Mechanical Strengths, Thermal Stability and Electrical Resistivity of Aluminum-Rare Earth Metal Binary Amorphous Alloys

By A. Inoue*, T. Zhang**, K. Kita*† and T. Masumoto*

Aluminum-rich amorphous alloys in Al-Ln (Ln=lanthanide element) binary systems were formed in the range of 7 to 16 at% Ln by melt spinning. The amorphous alloys exhibit good bending ductility in the entire compositional ranges. With increasing Ln content, the crystallization temperature, Vickers hardness number, tensile strength and electrical resistivity at room temperature of the Al-Ln amorphous alloys increase from 434 to 534 K, 150 to 270, 360 to 870 MPa and 48 to 109 $\mu\Omega\text{cm}$, respectively; thus there are no systematic changes of their values with the change in the atomic number of the Ln metals. It is noteworthy that the Al-Ln amorphous alloys have a high tensile strength of 870 MPa which exceeds the highest strength obtained with conventional Al-base crystalline alloys. The glass formation and compositional dependence of the amorphous binary alloys are explained on the basis of the assumption that the attractive bonding nature between Al and Ln metals is dominant rather than the volume mismatch resulting from the difference in atomic size.

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I. Introduction

It has recently been found that Al-rich alloys in the Al-Ln-TM (Ln=rare-earth metal and TM=transition metal) systems can be easily amorphized by liquid quenching⁽¹⁾⁻⁽³⁾ and the resulting amorphous alloys exhibit high mechanical strengths combined with low density⁽⁴⁾, good corrosion resistance⁽⁵⁾, large glass-forming capacity⁽⁵⁾ and distinctly appreciable glass transition phenomenon⁽⁶⁾. The good combined properties of the Al-Ln-TM amorphous alloys are expected for developing a new type of high-strength materials. As an amorphous phase in Al-rich binary alloys is formed only in the Al-Ln system, clarification of the compositional range and mechanical, thermal and electrical properties of Al-Ln binary amorphous alloys is very important for a better understanding of the origin for the appearance of

the above-described combined properties for the Al-Ln-TM ternary amorphous alloys. The present authors have previously reported on the glass formation of Al-Ln (Ln=Y, La, Ce, Pr, Nd, Sm or Gd) binary alloys by liquid quenching and the mechanical and thermal properties of their binary amorphous alloys in two short papers⁽⁷⁾⁽⁸⁾. In a series of researches on the glass formation in Al-rich Al-Ln binary alloys, the formation of an amorphous phase was also found in the other binary alloys of Al-Ln (Ln=Tb, Dy, Ho, Er or Yb). The aim of this paper is to present the compositional range, mechanical strength, thermal stability and electrical properties of Al-Ln (Ln=Tb, Dy, Ho, Er or Yb) amorphous alloys and to investigate a common effect of Ln elements on the glass formation and the above-described properties of the Al-Ln binary amorphous alloys.

II. Experimental Procedure

Al-Ln (Ln=Tb, Dy, Ho, Er or Yb) binary alloys in the compositional range below 25 at% Ln were used in the present study. The Al-Ln ingots were prepared by arc melting a

* Institute for Materials Research, Tohoku University, Sendai 980, Japan.

** Graduate School, Materials Science and Engineering Department, Tohoku University.

† On leave from Yoshida Kogyo K. K., Yoshida Kurobe, Toyama Pref. 938, Japan.

mixture of pure Al(99.99 mass%) and Ln metal in a purified argon atmosphere. The compositions are nominally expressed in atomic per cent. From the master alloy ingots, ribbons with a cross-section of about $0.02 \text{ mm} \times 1 \text{ mm}$ were prepared by a single roller melt-spinning technique. The amorphicity of the melt-spun ribbons was examined by X-ray diffraction using Cu-K α radiation, transmission electron microscopy (TEM) and differential scanning calorimetry (DSC). The DSC measurement was made at a heating rate of 40 K/min. The methods of characterizing the bending ductility, crystallization temperature and heat of crystallization have been described previously⁽⁹⁾. Hardness and tensile strength of the ribbon specimens were measured using a Vickers microhardness tester at a 50 g load and an Instron-type tensile testing machine at a strain rate of $8.3 \times 10^{-4} \text{ s}^{-1}$, respectively. The methods of determining the hardness and tensile strength values have been described in ref. 4. Electrical resistivity was measured in the temperature range of 77 K to room temperature by the four-probe technique. The thickness of the ribbons, necessary to calculate electrical resistivities, was estimated by a density-length-weight method. The absolute error in the present measurements is of the order of 15%.

