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Prediction of good glass formers in the Al-Ni-La and Al-Ni-Gd systems using topological instability and electronegativity

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A new criterion has been recently proposed combining the topological instability (λ) criterion and the average electronegativity difference (Δe) among the elements of an alloy to predict and select new glass-forming compositions. In the present work, this criterion (λ,Δe) is applied to the Al-Ni-La and Al-Ni-Gd ternary systems and its predictability is validated using literature data for both systems and additionally, using own experimental data for the Al-La-Ni system. The compositions with a high λ,Δe value found in each ternary system exhibit a very good correlation with the glass-forming ability of different alloys as indicated by their supercooled liquid regions (ΔTg) and their critical casting thicknesses. In the case of the Al-La-Ni system, the alloy with the largest λ,Δe value, La55Al26.5Ni17.5, exhibits the highest glass-forming ability verified for this system. Therefore, the combined λ,Δe criterion is a simple and efficient tool to select new glass-forming compositions in Al-Ni-RE systems. © 2011 American Institute of Physics. [doi:10.1063/1.3563099]

I. INTRODUCTION

Several parameters to evaluate the glass-forming ability (GFA) have been proposed over the years based on thermal behavior, such as \( T_{rg} (\equiv T_g/T_l) \), \( \Delta T_g (\equiv T_c - T_g) \), \( \gamma (\equiv T_g/(T_c + T_l)) \), \( \delta (\equiv T_g/(T_l - T_c)) \), \( \gamma_m (\equiv 2T_c - T_g/T_l) \), \( \phi (\equiv T_{rg} (\Delta T_g/T_l)^{1/2}) \), \( \beta (\equiv T_g/(T_l - T_c)^2) \), \( \alpha (\equiv T_g/T_l) \) and \( \omega (\equiv T_g/T_c - 2T_g/(T_c + T_l)) \), where \( T_c \), \( T_g \), and \( T_l \) are the onset crystallization, liquidus and glass transition temperatures, respectively. The most widely used indicators are \( T_{rg} \), \( \Delta T_g \), \( \gamma_m \), and \( \gamma \). In a recent work, Guo et al.10 evaluated these criteria using statistical and physical considerations and found the best indicator of the GFA for the \( \gamma_m \) criterion.5 The biggest short coming, however, is that they cannot predict new compositions, since they make use of thermal properties of the glassy state. Therefore, the forecast of GFA is impossible.

The selection of new bulk metallic glasses (BMGs) in the last years was usually based on empirical rules21,22, i.e., (1) the glassy should consist of multicomponent alloys of three or more elements with alloy composition, (2) the atomic radius mismatch between elements should be more than 12%, (3) negative heat of mixing between the main elements, (4) using an alloy composition close to a deep eutectic, where the liquid phase has high thermal stability. These rules do not strictly apply in many cases since new bulk metallic glasses have been discovered in some binary systems12-16 in compositions far from the eutectic points14,17-20 and in alloys where the addition of elements with a positive heat of mixing can induce phase separation and hence improve the GFA.21

The lack of accuracy of these empirical rules shows the necessity for new tools with a reliable predictability in order to facilitate the search of new glass-forming compositions. Thus, much attention has been devoted in the past few years to develop better criteria to predict the best glass-forming composition(s) for different alloy systems or groups of systems. Some of these criteria are based on the electronic structure,22 the formation enthalpy of amorphous and crystalline phases,23,24 topological instability or destabilization of the host crystalline lattice,25-27 the electronegativity difference,25,26-30 calculation of the liquidus temperature based on the computer calculation of phase diagrams (CALPHAD) technique23 and structural models.23,31,32 However, so far there is no universal model that can predict new amorphous compositions and, in fact, many researchers are presently evaluating the connection of the different criteria to refine and improve their forecast and to obtain a better agreement with experimental data.23,28,33

