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Phase selection in aluminum rare-earth metallic alloys by molecular dynamics simulations using machine leaning interatomic potentials

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Abstract

Aluminum-rare-earth (Al-RE) metallic alloys and glasses with Al-rich compositions have attracted much attention owing to their high strength-to-weight ratio and superior thermal stability. However, difference in phase selection and formation upon using light or heavy RE elements are still not well understood. Using $Al_{90}Ce_{10}$ and $Al_{90}Tb_{10}$ as prototype Al-rich light-RE and heavy-RE alloys, we study the similarity and difference of phase selection and glass formation ability (GFA) in the two metallic glasses by molecular dynamics (MD) simulations using accurate machine learning (ML) interatomic potentials and cluster alignment analysis. We showed that the glass-forming structural short-range order (SRO) motifs are much stronger in $Al_{90}Tb_{10}$ glass than in the $Al_{90}Ce_{10}$ glass (77% vs 47%). On other hand, there is a noticeable fraction of $Al_{11}RE_3$ and hexagonal Al_3RE crystalline SRO motifs in $Al_{90}Ce_{10}$ glass which are almost absent in $Al_{90}Tb_{10}$. The origin of the SRO difference is investigated by comparing the structure-energy landscapes in these two systems, where novel competing meta-stable structures are obtained from adaptive genetic algorithm (AGA) search. MD simulations using the ML potentials and a solid/liquid interface model are also performed to show that the crystal growth speed in the Al-Ce is about 2-3 time faster than that in the Al-Tb system at similar undercooling conditions. Diffusion constant in the undercooled liquids and the latent heat of several competing crystalline phases and their liquids are also calculated to elucidate the different crystallization behaviors in the two systems. Moreover, chemical order preference in the two systems is also analyzed. Our results suggest that the different phase selection and GFA in the two systems can be attributed to relative energetic stabilities of various competing metastable phases which lead to different structural and chemical SROs and different crystallization driving forces which influence the phase selection kinetics of the two systems.

I. Introduction

Light-weight high-temperature alloys are important to the transportation industry. In designing energy efficient vehicles and airplanes, weight, cost and operating temperature of the alloys are major factors of consideration. Aluminum-rare-earth (Al-RE) metallic alloys and glasses with Al-rich compositions have attracted much attention owing to their high strength-to-weight ratio. With more than 85 at. % Al and under rapid cooling, Al-RE alloys can form either metallic glass or metastable crystalline compounds in the process of solidification, depending on the details of cooling rates and protocols [1].

The formation of metallic glasses around $Al_{90}RE_{10}$ compositions have been extensively studied in the past several decades [2-12]. It has been shown that the glass formation ability (GFA) in these systems varies considerably with the RE elements. The best binary Al-RE glass formers are Al-Sm and Al-Tb, while Al-Ce alloy has a much weaker GFA. Recently, it has been have shown that alloying aluminum with cerium can produce crystalline alloys with dramatically improved high-temperature performance which are easily cast or readily "printed" using additive manufacturing compatible to traditional aluminum alloy additions [13-15]. Al-Ce based-alloys have the potential to replace heavier steel and cast-irons for use in elevated-temperature applications beyond current Al alloys. Understanding the fundamental interactions and mechanism governing the phase selection and stability in Al-RE based systems are therefore highly desirable for guiding the design, discovery, and synthesis of novel Al-RE binary and Al-RE-X (X = Mg, Zr, …) ternary intermetallic compounds for energy-efficient applications. In particular, La and Ce are 'in excess supply' relative to the other REs [16, 17]. More high-value uses for these abundant REs in alloy design will provide a more robust market for the RE industry.

Many questions about the role of different RE elements played in the phase selection and thermodynamic stability of Al-RE based alloys remain open. These questions include what is fundamentally different between light RE (LRE) and the heavy RE (HRE), and what role does size and valence play in the phase selection and stability? More specifically, it is vital to understand (a) How are the short-range order (SRO) and medium range order (MRO) in the liquids and during the rapid solidification affected by different RE elements in Al-RE based alloys? (b) What are the local structure motifs in competing stable and metastable crystalline phases of Al-RE based alloys with different RE elements and can the differences in the ground state structures tell us about GFA? and (c) How different RE element in Al-RE based alloys influence the nucleation and growth kinetics of various competing crystalline phases?

While many experimental efforts have been devoted to investigating the local structure order in metallic glasses [1, 18, 19], elucidating the detailed atomic structure in terms of SRO and MRO is still very challenging for experimental studies. On the other hand, in the last 10 years, we have demonstrated that by integrating molecular dynamics (MD) simulations with the local structure order analysis using the cluster alignment method [20] can provide a very powerful approach to quantify the SRO and MRO in metallic liquids and glasses at the atomic scale [21-23]. More importantly, MD simulations can also provide critical insight into the mechanisms controlling kinetics nucleation and growth [24]. However, the bottleneck in MD simulation studies is the availability of the reliable interatomic potentials for the systems of interest.

Recently, we have shown that accurate and transferable interatomic potentials for Al-RE systems can be developed through a deep machine learning (ML) strategy using an artificial neural network (ANN) [25]. Such ANN-ML interatomic potentials have been successfully generated for Al-Tb and Al-Ce systems [26-28]. The ANN-ML potential for Al-Tb system has also been used to perform MD simulation to investigate SRO and MRO in $Al_{90}Tb_{10}$ undercooled liquid and glass [26, 27]. We show that the SRO in $Al_{90}Tb_{10}$ undercooled liquid and glass are strongly correlated with the building blocks of several novel low-energy metastable crystalline structures in the vicinity of the $Al_{90}Tb_{10}$ composition [27]. However, none of these SRO motifs match well with the structures of the stable $Al_{11}RE_3$ or Al_3RE phases which are the known to be common stable crystalline phases of Al-LRE and Al-RE alloys respectively.

In order to understand the difference in GFA and phase selection in Al-rich alloys with LRE or HRE, in this paper we use $Al_{90}Ce_{10}$ and $Al_{90}Tb_{10}$ as prototypes for Al-LRE and Al-HRE metallic alloys and performed detail analysis for the SRO and MRO differences in $Al_{90}Ce_{10}$ and $Al_{90}Tb_{10}$ glasses. Both glass samples are generated by MD simulations using the reliable ANN-ML interatomic potentials, and cluster alignment analysis is performed to quantify the SRO and MRO in the two systems. We also performed crystal structure predictions based on adaptive genetic algorithm (AGA) and ANN-ML interatomic potentials to search for new energetically favorable metastable structures around the composition of $Al_{90}RE_{10}$. The results from our AGA search provide comprehensive energy landscapes which are very useful for understanding the thermodynamic driving forces for the phase selections in the two systems. Finally, we performed MD simulations (using the ANN-ML potentials) to study the crystallization process in the two systems to elucidate possible kinetic factors influencing the phase selection.

The results from our simulations and analysis show that although large fraction of glass forming motifs are also observed in $Al_{90}Ce_{10}$ glass, the percentage of such motifs is substantially smaller than that in $Al_{90}Tb_{10}$ glass. On the other hand, there are a noticeable fraction (~24%) of $Al_{11}RE_3$ crystalline SRO motifs in $Al_{90}Ce_{10}$ glass which are almost absent in $Al_{90}Tb_{10}$ glass, which is not surprising since the $Al_{11}RE_3$ is not a stable phase in the HRE binary systems [29]. Moreover, about 10% of the Ce-centered SRO clusters in $Al_{90}Ce_{10}$ glass belong to hexagonal Al_3Ce crystalline motif, while such clusters in $Al_{90}Tb_{10}$ glass is only about 2%. Crystal structure prediction using AGA also confirms that crystal structures with the building blocks similar to the motifs of $Al_{11}RE_3$ crystal structure are energetically more favorable in Al-Ce system than in Al-Tb system. The difference in the relative energy landscapes in the two systems would lead to different phase selection behaviors during solidification process. We also found that Tb-Al bond lengths of the glass forming SRO motifs in the glass is very different (~7.0% in average) from that of the nearby stable Al_3Tb phase, which would make the crystallization in Al-Tb system more difficult. Our MD simulation of crystallization from the solid-liquid interfaces also confirm that crystal growth proceeds faster in Al-

Ce system than in Al-Tb system. Our simulation results thus provide useful insights for understanding the different GFA and phase selection in Al-LRE and Al-HRE systems.

