are caused by the competition between the size effect and the electronic effect, as shown in Figure 8b. For alloying elements with $V_{GS}$ less than Mg, as En increases, $\gamma_{us}$ decreases. The greater the En is, the stronger the ability to attract electrons in the compound is, which reduces the electron distribution between Si1 and Si2, weakens the Si1−Si2 bond, and results in a decrease in $\gamma_{us}$. However, large $V_{GS}$ also reduces the $\gamma_s$ of the Mg$_2$Si system, which is not desirable, as shown in Table 1, because it will make the system easier to break. In summary, addition of alloying elements with appropriate $V_{GS}$ and higher En (such as In, Cu, Pd, etc.) would exhibit lower $\gamma_{us}$ and little change in $\gamma_s$. 

Figure 7. Correlation map between $\gamma_{us}$ and 13 features of alloying elements.

Figure 8. Variation of calculated SFE with respect to (a) $V_{GS}$ of alloying elements and (b) En of alloying elements.
which means that the Mg–Si–X system would yield a better ductility.

3. CONCLUSIONS

First-principles calculations in the context of DFT have been carried out to determine the SFE of 33 alloying elements across the periodic table in Mg2Si. It is found that the addition of alloying elements with various size effect and electronic effect properties led to a large variation of SFE and ductility. Among the properties of alloying elements, the variation of SFE was found to be strongly related to the $V_{GS}$ in the size effect. At the same time, the electronic effect will compete with the size effect to have an impact on SFE. An appropriate $V_{GS}$ and higher En would reduce the $\gamma_{us}$ in the Mg–Si–X system and yield a better ductility. The alloying elements In, Cu, Pd, etc. are identified to be beneficial to the ductility of Mg2Si alloys. We anticipate that the conclusions and rules obtained in the present work will provide profound guidelines for further alloying design in the promising Mg2Si alloys with better ductility.

4. METHODS

Mg2Si has an FCC structure corresponding to the space group of Fm–3m. Mg and Si atoms are located at the 4a (0,0,0) and 8c (1/4,1/4,1/4) sites, as shown in Figure 10a. The solute preferred insertion on the Mg site because of the smaller formation energy.40 To calculate the solution energies of Mg–Si–X systems, a $2 \times 2 \times 2$ supercell containing 63 Mg atoms, 32 Si atoms, and 1 single kind of solute element substituting one Mg atom was built.

To study the unstable SFs, we employed a slab model of the slip system with 18 layers and 4 atoms in each layer (Figure 10b). Although the interaction between stacking fault and solutes can be extended to several close-packed layers, most of the solutes tend to stably aggregate in the faulted region.18 Therefore, in the cases of Mg–Si–X systems, one Mg atom in the first nearest-neighbor plane with respect to the stacking fault plane was substituted with an X atom, as shown in Figure 10c. A vacuum layer with a 10 Å thickness was introduced into the slab to avoid image interactions. The atomic positions were only allowed to relax along the direction perpendicular to the slipping layers.

In this study, the DFT41 calculations were performed using the Vienna Ab initio Simulation Package.42,43 The interaction potential of the core electrons was described using the projector augmented wave method.44 The generalized gradient approximation with Pardew–Burke–Ernzerhof parameterization for the exchange correction function was adopted. The cutoff energy for the plane wave basis set was 450 eV. The $k$-points were meshed by $2 \times 2 \times 2$ for the calculation of solution energies and $3 \times 3 \times 1$ for the calculation of SFE.46 The semicore $p$ electrons for all of the elements were treated as valence electrons when available.44,47

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Figure 9. Distance between alloying elements and Si1 in the Mg2Si system as a function of the $V_{GS}$.

Figure 10. (a) Unit cell of Mg2Si, (b) schematic view of a slab model used for SFE calculations, and (c) displacement of the $\{111\}\langle 110 \rangle$ slip system to explain the formation of the stacking fault. Mg, Si, and alloying elements are colored in green, light blue, and red, respectively.
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References

1. Liu, W.; Tan, X. J.; Yin, K.; Liu, H. J.; Tang, X.; Shi, J.; Zhang, Q.; Uher, C. Convergence of conduction bands as a means of enhancing thermoelectric performance of N-Type Mg$_2$Si$_x$Sn$_{1-x}$ solid solutions. Phys. Rev. Lett. 2012, 108, No. 166601.

2. Jiang, G.; He, J.; Zhu, T.; Fu, C.; Liu, X.; Hu, L.; Zhao, X. High performance Mg$_2$(Si,Sn) solid solutions: A point defect chemistry approach to enhancing thermoelectric properties. Adv. Funct. Mater. 2014, 24, 3776–3781.

3. Hirayama, N.; Iida, T.; Funashima, H.; Morioka, S.; Sakamoto, M.; Nishio, K.; Kogo, Y.; Takashni, Y.; Hamada, N. First-principles study on structural and thermoelectric properties of Al-and Sn-doped Mg$_2$Sn. J. Electron. Mater. 2015, 44, 1656–1662.