Recently, a new criterion for the selection of amorphous alloys has been proposed with a combination of the topological instability (λ) criterion and the average electronegativity difference among the elements of an alloy (Δe) and a good correlation has been found for binary systems.25,28 The λ criterion was firstly proposed by Egami and Waseda34 to find the minimum solute concentration necessary to obtain amorphous phase by rapid quenching in a binary alloy system. An extension of this criterion was proposed to describe the crystallization behavior of Al-based alloys26 and to predict amorphization35 by trying to avoid compositional fields associated with primary crystallization in any particular metallic alloy composition. This extended criterion, termed λ criterion, can be calculated using the Eq. (1), where \( X_i \) is the atomic fraction of any solute element, \( Vm_i \) is the molar

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volume of any given solute and $V_m0$ is the molar volume of the phase:

$$\lambda = \sum X_j \left| \frac{V_m j}{V_m0} - 1 \right|.$$  \hspace{1cm} (1)

The molar volumes of phases can be obtained from the JCPDS database or estimated using Eq. (2), where $n_j$ is the number of $j$ atoms in the unit cell and $V_j$ is the atomic volume of the $j$ atom. $P_f$ is the packing factor and $N$ is the Avogadro’s number:

$$V_m = \frac{\sum n_j V_j}{P_f}. \hspace{1cm} (2)$$

The $\lambda$ value indicates the level of destabilization of an equilibrium phase (host crystalline lattice, considered in the equation by $V_m0$) due to the insertion of new elements into this phase. The larger this value, the larger will be the phase instability. Let us consider two adjacent phases in a binary phase diagram, i.e., an A-rich and a B-rich phase: the addition of B to the A-rich phase as well as the addition of A to the B-rich phase destabilizes each crystalline phase. At a certain composition the topological instability of both phases, or likewise their $\lambda$ values coincide, and this point corresponds to the maximum topological instability in the compositional region between them. For this composition the vitrification should be easiest. Based on this assumption and considering all equilibrium phases, a $\lambda$ plot is built.28

Nevertheless, as already discussed in,25,28 the predictability of this criterion is improved when the electronegativity is considered. Many works have described the influence of the electronegativity of the elements on the thermal behavior of amorphous alloys.56–58 which is related to the enthalpy of formation ($\Delta H$) and glass stability.7 The attractive force originates from the difference in electronegativity between the elements and is reflected in the heat of mixing. Elements with stronger attractive force exhibit larger heat of mixing, which favors the GFA, as also indicated by the well-known empirical rules.2 Therefore, it is reasonable to expect that the higher the average electronegativity difference among the elements is, the higher will be the glass-forming ability. Considering this, an average electronegativity map can be constructed for the system of interest using Eq. (3):

$$\Delta e = \sum X_i \left( \sum S_j |e_i - e_j| \right), \hspace{1cm} (3)$$

where $S_j = X_j V_m j ^{2/3}$ and $X_j$ and $X_i$ are the atomic fractions of the atoms $i$ and $j$, respectively. $V_m$ and $V_m j$ are the molar volumes of the elements $i$ and $j$, respectively and $e_i - e_j$ is the electronegativity difference between a central atom $i$ and its $j$ neighbors.

Both factors, the topological instability and the difference in the electronegativity are used together, assuming a synergistic effect, to produce a final criterion map, named $\lambda$. $\Delta e$ map.

In previous work40 we calculated this map for the Al-Ni-La system and compared with experimental data obtained from ribbons. The La$_{56}$Al$_{26.5}$Ni$_{17.5}$ alloy that presents the largest $\lambda.\Delta e$ value, exhibited the highest GFA verified so far for this system considering its thermal behavior. This composition is very close to the best BMG reported in literature for this system, La$_{55}$Al$_{25}$Ni$_{20}$. This alloy was one of the first BMGs that was found41 and was used as base composition for many other RE$_2$TM$_{30}$Al$_{125}$ (RE = rare earth elements and TM = transition metal elements) amorphous alloys.42,43 In this paper comparisons are done between the GFA of these two compositions and between the $\lambda.\Delta e$ values obtained for the Al-Ni-La system and the literature data. Additionally, we calculated the $\lambda.\Delta e$ map for the Al-Ni-Gd system and also compared with literature data. These results validate the $\lambda.\Delta e$ criterion as a good tool to select new glass formers in other Al-Ni-RE systems.