III. Results

Figure 1 shows the compositional dependence of the as-quenched phase in the Al-Tb, Al-Dy, Al-Ho, Al-Er and Al-Yb binary systems. The amorphous phase is formed in compositional ranges of 9 to 14%Tb and 9 to 12% of Dy, Ho, Er or Yb. From the previous data⁽⁷⁾⁽⁸⁾ that an amorphous phase of Al-Ln binary alloys is formed in the ranges of 9 to 13%Y, 7 to 10%La or Ce, 8 to 12%Nd or Gd and 8 to 16%Sm and in the vicinity of 10%Pr, the glass-formation range is the widest for Al-Sm, followed by Al-Tb, Al-(Y, Nd or Gd), Al-(La, Ce, Dy, Ho, Er or Yb) and then Al-Pr. Furthermore, the supersaturated fcc solid solution is formed in the concentration ranges below the glass-formation ranges instead of an equilibrium mixture of fcc Al and intermetallic Al-Ln compounds. In the concentration

Fig. 1 Compositional dependence of structure in rapidly solidified Al-Tb, Al-Dy, Al-Ho, Al-Er and Al-Yb binary alloys.

ranges by about 1 at% higher than the glass-formation ranges, the quenching-induced phases are amorphous plus unidentified x-phases in the Al-Dy, Al-Ho, Al-Er and Al-Yb systems and Al plus x-phases in the Al-Tb system. A further increase of the solute concentration gives rise to the formation of equilibrium phases of Al plus Al_3Ln ($\text{Ln} = \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$ or Yb).

Figure 2 shows the relation between the glass-formation range and the equilibrium phase diagram⁽¹⁰⁾ for the Al-Ln binary alloys.

Fig. 2 Composition ranges for the formation of an amorphous phase in the Al-Ln ($\text{Ln} = \text{Y}, \text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}, \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$ or Yb) binary systems. Their binary phase diagrams were adopted from Ref. (10).

The glass-formation ranges except for the Al–Nd system lie in the compositional range between a eutectic point and $\text{Al}_{11}\text{Ln}_3$ ($\text{Ln}=\text{La}$, Ce , Pd , Nd or Sm) or Al_3Ln ($\text{Ln}=\text{Y}$, Gd , Tb , Dy , Ho , Er or Yb) compounds. Furthermore, the solidus temperature and the temperature difference between the liquidus and solidus points in their glass formation ranges are in the range of 625 to 655 K and 305 to 627 K, respectively. The melt-quenched phase at the eutectic composition with the lowest melting temperature is composed only of the fcc solid solution in all the alloy systems. This result is in disagreement with the previous tendency that the solute-poor amorphous alloys in metal-metal systems such as $(\text{Fe}, \text{Co or Ni})\text{--Zr}^{(11)}$ and $(\text{Fe}, \text{Co or Ni})\text{--Hf}^{(12)}$ etc., lie in the composition ranges including their eutectic points. The reason why the fcc single phase forms in place of an amorphous phase in spite of its trough of melting temperature near the eutectic composition is presumably because their eutectic compositions are too low to construct a stable amorphous structure.