The paper is arranged as follows. In section II, we describe the computational methods used in our MD simulations and the SRO and MRO structure analyses. Methods for searching the lower-energy crystalline structures including AGA and *ab initio* calculations are also briefly described in section II. The results obtained from our computational studies analyses, for given chemical compositions, including the SRO and MRO in the two glasses as well as structure-energy landscapes and comparison of crystal growth kinetics in the two systems, are presented and discussed in section III. Finally, a summary is given in section IV.

II. Computational Methods

MD simulations - MD simulations to generate $Al_{90}Ce_{10}$/$Al_{90}Tb_{10}$ glass samples and to study crystal growth at solid/liquid interfaces are performed using ANN-ML interatomic potentials and the LAMMPS package [30]. Our ANN-ML interatomic potentials for Al-Ce and Al-Tb systems developed [26, 28] previously are further refined by including more training data from metastable crystalline structures through the AGA scheme described below so that the ANN-ML interatomic potentials describe accurately not only for liquid and glass structures but also for crystalline structures with various bonding topologies. The updated accurate and transferable ANN-ML potentials for Al-Ce and Al-Tb alloys are provided in the present the Supplementary Materials [31]. Atomistic structures of $Al_{90}Ce_{10}$/$Al_{90}Tb_{10}$ glass samples are generated by cooling from liquid states using 5000 atoms (4500 Al and 500 Ce or Tb) and with periodic boundary conditions. An isothermal-isobaric (NPT) ensemble at zero pressure and a Nose-Hoover thermostat [32, 33] are used in the simulations. The MD time step is taken as 2.5 fs. The liquid $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ samples are equilibrated at 2000K for 500 ps respectively, and then are continuously cooled down to 300K at cooling rates of 10^{10} K/s. The sample configurations averaged over 500 ps at 300K are used to analyze the structure order in glassy $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ by the template cluster alignment method. MD simulations of crystallization at solid/liquids interface are performed using the same LAMMPS package, the same ANN-ML potentials and the same time step of 2.5 fs. More details of these simulations will be given in section 3.3.

SRO and MRO analysis - In order to investigate the local SRO and MRO in the glass samples generated by the MD simulations, template cluster alignment method previously developed [20] is employed. Quai-spherical clusters with size of 70 atoms covering at least three atomic shells around each RE atom in the $Al_{90}RE_{10}$ samples from the MD simulations are extracted. We refer to these atomic clusters as sample clusters. Similar alignment scheme is also applied to Al-centered clusters as discussed in Supplementary Materials [31]. The first atomic shell of these sample clusters is then aligned one-by-one against several given templates (which are clusters containing only the first shell atoms around the center atom) to see how the SRO of the clusters extracted from the MD samples are similar to each of the given template, respectively. The similarity is measured by an alignment score s which is defined as,

$$s = \min_{0.8 \le \alpha \le 1.2} \left\{ \left[\frac{1}{N} \sum_{i=1}^{N} \frac{(\vec{r}_{ic} - \alpha \vec{r}_{it})^2}{(\alpha \vec{r}_{it})^2} \right]^{1/2} \right\} \qquad (1)$$

where N is the number of the first shell atoms in the template. $\vec{r}_{ic}$ and $\vec{r}_{it}$ are the atomic positions in the sample cluster and template, respectively. α is chosen between 0.8 and 1.2 to vary the size of the template for an optimal alignment. From the alignment score defined in Eq. (1), we can see that the smaller the value of s is, more similar is the sample cluster to the template. The alignment is performed by simulated annealing to minimize the alignment score s, using molecular dynamics simulations with a strong attractive potential between a pair of atoms in the sample cluster and the template respectively. The internal structures of both the sample and template clusters are fixed (except the bond lengths of the template which are allowed to breath by a factor $0.8<\alpha<1.2$ to give a better fit to that of the sample cluster) during the alignment, therefore only the rigid translation and rotation of whole cluster are allowed during the MD simulations. After all the clusters extracted from the glass samples have been aligned respectively with the templates using the procedure described above, the motif and degree of SRO in the glass samples can be determined through the histograms of the alignment score distribution with respect to each template. The sample clusters extracted from the glass samples can be classified into different template motifs according to their smallest alignment score to the template if this score is also below the given cutoff score. Moreover, by overlapping the center atoms of the sample clusters belonged to the same template motif while keeping orientations of the clusters in their best alignment positions, the distribution of the first cluster shell atoms will also give a visible measure of the degree of the SRO motif, while the distribution of the atoms in the 2nd and 3rd cluster shells (and beyond) provides the information about MRO motifs in the glass samples.

Crystal structure search – New stable and meta-stable crystalline structures in Al-Tb and Al-Ce alloys in the compositions close to $Al_{90}RE_{10}$ are explored using the adaptive genetic algorithm (AGA) code developed previously [35, 36]. In AGA, the most time-consuming structure relaxation and energy evaluation for offspring structures in the GA loop is done efficiently by using an auxiliary interatomic potential instead of direct *ab initio* calculations. The auxiliary interatomic potential is adjusted adaptively in an adaptive loop using feedbacks from *ab initio* calculations on some selected structures obtained by the GA search to ensure the accuracy and reliability of the GA predictions. Different from previous AGA studies where the auxiliary interatomic potentials are analytically expressed in a given mathematical formula (e.g., LJ or EAM) with some adjustable parameters, ANN-ML interatomic potential is used as the auxiliary interatomic potential in the present AGA search. By defining the total cost function in the ANN-ML training as a weighted sum of the errors in the energy and forces from each structure in the training database, the ANN-ML auxiliary potential during the AGA search is adaptively refined by adjusting the weight factors of different datasets. The ANN-ML potentials after such adaptive refinement describe well not only the liquid and glass structures but also crystalline structures with various bonding topologies, thus ensure the accuracy and reliability of the discovery of the low-energy competing meta-stable phases in these two systems. The refined ANN-ML potentials for Al-Tb and Al-Ce systems are given in the Supplementary Materials [31]. These potentials are also used in the MD simulations in this study. For a given ANN-ML potential, GA search starts from a random generated initial structure pool. The cut-and-paste operation with some mutations implemented in the AGA code is used to generate the offspring structures from generation to generation. Structures in the Al-Tb and Al-Ce structure pools are exchanged from time to time during the GA search to enhance the diversity of the structures for each system. The GA search with a given ANN-ML potential is stopped when the lowest-energy structure in the pool is unchanged in 200 consecutive generations. Such GA protocol is repeated with newly adjusted ANN-ML potentials or newly generated initial structure pools until sufficient number of low-energy structures with verification from *ab initio* calculations are obtained.

Ab initio calculations - The *ab initio* calculations during and AGA search and the final optimization of the meta-stable structures from AGA are performed based on density functional theory (DFT) using VASP package [36, 37]. Perdew-Burke-Ernzerhof (PBE) functional [38] combined with the projector-augmented wave (PAW) method [39] and default cutoff energy in pseudopotential are used. We use a k-point grid with a mesh size of $2\pi \times 0.02$ Å^{-1} generated by the Monkhorst-Pack scheme. This mesh size is fine enough to sample the first Brillouin zone for achieving better k-point convergence. In the final optimization of the meta-stable structures by DFT, the atomic positions and the lattice vectors of the unit cells are fully relaxed until the force on every atom is less than 0.01 eV/Å.

III. Results and Discussions

3.1 SRO and MRO in glasses

Fig. 1 shows the temperature dependence of instantaneous potential energy E-3k_BT for $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ system, respectively, during the continually cooling process with a cooling rate of 10^{10} K/s. In Fig. 1, we also plotted the white lines guided by eyes to show the slope of potential energy changes. For both $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$, the estimated glass transition temperature from the intersection point of the two white slope lines in the potential energy is about 600K. However, the slope (ΔE/ΔT) in the region of undercooled liquid (1000K-600K) is about 0.16 meV/K for $Al_{90}Tb_{10}$ and 0.23 meV/K for $Al_{90}Ce_{10}$, respectively. It shows that the decreasing of potential energy in $Al_{90}Ce_{10}$ is faster than $Al_{90}Tb_{10}$ upon quench process, suggesting that the atomic structure in $Al_{90}Tb_{10}$ is more sluggish than that in $Al_{90}Ce_{10}$ and the glass forming ability (GFA) of $Al_{90}Tb_{10}$ would be greater than $Al_{90}Ce_{10}$. The suggestion of better GFA in $Al_{90}Tb_{10}$ is consistent with experimental observation [1] and will be investigated in more details below.