II. EXPERIMENTAL PROCEDURES

Calculations of the $\lambda.\Delta e$ criterion for the Al-Ni-La and Al-Ni-Gd systems were carried out using Eqs. (1) and (3). We considered in the calculations all phases established in the binary and in the partial ternary phase diagrams and, trying to incorporate all phases in the ternary systems, all ternary phases described in the JCPDS database were considered. The molar volumes of the phases were calculated using the JCPDS database and, for only for two of them (Al$_{15}$Ni$_3$Gd$_2$ and Al$_7$Ni$_5$Gd$_3$), the calculation was made using Eq. (2), assuming a packing factor of 0.72. The phases and their respective molar volumes are summarized in supplementary table.51

Rods were cast for the best predicted glass-forming alloy in the Al-La-Ni system (La$_{56}$Al$_{26.5}$Ni$_{17.5}$) and, additionally, for the glass with the largest critical casting thickness found in literature, La$_{55}$Al$_{25}$Ni$_{20}$.42 These samples were prepared by suction casting. The surface of the elements Al and Ni was chemically cleaned but La could not be cleaned because of its irregular shape and a correct reagent for etching and cleaning could not be found. The elements were repeatedly arc-melted and then suctioned directly into a water-cooled copper mold, producing samples with 2 mm diameter and 7 cm length.

Three rods for each composition were produced and each of them was divided into three sections: upper, middle, and bottom part. The cross section of each part was investigated by optical microscopy (OM) and the amount of glassy phase was determined. The rod that presented the largest volume fraction of glass (considering the average in these three different regions) was selected for further analysis.

Optical images and phase quantification were performed using a NIKON EPIPHOT 300 optical microscope with polarized light and software a4i analysis. XRD analysis was carried out on the cross section of the rods using an STOE diffractometer in transmission mode (Mo-K$_{\alpha}$ radiation), and the thermal stability behavior was evaluated with a Diamond DSC (Pyris) using a heating rate of 40 K/min. Scanning electron microscopy (SEM) images and energy-dispersive x-ray analysis (EDX) were carried out employing a SEM-FEG Gemini 1530 with a Bruker Xflash 4010 spectrometer. The oxygen content in the rods was measured by inductively-coupled plasma optical emission spectroscopy (ICP OES) using a TC-436 DR LECO.
III. RESULTS AND DISCUSSION

A. Al-Ni-La

The final $\lambda$,$\Delta \varepsilon$ criterion map applied to the Al-Ni-La system is shown in Fig. 1, where the brightest spots indicate compositions with the highest glass-forming ability. The binary eutectic compositions and the best glass-forming alloy reported in the literature for this system with a critical casting thickness of 3 mm, \( \text{La}_{55}\text{Al}_{25}\text{Ni}_{20} \) \cite{42}, were plotted for comparison. This composition is located in a region near the La-rich corner and near the point with the maximum $\lambda$,$\Delta \varepsilon$ value (0.069). We also plotted in this figure the alloys with large critical casting thickness found in the Al-La-(Cu,Ni) system. \cite{52} Obviously, these alloys locate in a region with high $\lambda$,$\Delta \varepsilon$ values; in fact, the small deviation from the predicted compositions is probably associated with the fact that Ni and Cu were considered as the same element in order to make the comparison possible.

The $\Delta T_x$ values ($\Delta T_x = T_x - T_g$, where $T_x$ and $T_g$ are the onset crystallization and glass transition temperatures, respectively) for these alloys are compared with the $\lambda$,$\Delta \varepsilon$ values in Fig. 2. These values increase toward larger $\lambda$,$\Delta \varepsilon$ values. Large $\Delta T_x$ values imply that the supercooled liquid can occur in a wide temperature range without crystallization and has a high resistance against nucleation and growth of the respectively crystalline phases. \cite{53,54}