Figure 3 shows onset temperature of the first exothermic peak (crystallization temperature, T_x) as a function of Ln content for the Al–Ln amorphous alloys. With increasing Ln content from 7 to 12 at%, T_x increases significantly from 434 to 534 K and no further increase of T_x is seen in the Ln concentration range of 12 to 16 at%. Thus, the compositional effect of T_x is almost independent of the atomic number of the Ln elements, though the Al–Er and Al–Yb alloys exhibit lower T_x values. In order to clarify the reason for the significant increase of T_x for the Al–Ln amorphous alloys with increasing Ln content from 7 to 12 at%, crystallization behavior was examined for some Al–Ln amorphous alloys. As examples, Fig. 4 shows differential scanning calorimetric curves of amorphous $\text{Al}_{100-x}\text{Tb}_x$ and $\text{Al}_{100-x}\text{Dy}_x$ ($x=9, 10, 11$ and 12 at%) alloys. Two or three exothermic peaks are seen on the DSC curves, indicating that the crystallization takes place through two or three stages. From the X-ray diffraction analyses, it was confirmed that the broad exothermic peak at the low-temperature side for $\text{Al}_{91}\text{Tb}_9$, $\text{Al}_{90}\text{Tb}_{10}$ and $\text{Al}_{91}\text{Dy}_9$ alloys is due to the precipitation of fcc Al and the sec-

Fig. 3 Crystallization temperature (T_x) of amorphous Al–Ln ($\text{Ln}=\text{Y}$, La , Ce , Pr , Nd , Sm , Gd , Tb , Dy , Ho , Er or Yb) alloys as a function of Ln content.

Fig. 4 Differential scanning calorimetric curves of amorphous $\text{Al}_{100-x}\text{Tb}_x$ (a) and $\text{Al}_{100-x}\text{Dy}_x$ (b) ($x=9, 10, 11$ and 12 at%) alloys.

ond exothermic peak is due to the precipitation of Al_x and Al_3Tb or Al_3Dy phases from the remaining amorphous phase. The increase of T_x by the disappearance of the Al phase is also seen for the other Al–Ln ($\text{Ln}=\text{Y}$, La , Ce , Nd or Sm) amorphous alloys. Accordingly, the strong compositional dependence of T_x shown

in Fig. 3 is due to the disappearance of fcc Al phase resulting from the change of the crystallization process of $\text{Am.} \rightarrow \text{Am.} + \text{Al} \rightarrow \text{Al} + \text{Al}_3\text{Ln}$ to $\text{Am.} \rightarrow \text{Al} + x + \text{Al}_3\text{Ln} \rightarrow \text{Al} + \text{Al}_3\text{Ln}$ or $\text{Am.} \rightarrow \text{Al} + \text{Al}_3\text{Ln}$.

Figure 5 shows the change of Vickers hardness number (H_v) as a function of Ln content for the Al-Ln amorphous alloys. The H_v shows an almost linear dependence in the entire compositional range and increases significantly from 150 to 270 for the Al-Ln alloys. Although the Al-Sm, Al-Dy and Al-Tb amorphous alloys exhibit higher H_v values, no systematic change in H_v with the atomic number of the Ln elements is seen for the Al-Ln amorphous alloys. Here, it is important to describe

Fig. 5 Vickers hardness (H_v) of amorphous Al-Ln (Ln=Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er or Yb) alloys as a function of Ln content.

Fig. 6 Tensile fracture strength (σ_f) of amorphous Al-Ln (Ln=Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er or Yb) alloys as a function of Ln content.

that all the amorphous alloys in the Al-Ln binary system have a good ductility which is shown by a 180-degree bending without fracture. Figure 6 shows the change of tensile fracture strength (σ_f) as a function of Ln content for the Al-Ln amorphous alloys. The σ_f increases significantly from 360 to 870 MPa with increasing Ln content. It should be noted that all the amorphous alloys in the Al-Ln systems exhibit tensile strengths which exceed the highest value (530 MPa) obtained with optimally age-hardened Al-base alloys. Figure 7 shows the fracture surface appearance of an $\text{Al}_{88}\text{Tb}_{12}$ amorphous ribbon fractured by the uniaxial tensile test. The fracture surface consists of a smooth region (A) caused by shear sliding and a vein region (B) caused by final fracture after shear sliding. Furthermore, it is seen that the fracture takes place along the shear plane declined by 45 to 55 degrees to the longitudinal tensile direction. These features in the fracture behavior are the same as those reported previously⁽⁴⁾ for the other ductile amorphous alloys.