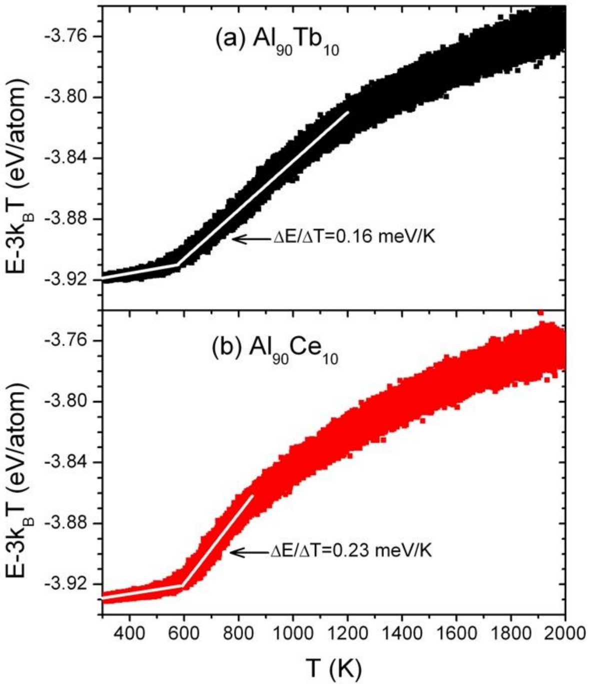


Fig. 1. The temperature dependence of potential energy E-3k_BT for (a) $Al_{90}Tb_{10}$ and (b) $Al_{90}Ce_{10}$ from MD simulations.

The pair correlation functions and the structure factors of $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ at T=300K obtained from our MD simulations are compared in Fig. 2. From the total and partial pair correlation functions shown in Fig. 2 (a) - (d), we can see that the splitting of the first peak in total g(r) is more obvious in $Al_{90}Ce_{10}$ than in $Al_{90}Tb_{10}$. This difference can be attributed to fact that the bond lengths between Al and RE atoms and that among the RE atoms are slightly longer in $Al_{90}Ce_{10}$ than in $Al_{90}Tb_{10,}$ as one can see from the corresponding Al-RE and RE-RE partial pair correlation functions shown in Fig. 2 (c) and (d). These results indicate that the atomic packing in $Al_{90}Tb_{10}$ glass is more compact than that in $Al_{90}Ce_{10}$ glass consistent with the well know Lanthanide contraction. The total and weighted partial structure factors (as defined in ref. 26) shown in Fig. 2 (e)-(h) suggest that the SRO and MRO in $Al_{90}Tb_{10}$ glass in more pronounce than those in $Al_{90}Ce_{10}$ glass, as the peaks in the structure factors of $Al_{90}Tb_{10}$ are sharper than those of $Al_{90}Ce_{10}$. These observations are also confirmed by the local structure order analysis using the cluster alignment method as discussed below.

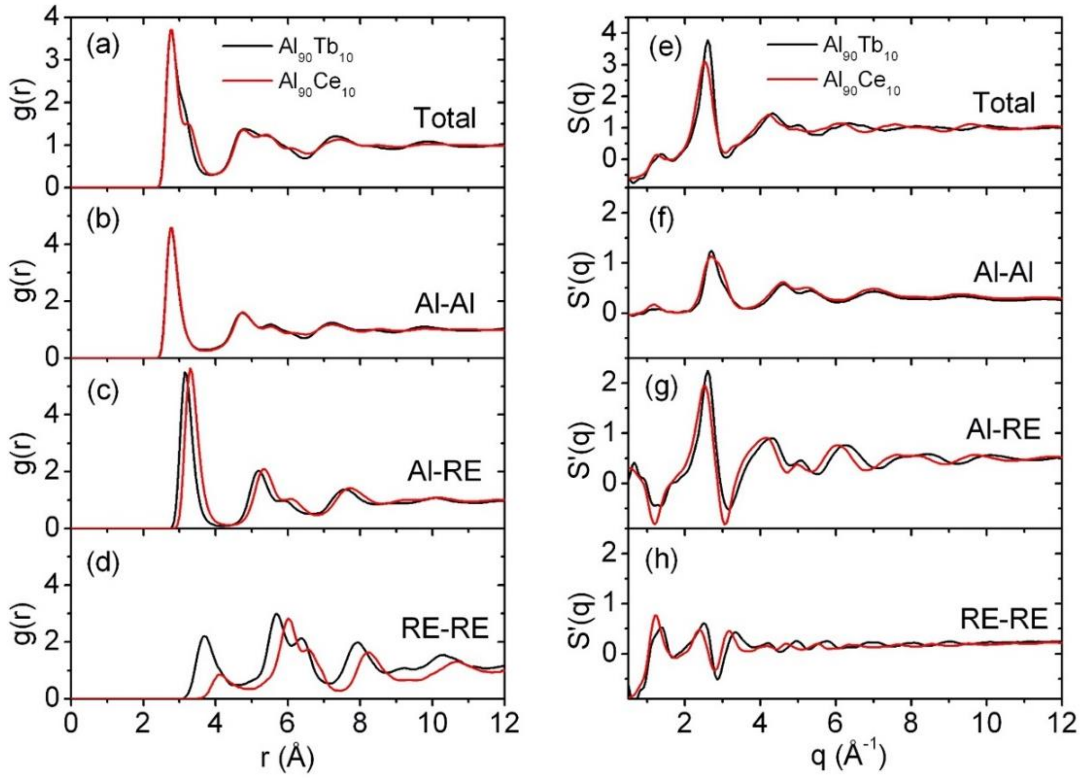


Fig. 2. (a)-(d) The total and partial pair correlation functions for $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ at T=300K. (e)-(h) The total and weighted partial structure factors for $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ at T=300K. The results of total structure factors show that the structure of $Al_{90}Tb_{10}$ is more ordered than that of $Al_{90}Ce_{10}$.

Fig. 3(a)-(b) show the alignment score distributions for the Tb-centered and Ce-centered clusters in the $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ glass samples with respect to several relevant SRO templates as labeled. The '3661' is a RE-centered cluster whose atomic shell consists of a top triangle, two consecutive hexagons in the middle and a single atom at the bottom. The '15551' is also a RE-centered cluster whose atomic shell has three consecutive pentagonal rings and capped by an atom on the top and bottom rings respectively. Our previous studies have shown that the '3661' and '15551' clusters are the two major RE-centered glass forming SRO motifs in Al-rich Al-RE (RE=Sm, Tb, Ce) liquids and glasses [21, 26-28, 40]. $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 are the two RE-centered SRO motifs around the two non-equivalent Ce sites in $Al_{11}Ce_3$ crystal structure. For Al_3RE crystalline phase, there are two RE-centered SROs structures. One is in cubic-Al_3RE crystal (with cubic unit cell) and the other is in hex-Al_3RE crystal (with hexagonal unit cell) respectively. The cubic-Al_3RE crystal has FCC structures where the Al atoms occupy the face centers and RE atoms occupy the corner of the cell. The hex-Al_3RE crystal are built by hexagonal closest-packed layers (as …ABABA… sequence) where one RE atom contacts six Al atoms. Two RE-centered clusters are extracted respectively from the cubic and hexagonal Al_3RE crystalline structures for the templates of the alignment analysis. By noting that the smaller alignment score indicates the sample cluster is more similar to the template, we can see from Fig. 3 (a) and (b) that although ‘3661’ and ‘15551’ clusters are dominate SROs in both glass samples, the degrees of such SROs are much stronger in the $Al_{90}Tb_{10}$ than in the $Al_{90}Ce_{10}$. On the other hand, $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 SROs are more pronounce in the $Al_{90}Ce_{10}$ than in the $Al_{90}Tb_{10}$ glass samples, although they are not as strong as the ‘3661’ and ‘15551’ SROs. If we use the score cutoff value of 0.16 to assign the sample clusters to the templates according our previous studies [26-28], the fractions of the 6 types of SRO clusters can be quantified as shown by the pie chart in Fig. 3(c) and (d). We can see that in the $Al_{90}Tb_{10}$ glass sample, the ‘3661’ and ‘15551’ clusters are predominant with 77% of the Tb-centered clusters, while $Al_{11}RE_3$-1/2 and Al_3RE SROs are only about 5%. By contrast, in the $Al_{90}Ce_{10}$ glass sample, the percentage of the ‘3661’ and ‘15551’ clusters are 47% while that of $Al_{11}RE_3$-1/2 SROs are as many as 24%. Moreover, the hex-Al_3RE motif in $Al_{90}Ce_{10}$ glass is also about 10% while there is only ~2.0% of such a motif in $Al_{90}Tb_{10}$ glass even hex-Al_3Tb is the nearby stable crystalline structure. Good GFA in $Al_{90}Tb_{10}$ would be attributed to its strong tendency in forming ‘3661’ and ‘15551’ SRO rather than crystalline SRO motifs upon rapid solidification. On the other hand, formation of substantial crystalline SRO motifs in the $Al_{90}Ce_{10}$ would be responsible for the weaker glass formation ability of Al-Ce alloy. For Al-centered clusters, ICO and disordered ICO motifs are predominate SRO motifs in both glasses as one can see from Fig. S2 in the Supplementary Materials [31].