Inoue \textit{et al.} \cite{55} measured the $\Delta T_x$ and $T_{rg}$ values ($T_{rg} = T_g/T_m$, where $T_g$ and $T_m$ are the glass transition temperatures and the melting temperature, respectively) values for a vast number of alloys in the ternary Al-La-Ni system. The obtained results are displayed together with the $\lambda$,$\Delta \varepsilon$ criterion map in Figs. 3(a) and 3(b). Also plotted in these figures are the best glass formers reported in the literature as well as the binary eutectic compositions \cite{45,48,50} for comparison. The regions with best $\Delta T_x$ and $T_{rg}$ are located near the brightest region of the $\lambda$,$\Delta \varepsilon$ map and it appears that the $\Delta T_x$ values indicate a better consistency with the criterion results than...
the $T_{rg}$ values. It is interesting to point here that the $T_{rg}$ values calculated by Inoue et al.\textsuperscript{55} was made considering the melting temperature ($T_m$) but not the liquidus temperature ($T_l$). It is known that the GFA exhibits a more reliable correlation with $T_{rg}$ when the liquidus temperature is taken into account.\textsuperscript{56}

In order to determine the actual critical casting thickness of the alloy with the highest $\lambda\Delta\epsilon$ value ($\La_{56}\Al_{26.5}\Ni_{17.5}$, $\lambda\Delta\epsilon = 0.069$) and attempt to compare with the best glass former verified in the literature ($\La_{35}\Al_{25}\Ni_{20}$), rods of these two compositions were prepared and analyzed. The results are shown in Figs. 4–8. We named the best composition predicted by the $\lambda\Delta\epsilon$ criterion as “criterion rod” and the best amorphous composition reported in the literature as “literature rod.” The cross sections of both rods are shown in Figs. 4(a) and 4(b) for the literature and the criterion rod, respectively. Two kinds of regions can be separated, with a brighter and darker contrast. The darker correspond to crystalline phases and the brighter consist of an amorphous phase with, most likely, crystalline phases in a small length-scale. The volume fraction of the brighter phase(s) is higher in the criterion rod than the literature rod [Figs. 4(a) and 4(b)]. Moreover, a big crack is observed in the cross section of the literature rod, originated from shrinkage during solidification of the respectively crystalline phases. This confirms a larger amount of crystalline phase in this sample, since no crack was found in the criterion rod.

The XRD results for the two types of rods are shown in Fig. 5. Even though the compositions are different, the specimens exhibit the same crystalline phases, namely $\La_3\Al$, $\La_2\O_3$ and $\La_7\Ni_3$. Yet, some peaks could not be identified. In the case of the criterion rod there are two broad but weak maxima on which the Bragg peaks are superimposed. For the literature rod these features are absent and, instead, the crystalline reflections are sitting on top of a flat baseline. This indicates the presence of more amorphous phase in the criterion rod and hence corroborates the results of the microscopy.

The DSC results for the two rod samples are shown in Fig. 6. Only the criterion rod exhibits a glass transition with an onset temperature of 476 K. The crystallization temperatures are 517 and 522 K for the composition predicted by our criterion and for the composition reported in the literature, respectively. The literature rod did not exhibit glass
transition, which means that the composition of the residual amorphous phase significantly changed (the $\Delta T_x$ for this alloy was reported as 69 K)\textsuperscript{42} as a result of the partial crystallization. This result indicates that the literature rod presents more crystalline phase than the criterion rod, which has a glass transition temperature similar to its ribbon\textsuperscript{40}.

The amount of the amorphous phase was estimated based on the enthalpies of crystallization. These values were $-46\pm 1$ and $-9\pm 1$ J/g for the criterion and literature rod, respectively, and $-119\pm 1$ and $-73\pm 1$ J/g (taken from Ref. \textsuperscript{41}) for their melt-spun ribbons, respectively. Assuming a fully amorphous structure in the ribbons, the calculated volume fraction of the amorphous phase was 38.7% and 12.3% for the criterion and literature rod, respectively, indicating more amorphous phase in the predicted alloy.

