On the Optimal Glass-Forming Composition of Al-Co-Y Amorphous Alloys

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Al-Co-Y alloys were suction cast under identical conditions into a wedge-shaped copper mold for investigating the compositional dependence of glass-forming ability (GFA). The optimal glass-forming composition was determined to be $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$, whose Al content is evidently larger than the findings of the optimal glass formers in other Al-TM-Y alloy systems. A reason is that Co-centered clusters tend to share vertexes and edges rather than faces with the surrounding clusters in Al-Co-Y amorphous alloy, and more Al atoms are required. When $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ amorphous alloy and its neighbors were isochronally annealed in a differential scanning calorimeter, no glass transition could be observed and the primary crystallization phase was invariably fcc-Al. [doi:10.2320/matertrans.M2018177]

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1. Introduction

Since the successful preparation of ductile Al-based amorphous alloys by Inoue and He *et al.* in 1988,^{1,2)} Al-based amorphous alloys have been drawing worldwide attention due to their high specific strength and good ductility.³⁾ Compared with other amorphous alloys, Al-based amorphous alloys show some distinctive features. For instance, the content of Al atoms exceeds 80 at%, being extraordinarily high even as the main element;³⁾ the best glass-forming compositions are far off eutectic points for many binary and ternary Al-based alloy systems; and the glass-forming ability (GFA) is very sensitive to the alloy composition.^{4–10)} Following the empirical rule that similar element substitution often have a positive effect on GFA,¹¹⁾ Wang *et al.*^{12–15)} prepared Al-based bulk amorphous alloys of 1 mm diameter from $\text{Al}_{86}\text{Ni}_6\text{Y}_{4.5}\text{Co}_2\text{La}_{1.5}$, $\text{Al}_{86}\text{Ni}_7\text{Y}_5\text{Co}_1\text{La}_1$ and $\text{Al}_{86}\text{Ni}_7\text{Y}_{4.5}\text{Co}_1\text{La}_{1.5}$ and 1.5 mm diameter from $\text{Al}_{86}\text{Ni}_{6.75}\text{Co}_{2.25}\text{Y}_{3.25}\text{La}_{1.75}$ and $\text{Al}_{86}\text{Ni}_6\text{Y}_{4.5}\text{Co}_2\text{La}_{1.5}$. Even so, the Al-based amorphous samples obtained are still much smaller than the Zr-, Pd-, Fe- and Cu-based amorphous alloys.^{16–18)}

The ability for an alloy to form a glass is equivalent to suppressing crystallization within the undercooled liquid. An alloy melt whose atoms are densely packed is known to be sluggish in crystallization kinetics. The efficient atomic packing (EAP) model was therefore proposed by Miracle *et al.*¹⁹⁾ to design amorphous alloys. For Al-based amorphous alloys, the solute atoms are exclusively surrounded by Al atoms owing to the strong chemical bonding between the Al and solute atoms⁴⁾ and the high content ratios of the Al to the solute. Sheng *et al.*⁴⁾ computer simulated the three-dimensional atomic configurations of numerous Al-based amorphous alloys, and gave the average coordination number of Al atoms for various solute atoms. With the above advances, Ma *et al.*²⁰⁾ proposed a cluster stability line intersection (CSLI) method to search the composition with the best GFA in ternary Al-based alloys. Zhang *et al.*^{9,21)} later modified this method by taking the effect of interaction between different elements on atomic radii into account. The

optimal glass-forming compositions of several Al-Ni-RE ternary systems were predicted to be $\text{Al}_{85.7}\text{Ni}_{9.5}\text{La}_{4.8}$ and $\text{Al}_{85.5}\text{Ni}_{9.5}\text{RE}_5$ (RE = Y, Ce, Gd, Dy), in good agreement with the experimental results.⁹⁾