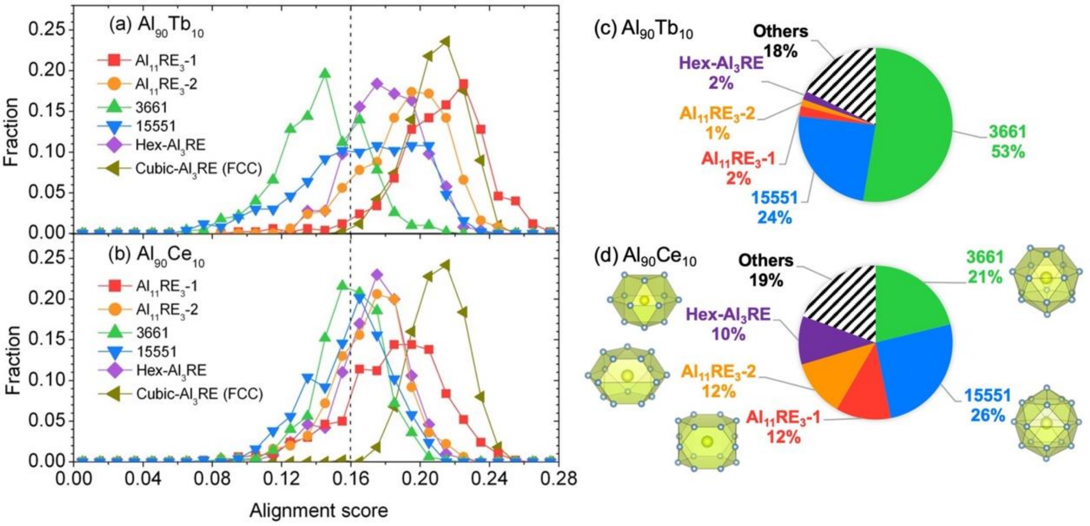


Fig. 3. The distributions of alignment scores against the relevant competing SRO templates for RE-centered clusters in the glassy (a) $Al_{90}Tb_{10}$ and (b) $Al_{90}Ce_{10}$ sample at T=300K. (c) and (d) The fractions of these RE-centered SROs in $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$, respectively. The corresponding structures of SRO templates are plotted. It shows ‘3661’ and ‘15551’ are the dominant SROs both in $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ glass samples, while only $Al_{90}Ce_{10}$ system have considerable SRO motifs as those in $Al_{11}Ce_3$ and Hex-Al_3Ce crystal.

By overlapping the center atoms of the 70-atom RE-centered superclusters belonged to the same template motif while keeping orientations of the clusters in their best alignment positions, we can quantify the degrees of not only SRO but also the MRO of the four RE-centered motifs in the $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ glass samples as shown in Fig. 4. Gaussian smearing [23] is applied to the atomic distribution and the intensity of the atomic density after the Gaussian smearing has been normalized (i.e., divided) by the number of clusters used in the overlapping, so that the plots show the averaged structural order of the four types of dominant motifs. As shown in Fig. 4(a)-(b) and (e)-(f), the ‘3661’ and ‘15551’ SRO extends outward to form Bergman-type MRO in the second and third shell of the superclusters. We can also see that both the first shell SRO and the MRO in the 2nd and 3rd cluster shells in the ‘3661’ and ‘15551’ motifs are very strong in both the $Al_{90}Tb_{10}$ and the $Al_{90}Ce_{10}$ glass samples. However, the MRO of the ‘3661’ motif is stronger the $Al_{90}Tb_{10}$ glass sample than in the $Al_{90}Ce_{10}$ glass sample, especially in the upper region of the superclusters as indicated by the red ovals in Fig. 4 (a) and (e). On the other hand, both the SRO and MRO of the $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 motifs are much stronger in the $Al_{90}Ce_{10}$ than in the $Al_{90}Tb_{10}$ glass samples, as one can see from the comparison in Fig. 4 (c)-(d) and (g)-(h) respectively. However, it is also interesting to note that the second and third shells of $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 motifs do not follow the packing motif in the crystalline $Al_{11}Ce_3$ structure, rather than packed in a closest packing pattern, i.e., the outer atoms fill the hollow sites among the inner atoms. Even the SRO packing of the first shell atoms in the superclusters has extra atoms capping to the $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 template motifs.

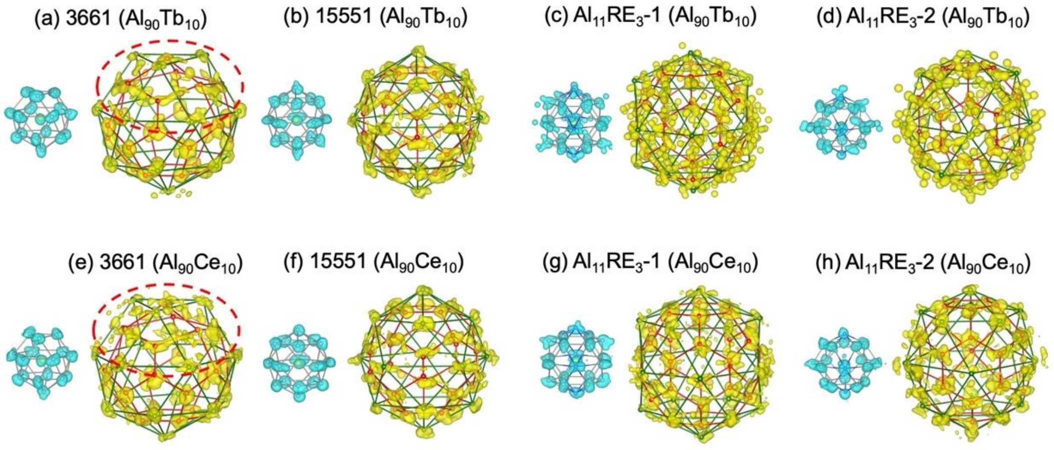


Fig. 4. The iso-surface of atomic distribution density (8×10^{-6}Å^{-3}) calculated by Gaussian smearing method for (a)(e) ‘3661’, (b)(f) ‘15551’, (c)(g) $Al_{11}RE_3$-1, and (d)(h) $Al_{11}RE_3$-2 superclusters (the first shell is plotted with blue and the second and third shell are in yellow) in glassy $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ sample, respectively. The average atomic structures are indicated by the balls and stickers in the plots. Here the stick frames outline either the SRO templates in the first shell or the averaged MRO structures in 2nd and 3rd shells. Note that extra atoms in the first shell go beyond the $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 SRO templates in (c), (g), (d) and (h).