4. Farahi, N.; Prabhudeva, S.; Bottom, G. A.; Salvador, J. R.; Kleinke, H. Nano-and microstructure engineering: An effective method for creating high efficiency magnesium silicide based thermoelectric devices. ACS Appl. Mater. Interfaces 2016, 8, 34431–34437.

5. Mao, J.; Kim, H. S.; Shuai, J.; Liu, Z.; He, R.; Saparamadu, U.; Tian, F.; Liu, W.; Ren, Z. Thermoelectric properties of materials near the band crossing line in Mg$_2$Sn–Mg$_2$Ge–Mg$_2$Si system. Acta Mater. 2016, 103, 633–642.

6. Yan, D.; Cuevas, F.; Latroche, M.; Percheron-Guégan, A. Influence of crystallinity on the structural and hydrogenation properties of Mg$_2$X phases (X = Ni, Si, Ge, Sn). Intermetallics 2006, 14, 163–169.

7. Udono, H.; Yamanaka, Y.; Uchikoshi, M.; Ishihara, M. Infrared photorepsonse from pn-junction Mg$_2$Si diodes fabricated by thermal diffusion. J. Phys. Chem. Solids 2013, 74, 311–314.

8. Li, A.; Zhao, X. P.; Huang, H. Y.; Ma, Y.; Gao, L.; Su, Y. J.; Qian, P. Fine-tuning the ductile-brittle transition temperature of Mg$_2$Si intermetallic compound via Al doping. Int. J. Miner., Metall. Mater. 2019, 26, 507–515.

9. Wang, Y.; Choo, H. Influence of texture on Hall–Petch relationships in an Mg alloy. Acta Mater. 2014, 81, 83–97.

10. Basu, S.; Dogan, E.; Kondori, B.; Karaman, I.; Benzeraga, A. Towards designing anisotropy for ductility enhancement: A theory-driven investigation in Mg-alloys. Acta Mater. 2017, 131, 349–362.

11. Zheng, R.; Bhattacharjee, T.; Shibata, A.; Sasaki, T.; Hono, K.; Joshi, M.; Tsujii, N. Simultaneously enhanced strength and ductility of Mg-Zn-Zr-Ca alloy with fully recrystallized ultrafine grained structures. Scr. Mater. 2017, 131, 1–5.

12. Mahjoub, R.; Laws, K. J.; Stanford, N.; Ferry, M. General trends between solute segregation tendency and grain boundary character in aluminum-an ab initio study. Acta Mater. 2018, 158, 257–268.

13. Li, G.; Gill, H.; Varin, R. Magnesium silicide intermetallic alloys. Metall. Trans. A 1993, 24, 2383–2391.

14. Chen, S.; Qu, X.; Lei, C.; Huang, B. Room temperature mechanical properties of ordered TiAl-La alloys. Acta Metall. Sin. 1994, 30, 20–24.

15. Qi, Y.; Mishra, R. K. Ab initio study of the effect of solute atoms on the stacking fault energy in aluminum. Phys. Rev. B 2007, 75, No. 224105.

16. Wang, C.; Zhang, H. Y.; Wang, H. Y.; Liu, G. J.; Jiang, Q. C. Effects of doping atoms on the generalized stacking-fault energies of Mg alloys from first-principles calculations. Scr. Mater. 2013, 69, 445–448.

17. Zhang, J.; Dou, Y.; Dong, H. Intrinsic ductility of Mg-based binary alloys: A first-principles study. Scr. Mater. 2014, 89, 13–16.

18. Wu, Y.; Li, S.; Ding, Z.; Liu, W.; Zhao, Y.; Zhu, Y. Effect of charge redistribution factor on stacking-fault energies of Mg-based binary alloys. Scr. Mater. 2016, 112, 101–105.

19. Tsuru, T.; Somekawa, H.; Chranz, D. Interfacial segregation and fracture in Mg-based binary alloys: Experimental and first-principles perspective. Acta Mater. 2018, 151, 78–86.

20. Dong, Q.; Luo, Z.; Zhu, H.; Wang, L.; Ying, T.; Jin, Z.; Li, D.; Ding, W.; Zeng, X. Basal-plane stacking-fault energies of Mg alloys: A first-principles study of metallic alloying effects. J. Mater. Sci. Technol. 2018, 34, 1773–1780.

21. Stanev, V.; Oses, C.; Russe, A. G.; Rodriguez, E.; Paglione, J.; Curtarolo, S.; Takeuchi, I. Machine learning modeling of superconducting critical temperature. npj Comput. Mater. 2018, 4, 1–14.

22. Pei, Z.; Yin, J.; Hawk, J. A.; Alman, D. E.; Gao, M. C. Machine-learning informed prediction of high-entropy solid solution formation: Beyond the hume-rothery rules. npj Comput. Mater. 2020, 6, 1–8.