Co and Ni are similar to each other in the physical and chemical properties. The optimal glass-forming composition of Al-Co-Y calculated by the EAP model in combination with the CSLI method is $\text{Al}_{85.5}\text{Co}_{9.5}\text{Y}_{5.0}$ (the derivation is shown below). The atomic ratio among the three elements is very close to the value of the optimal glass-forming composition of Al-Ni-Y. On the other hand, the optimal glass former in Al-Co-RE was experimentally determined by Yang *et al.*²²⁾ to be $\text{Al}_{89}\text{Co}_6\text{RE}_5$ and $\text{Al}_{90}\text{Co}_6\text{RE}_4$, and by Dong *et al.*²³⁾ to be $\text{Al}_{88}\text{Co}_5\text{Y}_7$. In the present work, we first examined the GFA of Al-Co-Y alloys in a wide composition range by carefully controlling the preparation condition. Then theoretical analyses were made to unravel what results in the high Al content in the optimal glass-forming compositions of Al-Ni-Y.

2. Experimental Procedure

The Al-Co-Y alloys investigated in this work are shown in Fig. 1. The alloy compositions distribute in two separate regions: one around the optimal composition predicted by the CSLI method, where two lines represent the content ratios of Al/Y and Al/Co; the other covering the compositions of $\text{Al}_{88}\text{Co}_5\text{Y}_7$, $\text{Al}_{89}\text{Co}_6\text{Y}_5$ and $\text{Al}_{90}\text{Co}_6\text{Y}_4$. Alloy ingots were prepared by arc melting the mixture of Al (99.999%), Co (99.99%) and Y (99.9%) in a water-cooled copper crucible under the Ti-gettered argon atmosphere. All the ingots were melted six times to ensure their compositional homogenization. The oxides on the as-prepared ingots were eliminated before the ingots were cut into small pieces weighing about 2 grams. Each piece of the alloy ingots was remelted and suction cast into a copper mould with an included angle of 5 degrees and a width of 10 mm to form a wedge-shaped sample in the arc melting furnace. The arc current and melting duration were controlled to be the same for all the casting operations. At least two wedge-shaped samples were prepared for each alloy composition to make

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Fig. 1 Al–Co–Y compositions investigated and corresponding critical thickness t_c (μm) of amorphous region in the wedge-shaped Al–Co–Y samples, where two straight lines ($\text{Al}/\text{Y} = 17$ and $\text{Al}/\text{Co} = 9$) represent the cluster stability lines.

sure that the results would be reliable. The as-cast wedge-shaped samples were then cut into two parts using a diamond saw along the central section and perpendicular to the inclined planes. After that, the samples were carefully mounted, polished, and etched using the Keller's reagent. The maximum thickness of the amorphous region was measured using an optical microscope as an indicator of GFA. To check the amorphous nature of the tip region, a JOEL JEM-2100F high-resolution transmission electron microscope (HRTEM) was used to examine the foils prepared by twin-jet polishing in a solution of 25% HNO_3 and 75% methanol at 233 K.

To study the crystallization behavior of the Al–Co–Y amorphous alloys, ribbons of 20–30 μm thickness and 5 mm width were spun at a copper-wheel surface velocity of 40 m/s in a vacuum single roller melt-spinning apparatus. After their amorphous nature was identified by X-ray diffraction (XRD)

with Cu $K\alpha$ radiation, the ribbons were annealed in a Perkin Elmer Pyris Diamond differential scanning calorimeter (DSC) at a heating rate of 20 K/min under a flow of highly-purified argon atmosphere to detect the crystallization. The ribbons annealed to the end of the first crystallization peak were also examined by XRD to determine the primary phase.

3. Results

3.1 GFA

An optical micrograph of the cross-section of an $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ wedge-shaped sample is shown in Fig. 2. Two distinct regions representing the amorphous and crystalline parts respectively are observed. The HRTEM image and the selected area electron diffraction (SAED) pattern of the amorphous part are shown in Fig. 3. No visible crystals in the HRTEM image and the broad halo in the SAED pattern confirm the fully amorphous nature of the sample tip. Thus, it was reliable to use the average maximum thickness (critical thickness) t_c of the amorphous parts produced from the same alloy as the GFA indicator of the alloy.