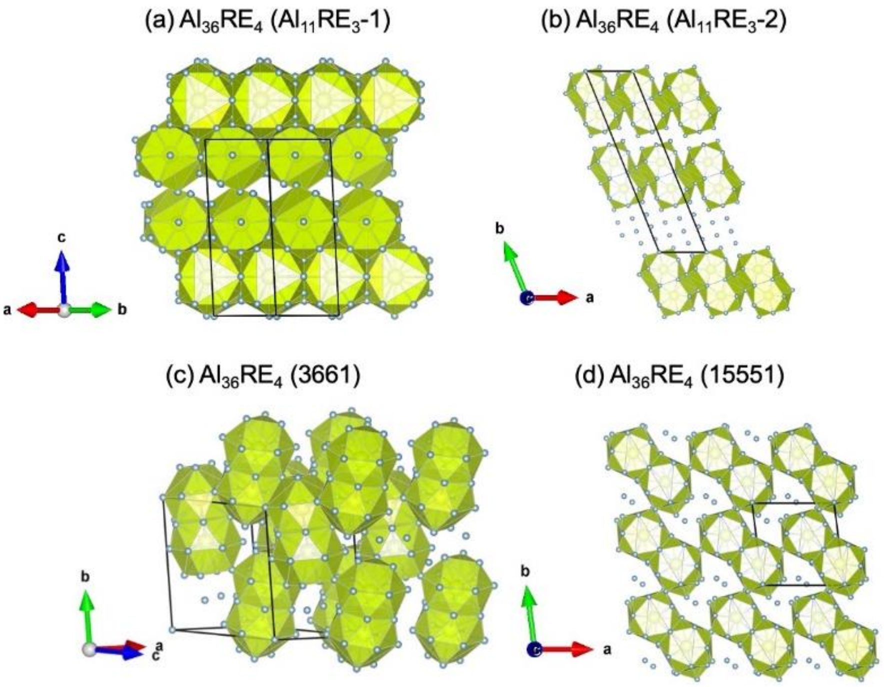
3.2 Structure-energy landscapes

The SRO and MRO analysis suggest that while the ‘3661’ and ‘15551’ orders are more favored in $Al_{90}Tb_{10}$ glass, $Al_{90}Ce_{10}$ glass exhibits stronger tendency in forming $Al_{11}Ce_3$-like structures. In order to see whether the different SRO and MRO in $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ glasses are driven by the different thermodynamic stabilities in the two systems, we investigate possible competing stable and metastable crystalline phases in these two systems in the vicinity of $Al_{90}RE_{10}$ composition, and correlate the structure-energy landscape of these stable and meta-stable crystalline structures with the dominant motifs found in the glass samples.

There are several known stable binary phases in these two systems. These known structures include $Al_{11}Ce_3$, Al_3Ce, Al_2Ce for Al-Ce binaries and Al_3Tb, Al_2Tb, AlTb, $AlTb_2$ for Al-Tb binaries. Using the formation energies of these known phases, the convex hulls for these two systems are shown in Fig. S3 in the Supplementary Materials [31]. Our previous studies on Al-Sm [40] and Al-Tb systems [27] also identify a number of low-energy metastable Al-RE phases in the vicinity of $Al_{90}RE_{10}$ composition. These phases include $Al_{17}Tb_2$, $Al_{120}Tb_{22}$ ('big cubic phase'), α-Al_4Tb, β-Al_4Tb, and $Al_{11}Tb_3$. By placing Tb or Ce atoms at the RE positions in these structures, we optimize the structures with both *ab initio* and ANN-ML potential calculations. The formation energies for these structures obtained from our *ab initio* and ANN-ML potentials calculations are also shown in Fig. S3 labeling with the cross symbols. We can see that the formation energies of these stable and meta-stable structures predicted from *ab initio* and ANN-ML potential calculations agree well with each other, indicating the ANN-ML potentials are accurate also in describing the energetic stability of crystalline phases in these two systems.

Based on the accurate ANN-ML potential model, we are able to perform efficient and reliable AGA [34, 35] search, as described in the section II, to explore additional competing meta-stable structures around the $Al_{90}RE_{10}$ composition in these two systems. The unit cells used in our GA search contain 39 to 41 atoms with composition of $Al_{35}RE_4$, $Al_{36}RE_4$ and $Al_{37}RE_4$ respectively. The size of the structure pool during the AGA search is maintained at the level of 200 structures. The volume and shape of the unit cells are allowed to vary but the change in the length of each lattice constant does not exceed ± 20% of the length of a cube of the same volume at each GA generation. The motifs of the crystal structures obtained by the AGA search are classified using the cluster alignment method described in section II. The value of alignment score cutoff 0.1 is used to assign the types of crystals obtained from GA searches.

In Fig. 5, we show the lowest-energy meta-stable structures built by each motif from our AGA search with $Al_{36}RE_4$ unit cell. It is interesting to note that for each of the 4 motifs, the lowest-energy structures in $Al_{36}Tb_4$ and $Al_{36}Ce_4$ are the same. The corresponding lowest-energy structures for $Al_{35}RE_4$ and $Al_{37}RE_4$ unit cells are also plotted in Fig. S4 and S5 in the Supplementary Materials [31]. All the lowest-energy structures in POSCAR format are given in the Supplementary Materials [31]. The formation energies of these lowest-energy meta-stable structures with respect to the convex hull from *ab initio* and our ANN-ML potential calculations are also plotted in Fig. S3. We can see that the formation energies of these structures are very close to the corresponding convex hull (within about 80 meV/atom of the convex hull). The formation energies obtained from our ANN-ML potentials are also in very good agreement with those from *ab initio* calculations.


(a) $Al_{36}RE_4$ ($Al_{11}RE_3$-1)

(b) $Al_{36}RE_4$ ($Al_{11}RE_3$-2)

(c) $Al_{36}RE_4$ (3661)

(d) $Al_{36}RE_4$ (15551)

Fig. 5. Lowest-energy metastable crystal structures with different building blocks as indicated for $Al_{36}RE_4$ (RE=Tb, Ce) obtained from our AGA search. The building blocks in these crystals are (a) $Al_{11}RE_3$-1, (b) $Al_{11}RE_3$-2, (c) '3661' and (d) '15551' clusters, respectively. Noted that the lowest-energy metastable crystal structures are the same for both $Al_{36}Tb_4$ and $Al_{36}Ce_4$.

In order to gain more comprehensive insights into the structure-energy landscape difference in the two systems, we collect the low-energy meta-stable structures (within 100 meV/atom above the convex hull) belong to the four motifs (i.e., '3661', '15551', $Al_{11}RE_3$-1, and $Al_{11}RE_3$-2) respectively from our AGA search, and then plot their formation energy distributions as shown in Fig. 6. For Al-Tb crystals, we have 1188, 43, 527, and 27 structures in the $Al_{11}RE_3$-1, $Al_{11}RE_3$-2, '3661' and '15551' motifs respectively, while these numbers in Al-Ce crystals are 1790, 17, 464, and 29, respectively. The energies in the Fig. 6 are from our ANN-ML calculations. We can see that for formation energies less than 60 meV/atom above the convex hull, only the structures with $Al_{11}RE_3$-1 motifs in Al-Ce system are found. The formation energies of the structures composed of either '3661' or '15551' motifs in both Al-Tb and Al-Ce systems are all higher than 60 meV/atom. Moreover, in low-energy range (< 70 meV/atom), the number of the $Al_{11}RE_3$-1 crystal in Al-Ce is 1067 while the '3661' and '15551' crystals in Al-Tb and Al-Ce systems are only 173 and 133 respectively. These results suggests that crystal structures with $Al_{11}RE_3$ motif are energetically much more favorable than those with '3661' or '15551' motifs in Al-Ce system. These results also strongly correlate with the SRO in the glass samples where there is significant fraction (24%) of RE-centered clusters in $Al_{90}Ce_{10}$ glass are classified as either $Al_{11}RE_3$-1 or $Al_{11}RE_3$-2, while '3661' and '15551' motifs are predominated in $Al_{90}Tb_{10}$ glass sample.

In addition, Table. I show the averaged bonding lengths of SROs in $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ glass as compared to that of the various competing crystalline phases at T=300K. The Tb-Al bond length of the major glass forming SRO motifs in the glass is very different (7.0%) from that of the nearby stable Al_3Tb phase, which would make the crystallization of Al_3Tb phase more difficult. The Ce-Al bond length of the major ('3661' and '15551') SRO motifs in the glass is very similar to that of the nearby stable $Al_{11}Ce_3$ phase, which would make the crystallization easier. Although the Ce-Al bond length of the $Al_{11}Ce_3$ motifs in the glass are about 4.7% larger than that in stable $Al_{11}Ce_3$ crystal, their shapes are similar which may also make the transformation easier. Therefore, thermodynamic stabilities of various competing metastable phase in the glass formation composition region have significant influence in the glass formation and local structure orders in the glass.