The criterion rod presents a supercooled liquid region of 41 K. Comparing with the result obtained for the ribbon ($\Delta T_x$ of 79 K)\textsuperscript{40} reveals a significant difference. The composition of the amorphous phase measured by EDX was La\textsubscript{52±2}Al\textsubscript{29±2}Ni\textsubscript{19±1}, indicating a significant difference from the nominal composition (La\textsubscript{56}Al\textsubscript{26.5}Ni\textsubscript{17.5}) due to partial crystallization during solidification. Another point is that the quenched-in crystalline phases may act as nucleation sites for subsequent crystallization upon heating, and thus decrease the crystallization temperature, as it has been observed for the cast rod.

Figures 7 and 8 show SEM images and corresponding EDX maps for the two different rods. The micrograph [Fig. 7(a)] shows a homogeneous glassy phase without any contrast, and crystalline phases with a dark contrast. The EDX analysis indicates that the elements are homogeneously distributed in the glassy matrix, as can be seen from the EDX maps [Figs. 7(b)–7(e)], and that the crystalline phase is an O-rich particle, with a composition near the La\textsubscript{2}O\textsubscript{3} oxide. Some of these particles are located at the surface, which means that they probably appeared after the metallographic procedure.

Figure 8 shows a SEM micrograph of the literature rod, revealing the presence of five crystalline phases. The analyzed region corresponds to the center of the cross section, and represents a magnification of the microstructure visible in Fig. 4(a). The phases were identified by EDX as La\textsubscript{3}Al, La\textsubscript{2}O\textsubscript{3}, Al\textsubscript{12}Ni\textsubscript{42}La\textsubscript{2}O\textsubscript{3}, and La\textsubscript{7}Ni\textsubscript{3} as indicated in Fig. 8 by numbers 1, 2, 3, and 4, respectively. The last crystalline phase’s composition, marked as number 5, could not be precisely measured, but the EDX maps in Figs. 7(b)–7(e), reveal a large amount of oxygen, indicating this phase an oxide. This oxide is probably related to the one observed in the ribbons\textsuperscript{40} and can be related with some peaks not identified in the rods diffraction pattern, maybe exhibiting some similar peaks like La\textsubscript{2}O\textsubscript{3}. It is located in eutectic agglomerates with the La\textsubscript{7}Ni\textsubscript{3} phase, as can be seen in Fig. 8. The La\textsubscript{2}O\textsubscript{3} oxide (phase number 2) was found at the surface, probably being formed during sample polishing. The very intense unidentified peaks described in the rods diffraction pattern are related to the Al\textsubscript{25±1}Ni\textsubscript{42.1±0.6}La\textsubscript{32.3±0.4} phase, marked with number 3.
Both literature data and results from ribbons indicate that the values of the $\lambda,\Delta e$ criterion are proportional to the $\Delta T_k$ values: larger values indicate a wider supercooled liquid region. A correlation between the electronegativity difference and atomic size with $\Delta T_k$ values was also reported by Fang et al. They showed that $\Delta T_k$ values are proportional to three parameters: the electronegativity difference, the atomic size and the valence electron difference, and a good correlation between calculated and experimental $\Delta T_k$ values were described for Fe-, Pd-, and Mg-based alloys. Considering that the larger the $\Delta T_k$ value, the higher the critical diameter for amorphization, we can say that the La$_{56}$Al$_{26.5}$Ni$_{17.5}$ alloy presents the best GFA in this system. This agrees well with previous results for Al-La-Ni and Al-La-(Ni,Cu) alloys in the literature.\textsuperscript{5,52,55}

The results of the rods of this composition (La$_{56}$Al$_{26.5}$Ni$_{17.5}$) and the best composition reported in the literature (La$_{55}$Al$_{25}$Ni$_{20}$, with a critical diameter of 3 mm for amorphization) are in agreement with such conclusion, since more amorphous phase was found in the predicted composition. The critical casting thickness obtained for the literature composition was smaller in our experiments because of a different processing and maybe because of the use of slightly impure lanthanum pieces, which might have led to the formation of the unidentified oxide (phase described as number 5 in Fig. 8). We additionally performed oxygen analyze to quantify the amount of oxygen in these samples. The results were 0.68 and 0.61 at.\% O for the literature and criterion rod, respectively. It is interesting to mention here that this amount was underestimated, since the oxide formed on the surface also contributed to the measured signal. Thus, these values suggest that the composition of the rods did not change too much, which in turn should not affect glass-forming ability significantly.