All the t_c of the investigated Al–Co–Y alloys is marked in Fig. 1. The error of the measurements is within 10 μm . It can be seen that t_c is very sensitive to the alloy composition. Among all the alloys investigated, $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ exhibits the best GFA, and its t_c is up to 405 μm . It is clear that $\text{Al}_{89}\text{Co}_6\text{Y}_5$, $\text{Al}_{90}\text{Co}_6\text{Y}_4$ and $\text{Al}_{88}\text{Co}_5\text{Y}_7$ that were declared by some researchers^{20,21)} to have the best GFA present far smaller t_c . With a no more than 120 μm t_c , the alloys around $\text{Al}_{85.5}\text{Co}_{9.5}\text{Y}_5$ exhibit worse GFA.

Fig. 2 Optical micrograph of a sectioned $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ wedge-shaped sample.

Fig. 3 (a) TEM bright-field image and SAED pattern of the amorphous part of an $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ wedge-shaped sample and (b) the corresponding HRTEM image.

Fig. 4 XRD patterns of Al-Co-Y ribbons: (a) as-prepared, (b) annealed up to the end of the first exothermic peak respectively at a rate of 20 K/min.

Fig. 5 DSC curves of the Al-Co-Y amorphous alloys at a heating rate of 20 K/min.

3.2 Crystallization behavior

Typical XRD patterns of the as-prepared Al-Co-Y ribbons are shown in Fig. 4(a). There are only broad peaks reflecting amorphous nature to be observed. The DSC curves of the ribbons at a heating rate of 20 K/min are shown in Fig. 5. No endothermic event took place before crystallization for all the Al-Co-Y amorphous alloys, indicating that no distinct glass transition was involved. Each DSC curve consists of at least two exothermic peaks. In general, the onset temperature of crystallization T_{x1} decreases with increasing Al content. The XRD patterns of the Al-Co-Y ribbons annealed to the end of the first crystallization peak are shown in Fig. 4(b).

Except for some alloys surrounding $\text{Al}_{85.5}\text{Co}_{9.5}\text{Y}_{5.0}$, the primary phase is composed of only fcc-Al for the other alloys.

4. Discussions

Metallic glass forms when crystallization does not occur during cooling of a molten metal to below the glass transition temperature T_g . Some researchers therefore take glass formation as a result of competition with crystallization during the cooling process.²⁴⁾ For the good glass formers, their compositions should be different from the composition of the primary crystallization phase so that the crystallization is easy to be hindered because of the necessary long-range solute diffusion. In general, more than one phases involve in the crystallization of a given alloy system, and obviously their compositions are different from each other. This is also the reason why the alloys with a near-eutectic composition have a good GFA.^{25–27)} Correspondingly, the optimal glass-forming composition is often located at the boundary separating different sorts of primary crystallization phases in the composition triangle.⁹⁾ In the present work, however, the DSC and XRD results indicate that all the Al-Co-Y amorphous alloys around $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ do not exhibit a glass transition during isochronal heat treatment, and the primary phase is constantly fcc-Al. That is to say, they all belong to the nanocrystalline alloys.²⁸⁾ Therefore, it seems that the composition of the optimal glass former in Al-Co-Y does not further deviate from the composition of the primary phase.

According to the X-ray absorption fine structure (XAFS) analysis results of local atomic structures of Al-TM-RE amorphous alloys (TM-transition metals such as Fe, Co and Ni, and RE-rare earth metals such as La, Ce and Y), the solute atoms are mostly surrounded by Al atoms.^{29,30)} The efficient atomic packing model proposed by Miracle *et al.*¹⁹⁾ can be used to describe the atomic arrangement in the good glass formers. Assuming that a solute atom is only surrounded by solvent atoms, there is the following relationship between the optimum coordination number N^T and R , the atomic radius ratio of the solute to solvent elements,¹⁹⁾

Fig. 6 Comparison of the TM-centered clusters in Al_3Ni (a) and Al_9Co_2 (b). The black, dark grey and light grey atoms are nickel, cobalt and aluminum, respectively. The white atoms with a cross are aluminum shared by two clusters. Common face occurs in Al_3Ni while common edge occurs in Al_9Co_2 .