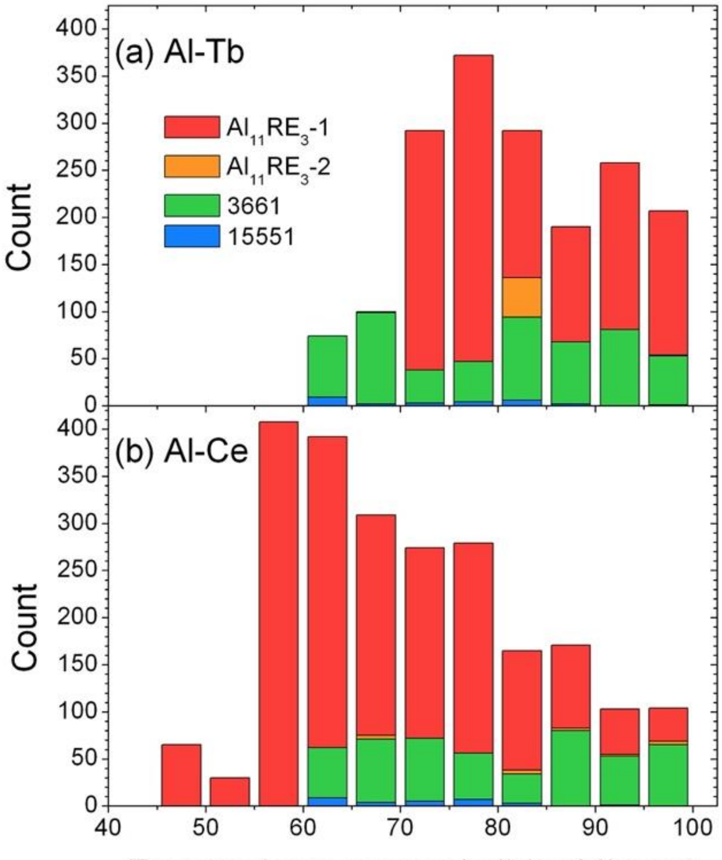


Fig. 6. The distributions of formation energies respect to convex hull for the low-energy metastable structures obtained from our AGA search. The structures are classified according to the 4 SRO motifs. The results for Al-Tb are shown in the top panel while those for Al-Ce are in the bottom panel.

Table. I. The averaged bonding lengths of the '3661' and '15551' motifs as well as the $Al_{11}RE_3$-1/2) motifs in $Al_{90}RE_{10}$ glass as compared to that of the various competing crystalline phases at T=300K. Here, the numbers in the brackets are the bonding length differences between $Al_{90}RE_{10}$ glass and the nearby stable crystalline phase. It can be seen that the Tb-Al bond length difference between ’3661’+’15551’ motifs in $Al_{90}Tb_{10}$ and nearby stable Hex-Al_3Tb crystal is about 0.22Å (7.0%), while the Ce-Al bond length difference between ‘3661’+’15551’ motifs in $Al_{90}Ce_{10}$ and nearby stable $Al_{11}Ce_3$ crystal is about only 0.02 Å (0.59%).

	‘3661’+’15551’ motifs in $Al_{90}RE_{10}$ (Å)	$Al_{11}RE_3$-1/2 motifs in $Al_{90}RE_{10}$ (Å)	$Al_{11}RE_3$ (Å)	Hex-Al_3RE (Å)	Cubic-Al_3RE (Å)
Al-Tb	3.324 (+0.216)	3.505 (+0.397)	3.328	3.108	3.034
Al-Ce	3.418 (+0.020)	3.558 (+0.160)	3.398	3.297	3.269

3.3 Crystal growth kinetics

Using the ANN-ML potentials, we also performed MD simulations to study the growth of $Al_{11}RE_3$ and big-cubic $Al_{120}RE_{22}$ phases at the interface with $Al_{90}RE_{10}$ liquids where RE are Tb and Ce respectively. While $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 SRO motifs are the building blocks of $Al_{11}RE_3$ crystalline phase, the RE-centered SRO motifs in big-cubic $Al_{120}RE_{22}$ phase are ‘3661’ and ‘16661’ respectively. The ‘16661’ motif is not the same as but with very similar structure topology as ‘15551’ clusters observed in the $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ glasses. Therefore, comparing the growth behavior and speed of these two crystalline phases from solid/liquid interface would provide useful insights into the phase selection kinetics in Al-Tb system verse Al-Ce system.

The temperature in the simulations of crystal growth is 700K for both Al-Tb and Al-Ce system. We chose 700K for the crystal growth simulation because it is just slightly above the T_g in the two systems. From Fig. 1 the glass transition temperature of $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ estimated from the intersection point of the two white slope lines in the potential energy is very close to each other around 600K. According to the phase diagram in the supplemental material [31], the melting temperatures of Al-Tb and Al-Ce at 90-10 composition are also very close to each other. Therefore, the degree of undercooling at 700K ($T/T_m \approx 0.5$) should be similar in the two systems. As discussed above, although the melting and glass transition temperatures are similar in these two systems, the change in the potential energy with temperature in the undercooled liquid region is more rapid in $Al_{90}Ce_{10}$ than in $Al_{90}Tb_{10}$, suggesting $Al_{90}Ce_{10}$ should have better crystallization ability. Therefore, the main purpose of crystal growth simulations is to estimate the crystal growth speed at the temperature just above the T_g in order to compare the GFA in the two systems.

To prepare an initial solid/liquid interface, we first perform MD simulation for the crystalline structure at T=700K for 10 ps. The MD simulation box is in a rectangular shape and contains 2016 atoms for the $Al_{11}RE_3$ phase and 2272 atoms for the big-cubic-$Al_{120}RE_{22}$ phase respectively. Periodic boundary conditions are applied in the three directions. Then, the atoms in the left half of the MD simulation box are kept fixed at their instant positions at 700 K, while the atoms in the right half of the MD simulation box are heated to 2000K for 1 ns to ensure the structure in this half of the box is completely melted into a liquid state. During the MD simulation at 2000K, some of the Al atoms are randomly selected to switch to RE atoms in order to have the liquid composition at $Al_{90}RE_{10}$. Finally, the liquid is cooled down to T=700K with a cooling rate of 10^{13} K/s. Thus, an initial solid/liquid interface is prepared at T=700K where the solid part is in the given crystalline structure ($Al_{11}RE_3$ or $Al_{120}RE_{22}$) while the liquid part have a composition of $Al_{90}RE_{10}$. We denote this initial interface structure as t=0 configuration. Starting from the t=0 configuration, MD simulations are performed at a constant temperature of 700K and constant P=0 to study the growth of the crystal.

In Fig. 7, we compare the growth speed of the $Al_{11}RE_3$ and $Al_{120}RE_{22}$ phases for RE= Tb or Ce respectively. Only the growths along the (001) direction are shown in Fig. 7. Growth behavior along other directions is similar to that along the (001) directions, as shown by Fig. S6 in the Supplementary Materials [31]. The short movies of crystal growth (t=0-200ns) are also given in the Supplementary Materials [31]. From the plots in Fig 7, we can see that the growth speed of the $Al_{11}RE_3$ phase in Al-Ce system is slightly faster than the growth in Al-Tb system. At 60 ns, crystallization in the Al-Ce system is essential completed which it takes about 130 ns for Al-Tb system to reach the same degree of crystallization. Because RE concentration is about 21.4% in the $Al_{11}RE_3$ phase, the crystallization of $Al_{11}RE_3$ phase in the MD simulation will saturate when all the RE atoms in the $Al_{90}RE_{10}$ liquid have been incorporated into the crystalline phase, thus leave a pure Al slab in the MD simulation box as one seen from Fig. 7. For the big-cubic $Al_{120}RE_{22}$ phase, the crystal growth in much faster in Al-Ce than in Al-Tb system. Crystallization in the whole MD simulations cell has already reached before 60 ns in Al-Ce system, while the crystal growth in Al-Tb system is still not quite completed even at the simulation time of 130 ns as one can see from the plots in Fig. 7.

In order to confirm the results of faster crystal growth rate for Al-Ce, we also simulated the crystal growth at T=800K for Al-Ce, where the degree of undercooling is smaller than that of 700K. As shown in Fig. S7 of the supplemental material [31], the crystal growth on Al-Ce at 800K is still faster than Al-Tb system at 700K.