B. Al-Ni-Gd

The final $\lambda,\Delta e$ criterion map calculated for the Al-Ni-Gd system is shown in Fig. 9, together with the binary eutectic points.\textsuperscript{44,46,47,50} Chen et al.\textsuperscript{57} investigated the glass-forming ability of the Al-Ni-Gd system and they obtained many BMGs in the Gd-rich corner. These data are also plotted in Fig. 9 for comparison. A small change in the composition; for example, from Gd$_{60}$Ni$_{15}$Al$_{25}$ to Gd$_{60}$Ni$_{15}$Al$_{25}$, can improve the GFA tremendously, which increase the critical diameter of amorphization from 1 to 4 mm.

The BMGs are located exactly near the three peaks of the $\lambda,\Delta e$ criterion at the compositions Gd$_{50.5}$Ni$_{19}$Al$_{30.5}$, Gd$_{50.5}$Ni$_{16.5}$Al$_{23}$, and Gd$_{50}$Ni$_{28.5}$Al$_{21.5}$ with $\lambda,\Delta e = 0.05852$, 0.05684, and 0.05593, respectively. The best glass former, Gd$_{60}$Ni$_{15}$Al$_{25}$ alloy with a critical casting thickness of 4 mm, is located near the peak with second highest value, at the composition of Gd$_{60.5}$Ni$_{16.5}$Al$_{23}$.

IV. LIMITATIONS OF THE NEW $\lambda,\Delta e$ CRITERION

In the following, qualitative arguments shall be considered, which could improve the predictability of the present $\lambda,\Delta e$ criterion.

To perform the calculations, all phases in the respective system need to be known. Nowadays this information can be obtained for many binary and ternary systems, but it is still quite poorly established for systems with four or more elements.

Another point is that this criterion cannot differentiate the glass-forming ability of stoichiometric intermetallic compounds which exhibit all the same value by definition. Considering, for example, the $\Delta T_k$ regions described in Fig. 3(a), it can be seen that the AILaNi intermetallic is located in a region with a high glass-forming ability but cannot be described by its zero $\lambda,\Delta e$ value (the $\lambda$ value is assumed to be 0 because there is no addition of elements to promote the topological instability in this specific composition). One factor that could be considered is an additional parameter that takes into account the changes in the liquidus temperature, which reflect the stability of the intermetallic phases.

Many works describe that the best region for glass formation near the Al-rich corner of the Al-Ln-TM systems (Ln = Lanthanides and TM = transition metals) is on the $\lambda = 0.1$ line,\textsuperscript{26,34,38,58} which was also added in Fig. 3(a). This region could not be predicted by the combined $\lambda,\Delta e$ criterion. Ln- and TM-centered icosahedral coordination clusters can be formed in this region,\textsuperscript{38,39,59} which is incompatible with primary crystallizing phase. Such a structure hampers the precipitation of the crystalline equilibrium phase(s),\textsuperscript{60} which in turn could explain the improvement of the glass-forming ability.

As we can see, different factors take part in a ternary alloy system and, for a complete GFA description, all these factors need to be considered together, which makes this task difficult and still requires further experimental and theoretical efforts.

V. CONCLUSIONS

The synergism between electronegativity and topological instability as considered by the $\lambda,\Delta e$ criterion is a good tool to select new glass formers in the Al-Ni-RE systems.

The results confirmed that the La$_{56}$Al$_{26.5}$Ni$_{17.5}$ alloy is the best glass former verified so far in the Al-Ni-La system.
A good correlation between the $\lambda, \Delta e$ criterion with literature data and own measurements was obtained. Also a clear correlation between $\Delta T_e$ and the $\lambda, \Delta e$ values was found for the Al-La-Ni system.

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23. See supplemental material at http://dx.doi.org/10.1063/1.3563099 for phases and their respective molar volumes.