$$N^T = \begin{cases} \frac{4\pi}{6 \arccos\{\sin(\pi/3)[1 - 1/(R+1)^2]^{1/2}\} - \pi} & \text{for } 0.225 \leq R \leq 0.414 \\ \frac{4\pi}{8 \arccos\{\sin(\pi/4)[1 - 1/(R+1)^2]^{1/2}\} - 2\pi} & \text{for } 0.414 \leq R \leq 0.902 \\ \frac{4\pi}{10 \arccos\{\sin(\pi/5)[1 - 1/(R+1)^2]^{1/2}\} - 3\pi} & \text{for } R \geq 0.902 \end{cases} \quad (1)$$

In view of the fact that most Al-based amorphous alloys have three basic constituent elements, Ma *et al.*²⁰⁾ developed the CSLI method to predict the optimum glass-forming compositions of Al-TM-RE metallic glasses. The expressions of the molar content of TM and RE given by them are as follows,

$$\begin{aligned} x_{\text{TM}} &= \frac{1}{N_{\text{TM}}^T \times (1 + 1/N_{\text{TM}}^T + 1/N_{\text{RE}}^T)} \\ x_{\text{RE}} &= \frac{1}{N_{\text{RE}}^T \times (1 + 1/N_{\text{TM}}^T + 1/N_{\text{RE}}^T)} \end{aligned} \quad (2)$$

where N_{TM}^T and N_{RE}^T are the optimum coordination numbers of TM and RE atoms, respectively. From the point of view of Wang *et al.*, the TM and RE solute atoms should be uniformly dispersed among the Al base atoms, and do not contact directly. Meanwhile, each Al atom needs to be shared by two neighboring solute-centered clusters. Considering that the Al-TM bond length is obviously smaller than the sum of the nominal atomic radii of Al and TM in Al-TM-RE amorphous alloys, Zhang *et al.*^{9,21)} introduced the idea of effective atomic radii for improving Miracle's equation. The optimal glass-forming compositions of Al-Ni-RE systems were determined to be $\text{Al}_{85.7}\text{Ni}_{9.5}\text{La}_{4.8}$ and $\text{Al}_{85.5}\text{Ni}_{9.5}\text{RE}_5$ (RE = Y, Ce, Gd, Dy). The predictions are in good agreement with the experimental results.⁹⁾

The physical and chemical properties of Co and Ni are very similar to each other. Their nominal atomic radii and Pauling electronegativities are 0.125 and 0.124 nm, and 1.91 and 1.88, respectively. The mixing enthalpy of liquid Al-Co (−18.8 kJ/mol) is also very close to that of liquid Al-Ni (−22.3 kJ/mol).³¹⁾ The Al-Co bond length is 0.244 nm, shortened by 10% compared to the sum of their nominal atomic radii, and the Al-RE bond length is close to the sum

of their nominal atomic radii.^{30,32)} Letting the effective atomic radius of Co be 0.101 nm, and those of Al and Y be their nominal atomic radii 0.143 nm and 0.180 nm, respectively, then the optimal glass-forming composition of Al-Co-Y alloys was calculated by the EAP model and the CSLI method to be $\text{Al}_{85.5}\text{Co}_{9.5}\text{Y}_{5.0}$. However, as described above, the t_c of $\text{Al}_{85.5}\text{Co}_{9.5}\text{Y}_5$ and its neighboring compositions is much less than that of $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ and its neighbors. It seems that the CSLI model fails to predict the optimal glass forming composition of Al-Co-Y alloys.