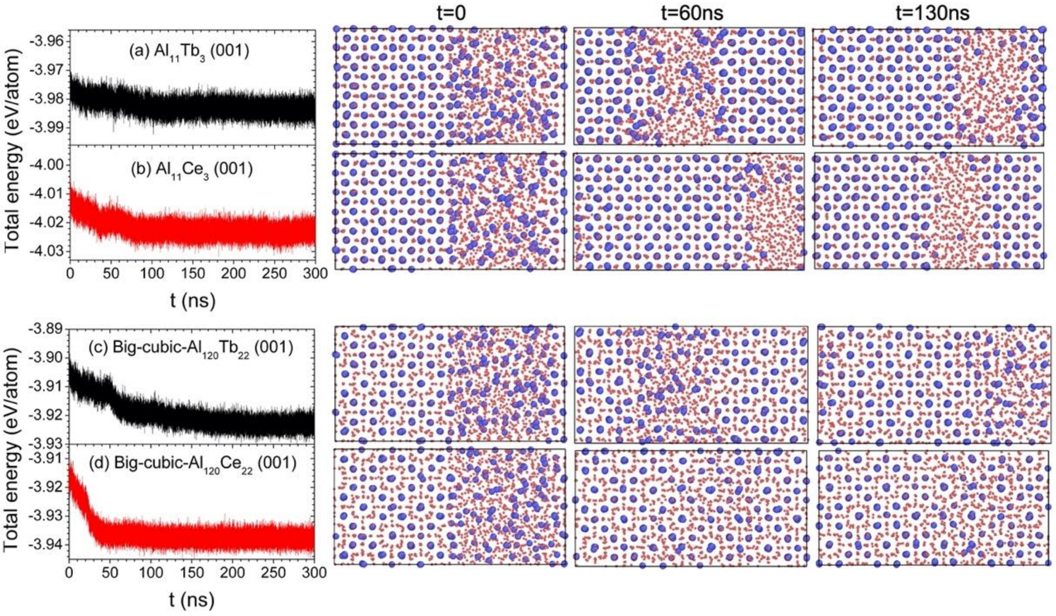


Fig. 7. The change in the total energy during the annealing process of (a)(b) $Al_{11}RE_3$ and (c)(d) big-cubic-$Al_{120}RE_{22}$ at T=700K. The solid/liquid interface are along the

directions of (001), where the liquid is $Al_{90}RE_{10}$. The snapshots at initial t=0, annealing time t=60 ns and t=130 ns are also shown in the right. The blue and red dots represent the RE and Al atoms respectively.

We found that the difference in the crystal growth speed of the two system is correlated with the diffusivity of the RE atoms in the undercooled liquid. The mean square displacements (MSD) as the function of time for Al atoms and RE atoms in the undercooled liquid at 700K are shown in Fig. 8. Using the Einstein formula, the diffusion constants of Al and Tb atoms in the $Al_{90}Tb_{10}$ undercooled liquid are 0.44×10^{-5} cm^2/s and 0.032×10^{-5} cm^2/s respectively, while the diffusion constants of Al and Ce atoms in the $Al_{90}Ce_{10}$ undercooled liquid are 0.35×10^{-5} cm^2/s and 0.055×10^{-5} cm^2/s. We can see that although Al atoms moves faster in $Al_{90}Tb_{10}$ undercooled liquid at 700K, Tb atoms moves considerably slower than Ce atoms. The limiting factor for the growth of the crystalline phases is the mobility of the RE atoms. These results indicate that the crystallization kinetics for Al-Tb is slower than that for Al-Ce crystals, which again implies that the Al-Ce system has strong tendency of forming crystal than Al-Tb system.

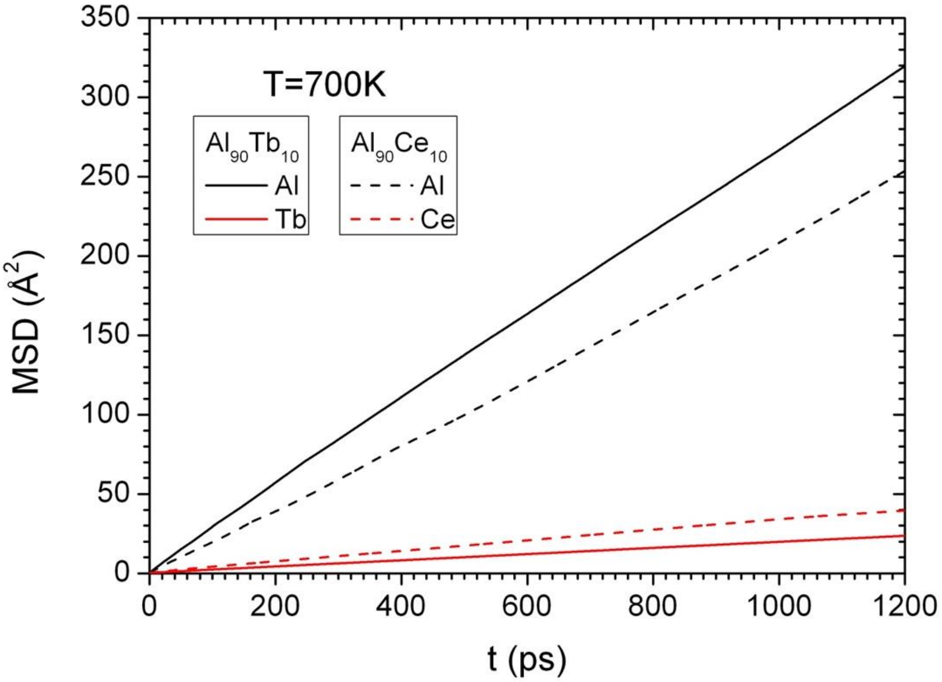


Fig. 8. The mean square displacements (MSD) as the function of time for Al atoms and RE atoms in the undercooled $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ liquids at 700K.

Based on Wilson-Frenkel theory [41, 42], the crystal growth rate is proportional to diffusion coefficient. Although the diffusion constant of Ce is slightly larger than that of Tb as shown in Fig. 8, the difference would not be sufficient to explain the crystal growth difference in these two systems. In order to further understand the difference of crystal growth between Al-Tb and Al-Ce system, we calculated the enthalpy difference between liquid and crystal (the latent heat) for $Al_{11}RE_3$ and Big-cubic-$Al_{120}RE_{22}$. A 9x4x3 $Al_{11}RE_3$ (total 3024 atoms) and a 3x3x3 Big-cubic-$Al_{120}RE_{22}$ (total 3834 atoms) supercell samples are generated for MD simulation, respectively. The samples are first equilibrated with NPT ensemble at T=300K for 500ps and then heated to 2000K in a rate of 10^{12} K/s. During the heating process where the samples are not melted, snapshots at T=700 to 1100K with 100K interval are taken as initial structures to perform MD simulations at the given temperature for 25ps to obtain the averaged enthalpy of crystal at this temperature. At high temperature T=2000K, the samples are melted into liquid and then equilibrated for 500ps. The liquid samples are then cooled in a rate of 10^{12} K/s to T=700K. The snapshots of undercooled liquid samples from T=700 to 1100K with 100K interval are also selected as initial configuration for 25ps MD simulation to obtained the averaged enthalpy of the liquid at the given temperature.

The enthalpy difference between the liquid and crystal scaled by thermal energy for $Al_{11}RE_3$ and Big-cubic-$Al_{120}RE_{22}$ at temperatures is shown in Fig. 9. It shows that for Al-Tb crystals, the driving force of $Al_{11}Tb_3$ and Big-cubic-$Al_{120}Tb_{22}$ is close to each other. Therefore, these two crystalline phases both have no competitive advantages in crystal growth at solid/liquid interface, and leads to frustration and inefficient crystallization. However, the crystallization driving force of $Al_{11}Ce_3$ is about 1.5 time as large as that of the other crystalline phases. Thus, the crystallization of $Al_{11}Ce_3$ should be more efficient (see our crystal growth simulations in Fig. 7). Although the driving force of Big-cubic-$Al_{120}Ce_{22}$ is smaller and close to that of Al-Tb system, the crystallization is still more efficient than that in Al-Tb system since the chemical order of the SRO clusters in $Al_{90}Ce_{10}$ liquid is closer to that in crystal structures. On the contrary, the chemical frustration in Al-Tb system could hinder the crystallization and lead to better GFA in Al-Tb system.

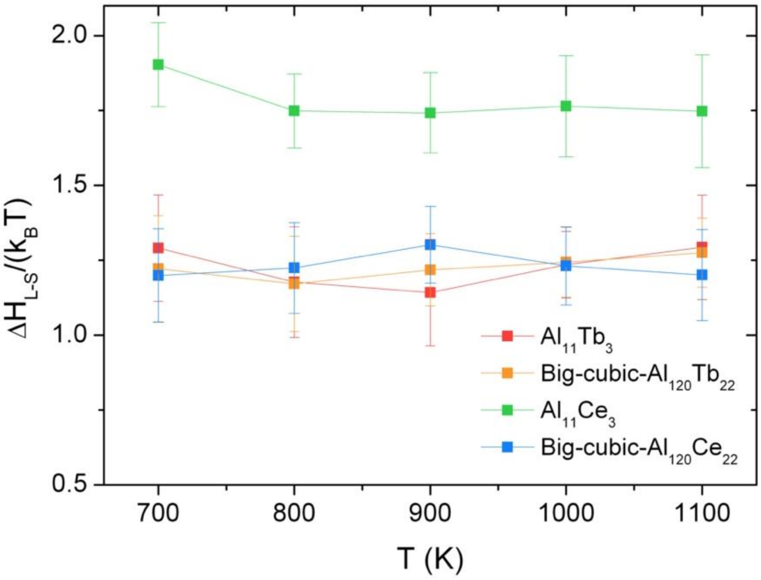


Fig. 9. The latent heats of $Al_{11}RE_3$ and Big-cubic-$Al_{120}RE_{22}$ scaled by the thermal energy.