23. Kong, X. S.; Wu, X.; You, Y. W.; Liu, C.; Fang, Q.; Chen, J. L.; Luo, G. N.; Wang, Z. First-principles calculations of transition metal–halide interactions with point defects in tungsten. Acta Mater. 2014, 66, 172–183.

24. Tatsuoka, H.; Takagi, N.; Okaya, S.; Sato, Y.; Inaba, T.; Ohishi, T.; Yamamoto, A.; Matsuyama, T.; Kuwabara, H. Microstructures of semiconducting silicide layers grown by novel growth techniques. Thin Solid Films 2004, 461, 57–62.

25. Rice, J. R. Dislocation nucleation from a crack tip: An analysis based on the piersi concept. J. Mech. Phys. Solids 1992, 40, 239–271.

26. Shi, S.; Zhu, L.; Zhang, H.; Liu, G.; Jiang, Q.; Sun, Z.; Ahuja, R. Mapping the relationship among composition, stacking fault energy and ductility in Nb alloys: A first-principles study. Acta Mater. 2018, 144, 853–861.

27. Ni, A. Effects of alloy elements on the ductilebrittle transition temperature of Mg$_2$Si. Master Dissertation, University of Science and Technology Beijing, Beijing, China, 2018. http://thesis.ustb.edu.cn/docinfo.action?id=144e9d5f8f9b4a553f34464141cb2d6c&cid=2SvysbPfXokj%2520S

28. Xu, M.; Cheng, Y.; Sheng, H.; Ma, E. Nature of atomic bonding and atomic structure in the phase-change Ge$_2$Sb$_2$Te$_5$ glass. Phys. Rev. Lett. 2009, 103, No. 195502.

29. Shang, S.; Wang, Y.; Du, Y.; Tschoopp, M. A.; Liu, Z. K. Incorporating computational modeling and first-principles calculations to predict stacking fault energy of dilute multicomponent Ni-base alloys. Comput. Mater. Sci. 2014, 91, 50–55.

30. Achmad, T. L.; Fu, W.; Chen, H.; Zhang, C.; Yang, Z. G. Cobased alloys design based on first-principles calculations: Influence of transition metal and rare-earth alloying element on stacking fault energy. AIP Conf. Proc. 2017, 1805, No. 060004.

31. Webelements. http://www.webelements.com (accessed Feb 07, 2020).

32. Pauling File. http://www.paulingfile.com/ (accessed Feb 07, 2020).

33. Ward, L.; Agrawal, A.; Choudhary, A.; Wolverton, C. A general-purpose machine learning framework for predicting properties of inorganic materials. npj Comput. Mater. 2016, 2, 1–7.

34. PeriodicTable. https://www.periodictable.com/ (accessed Feb 07, 2020).

35. Breiman, L. Random forests. Mach. Learn. 2001, 45, 5–32.
(36) Svetnik, V.; Liaw, A.; Tong, C.; Culberson, J. C.; Sheridan, R. P.; Feuston, B. P. Random forest: A classification and regression tool for compound classification and qsar modeling. *J. Chem. Inf. Model.* 2003, 43, 1947-1958.

(37) Loh, W. Y. Fifty years of classification and regression trees. *Int. Stat. Rev.* 2014, 82, 329-348.

(38) Loh, W. Y. Classification and regression trees. *Wiley Interdiscip. Rev.: Data Min. Knowl.* 2011, 1, 14-23.

(39) Cutler, A.; Cutler, D. R.; Stevens, J. R. Random forests. In *Ensemble Machine Learning*; Springer: Boston, MA, 2012, pp 157-175.

(40) Tani, J. I.; Kido, H. First-principles and experimental studies of impurity doping into MgSi. *Intermetallics* 2008, 16, 418-423.

(41) Kohn, W.; Sham, L. J. Self-consistent equations including exchange and correlation effects. *Phys. Rev.* 1965, 140, No. A1133.

(42) Kresse, G.; Hafner, J. Ab initio molecular dynamics for open-shell transition metals. *Phys. Rev. B* 1993, 48, No. 13115.

(43) Kresse, G.; Furthmüller, J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. *Phys. Rev. B* 1996, 54, No. 11169.

(44) Blöchl, P. E. Projector augmented-wave method. *Phys. Rev. B* 1994, 50, No. 17953.

(45) Perdew, J. P.; Burke, K.; Ernzerhof, M. Generalized gradient approximation made simple. *Phys. Rev. Lett.* 1996, 77, No. 3865.

(46) Monkhorst, H. J.; Pack, J. D. Special points for brillouin-zone integrations. *Phys. Rev. B* 1976, 13, No. 5188.

(47) Kresse, G.; Joubert, D. From ultrasoft pseudopotentials to the projector augmented-wave method. *Phys. Rev. B* 1999, 59, No. 1758.