We can find other clues from the binary phase diagrams of Al-Co and Al-Ni to explain the difference in the best glass-forming composition between Al-Co-Y and Al-Ni-Y alloys. According to the binary phase diagrams, the intermetallic compound in the Al-rich region is monoclinic Al_9Co_2 for Al-Co alloys but orthorhombic Al_3Ni for Al-Ni alloys. The TM-centered clusters of Al_9Co_2 and Al_3Ni are shown in Fig. 6. The lattice parameters of the unit cell of monoclinic Al_9Co_2 are $a = 8.565 \text{ \AA}$, $b = 6.290 \text{ \AA}$, $c = 6.213 \text{ \AA}$, $\alpha = 90^\circ$, $\beta = 94.760^\circ$ and $\gamma = 90^\circ$, and two (Al_9Co_2) molecules are contained in one unit cell.³³⁾ Defining that two TM-centered clusters whose central atom distance is smaller than the double value of the maximal length of TM-Al bonds are adjacent, then it can be seen that in Al_9Co_2 a Co-centered cluster has 8 adjacent other Co-centered clusters at the distances of 4.171~4.753 Å or the average distance of 4.590 Å. In the case of Al_3Ni , a Ni-centered cluster connects with 12 adjacent Ni-centered clusters, and the distance to them changes between 3.793~4.801 Å with an average value of 4.405 Å. This means that more Ni-centered clusters can be filled within a certain volume than Co-centered clusters, i.e. the average distance between two adjacent Co-centered clusters is larger. Reflected in the common atoms between two adjacent TM-centered clusters, as shown in Fig. 6, it can be seen that two faces and two edges of each Ni-centered cluster are shared with other Ni-centered clusters. In contrast, only one edge and no face of a Co-centered cluster are shared by the adjacent Co-centered clusters in Al_9Co_2 . That is to say, less Al atoms are shared between two adjacent Co-centered clusters compared with Ni-centered clusters, and as a result the Al_9Co_2 compound contains more Al atoms. This can be further unraveled by the partial radial distribution functions of the TM central atoms in Al_3Ni and Al_9Co_2 , as illustrated in

Fig. 7 The partial radial distribution function of Al_3Ni and Al_9Co_2 : (a) Al atoms which surround the central TM atoms, (b) TM atoms which surround the central TM atoms.

Fig. 7. To get the RDF in Fig. 7, the structural data of Al_3Ni (ICSD #58040) and Al_9Co_2 (ICSD #57598) were input into the software “Atom of Demeter”, developed by B. Ravel,³⁴⁾ to calculate the distances from the TM central atom to its surrounding atoms, and then the RDF was gained by statistically analyzing the relationship between the species and quantity of atoms and their distance to the central atom. It can be seen that a similar number of Al atoms surround the Co and Ni central atoms, but there are more TM atoms surrounding a central TM atom in Al_3Ni than in Al_9Co_2 . So the Al_9Co_2 phase contains relatively more Al atoms.

We believe that the feature for TM-centered clusters to connect each other in Al_9Co_2 and Al_3Ni intermetallic compounds is also reflected in the Al-based amorphous alloys. Less common Al atoms are shared by two adjacent TM-centered clusters in Al–Co–Y amorphous alloys than in Al–Ni–Y amorphous alloys, meaning that more Al atoms are required for constructing the Co-centered clusters. Consequently the best glass former of Al–Co–Y alloys, $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$, contains a higher content of Al. The present work indicates that although the coordinate number of a TM atom is mainly determined by the solute/solvent atomic radii in Al–TM–RE amorphous alloys, the packing of the TM-centered clusters is closely related with the species of the TM elements, due to which the optimal glass-forming composition changes. Since there are more Al atoms in $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ alloy and its neighbors, only α -Al forms as the primary phase during annealing of the amorphous alloys.

5. Conclusion

The optimal glass-forming composition of Al–Co–Y alloys is $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$, from which amorphous samples up to 405 μm thickness can be produced. This composition is evidently different from $\text{Al}_{85.5}\text{Co}_{9.5}\text{Y}_{5.0}$, the prediction by the CSLI method and the EAP model. The reason is that Co-centered clusters tend to share vertexes and edges with each other in Al–Co–Y alloys, and relatively more Al atoms are required. It is suggested that the species of TM atoms can greatly influence the packing of the TM-centered clusters, and therefore influence the optimal glass-forming

composition. With more Al atoms, the amorphous alloys of Al–Co–Y around $\text{Al}_{89}\text{Co}_{5.5}\text{Y}_{5.5}$ exhibit no glass transition during isochronal annealing, and the primary crystallization phase is constantly fcc-Al.

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