The crystal growth difference between Al-Tb and Al-Ce can also be attributed to the different chemical orders in RE-centered SRO clusters in the undercooled liquids. To examine the chemical order in the SRO clusters, we plotted the distributions of the atoms in the first shell of the RE-centered SRO clusters after the alignment, as shown in Fig. 10(a). The structures and chemical orders of some competing crystalline Al-RE phases are also plotted in Fig. 10(b)-(d) for references. As shown in Fig. 10(a), the RE atoms are nearly uniformly distributed around all the vertex of the first shell in SRO clusters, except in ‘3661’ SROs the RE atoms prefer occupying the bottom vertex indicated as ‘1’ in ‘3661’. This feature is also observed in Big-cubic-$Al_{122}RE_{22}$ crystal where the ’3661’ SRO is the building block of the structure. The averaged composition of RE atoms in the first shell of SROs are also calculated for $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$, where atoms at vertex ‘1’ of ’3661’ SRO are excluded from statistical analysis. The results show the RE composition in $Al_{90}Tb_{10}$ SRO clusters is larger than that for $Al_{90}Ce_{10}$. For the glass-forming ‘3661’ and ‘15551’ SROs (which are the majorities in $Al_{90}RE_{10}$), RE composition is ~8% and ~5% respectively in $Al_{90}Tb_{10}$ glass as compared to ~3% in $Al_{90}Ce_{10}$ glass. Even for crystal-SRO clusters in the glass phase, the RE composition at the first shell is larger in $Al_{90}Tb_{10}$ than in $Al_{90}Ce_{10}$. Note that all the atoms are Al in the first shell of RE-centered motifs in competing crystalline phases (noted that vertex ‘1’ is excluded from the statistics), as shown in Fig. 10(b)-(d).

Therefore, in comparison with $Al_{90}Tb_{10}$ glass, the SRO clusters in $Al_{90}Ce_{10}$ glass is not only structural more similar to the crystal phases but also with chemical order closer to that of the crystal phases. Less deviation of structural and chemical orders from competing crystalline phases for the SROs in $Al_{90}Ce_{10}$ undercooled liquid can reduce both the geometrical and chemical frustration against crystallization [43-47]. Because chemical order difference in the liquid and crystalline phases determines the interface energy which dominates the crystallization [43], the primary barrier to crystallization is lowered for glass SROs in $Al_{90}Ce_{10}$. Thus, besides the fact that there is noticeable fraction of crystal SROs in $Al_{90}Ce_{10}$, the less deviation of chemical order from competing crystals for all the SROs in $Al_{90}Ce_{10}$ would also enhance the crystallization and weaken the GFA in $Al_{90}Ce_{10}$ compared to the case of $Al_{90}Tb_{10}$.

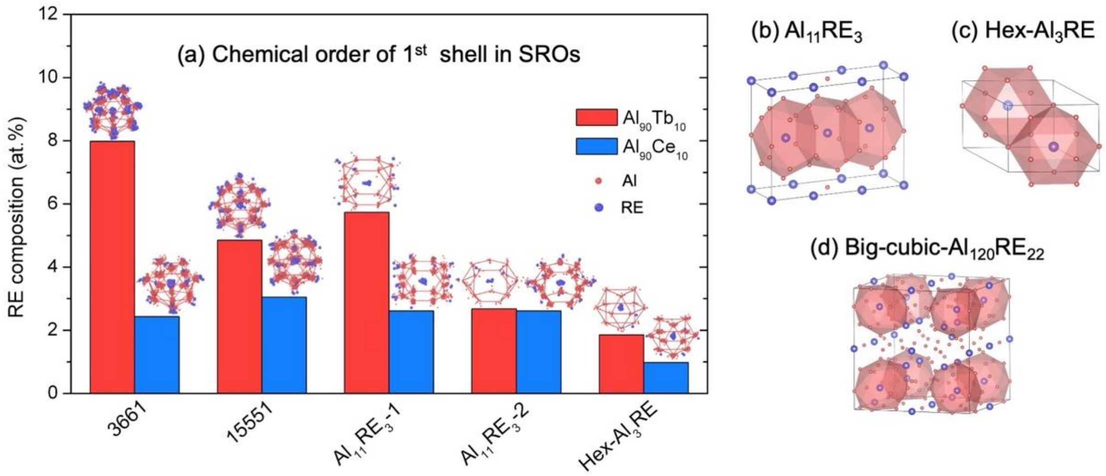


Fig. 10. (a) The RE composition of the first shell in RE-centered SROs for $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$. The corresponding atoms distributions of SROs after aligned against templates are also plotted in the figure (a). The structures of some competing crystalline phases in $Al_{90}RE_{10}$ liquid are plotted in (b)-(d), where the red polyhedral with Al atoms vertex are the RE-centered SRO clusters as building blocks in crystals. (b) $Al_{11}RE_3$ (c) Hex-Al_3RE (d) Big-cubic-$Al_{120}RE_{22}$, where the red polyhedral are '3661' clusters.

IV. Summary

Using the accurate and transferable interatomic potentials based on ANN-ML, we have performed reliable MD simulations and cluster alignment analysis to compare the similarity and difference of SRO and MRO in $Al_{90}Tb_{10}$ and $Al_{90}Ce_{10}$ metallic glasses. Our results show that although the glass forming Ce-centered '3661' and '15551' clusters are also the major motifs in $Al_{90}Ce_{10}$ glass, the percentage of such motifs (47%) is substantially smaller than that in $Al_{90}Tb_{10}$ glass which encompasses 77% of the Tb-centered clusters. On the other hand, there are significant amount of $Al_{11}Ce_3$ (24% of Ce-centered clusters) and Al_3Ce (10% of Ce-centered cluster) crystalline SRO clusters in $Al_{90}Ce_{10}$ glass, while the crystalline motifs are negligible in $Al_{90}Tb_{10}$ glass. The SRO and MRO differences in the two glasses can be attributed to the preference of different crystalline phases in Al-Ce system vs Al-Tb system. We explored possible competing stable and metastable crystalline phases in these two systems in the vicinity of $Al_{90}RE_{10}$ composition using AGA method and the ANN-ML interatomic potentials. A significant number of metastable crystalline structures with formation energy within 100 meV/atom above the corresponding convex hulls have been discovered from our AGA search. These low-energy structures can be classified into the four major structure motifs, i.e, '3661', '15551', $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2, which are observed in the glass samples. We found that the structures with $Al_{11}RE_3$-1 and $Al_{11}RE_3$-2 motifs are energetically much favorable in Al-Ce system, while those with '3661' and '15551' are preferred in Al-Tb system. These structure-energy landscapes are strongly correlated with the SRO and MRO found in the two glasses, suggesting thermodynamic driving forces play an important role in the phase selections. We also found that due to the difference in the atomic size and interatomic interaction between Al-Ce and Al-Tb systems, the average Tb-Al bond lengths in the glasses is more than 7.0% larger than that in the nearby stable crystalline Al_3Tb phase. Such big RE-Al bond length difference would also make the crystallization in Al-Tb system harder. Crystallization simulations using MD based on a solid/liquid interface model also show that the speed of crystal growth in Al-Ce system is about 2-3 time faster than that in the Al-Tb system at a similar undercooling condition. By comparing atomic diffusion constants in the undercooled liquids and the crystal-liquid latent heats of several competing crystalline structures, we found that the crystallization driving forces are mainly from the enthalpy difference between liquid and crystal structures. Moreover, we showed that less deviation of chemical order from competing crystals for all the SROs in $Al_{90}Ce_{10}$ would also enhance the crystallization and weaken the GFA in $Al_{90}Ce_{10}$ as compared to the case of $Al_{90}Tb_{10}$. The results from our studies are consistent with experimental observation that Al-Tb exhibit much better GFA than Al-Ce, and crystalline phase formations are more robust in the Al-Ce system. Our studies provide in-depth insights into the phase selection and formation difference between Al-LRE and Al-HRE metallic alloys.

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Data availability

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

Conflict of interest

The authors declare no competing interests.

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