Figure 8 shows the temperature dependence of electrical resistivity in the range of 77 K to room temperature for amorphous $\text{Al}_{90}\text{Ln}_{10}$ (Ln=Nd, Sm, Gd, Tb, Dy, Ho, Er or Yb) alloys. It can be seen for all the amorphous alloys that the resistivity change is positive in the entire temperature range and almost linear

Fig. 7 Scanning electron micrograph of tensile fracture surface for an amorphous $\text{Al}_{88}\text{Tb}_{12}$ alloy.

Fig. 8 Electrical resistivity of amorphous Al-Ln (Ln=Nd, Sm, Gd, Tb, Dy, Ho, Er or Yb) alloys as a function of temperature.

at temperatures above 150 K. As the Ln content increased from 8 to 12 at%, the resistivity at room temperature was measured to increase from 48–65 to 76–109 $\mu\Omega\text{cm}$. Furthermore, the temperature coefficient of resistivity defined by $1/\rho_{RT}(d\rho/dT)$ for the $\text{Al}_{90}\text{Ln}_{10}$ amorphous alloys is in the range of 0.134×10^{-4} to $6.29 \times 10^{-4} \text{ K}^{-1}$. Thus, the feature of the resistivity behavior is also independent of the atomic number of the Ln elements.

IV. Discussion

1. Glass formation of the Al-Ln binary alloys

In this section, the reason for the formation of the binary Al-Ln (Ln=Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er or Yb) amorphous alloys is discussed on the basis of the previous empirical factors for the glass formation of binary alloys by liquid quenching; namely, (1) the atomic size ratio of the constituent atoms is below about 0.8, and (2) the interaction between the constituent atoms is attractive and the mixing enthalpy is relatively large. Egami and Waseda⁽¹³⁾ proposed the concept that the

Table 1 The minimum solute concentration ($C_b^{\min}$) for the glass formation in the Al-Ln (Ln=Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er or Yb) binary alloys by liquid quenching, the volume mismatch between Al and Ln atoms (ΔV) and the critical size factor (λ_0) obtained by the product of $C_b^{\min}$ and ΔV .

Ln metal	$C_b^{\min}$	ΔV	λ_0
Y	0.09	1.062	0.096
La	0.07	1.272	0.087
Ce	0.07	1.096	0.077
Pr	0.09	1.096	0.098
Nd	0.08	1.062	0.084
Sm	0.08	0.961	0.077
Gd	0.08	0.929	0.083
Tb	0.09	0.864	0.078
Dy	0.09	0.833	0.075
Ho	0.09	0.833	0.075
Er	0.09	0.802	0.072
Yb	0.09	1.497	0.135

change from a crystalline solid solution to an amorphous phase in rapidly solidified alloys takes place when a volume mismatch due to the difference in atomic size exceeds a critical size factor (λ_0). Furthermore, it has empirically been shown that the λ_0 value is nearly equal to 0.1 for the binary amorphous alloys. Here, the λ_0 value is given by the product of the minimum solute concentration ($C_b^{\min}$) for the glass formation in a binary alloy system by liquid quenching and the volume mismatch of the constituent atoms (ΔV). The λ_0 values at the minimum concentration for the Al-Ln alloys were calculated and summarized in Table 1 together with the data of $C_b^{\min}$. As shown in Table 1, the λ_0 values except that of Al-Yb alloy tend to decrease from 0.098 to 0.072 with increasing atomic number of the Ln metals because of the decrease in atomic size. The λ_0 values for the Al-based alloys containing Ln metals with atomic numbers below 60 are in the range of 0.08 to 0.10, being roughly consistent with the empirical λ_0 value of ≈ 0.1 for the glass formation. However, the λ_0 values for the Al-Ln alloys containing heavier Ln metals except Yb are in the range below 0.08 and the Al-Yb alloy has a high value of 0.135 which deviates largely from the critical λ_0 value (≈ 0.10), being inconsistent with the empirical rule. Furthermore, although the atomic size ratios of the Al-Ln alloys are in the range of

0.73 to 0.80 for Y, La, Ce, Pr, Nd, Sm, Gd or Yb, the ratios for the other Ln metals are larger than the empirical critical size ratio of 0.80 for the glass formation by liquid quenching. The inconsistency of the λ_0 and atomic size ratio for the Al-Ln (Ln=Tb, Dy, Ho or Er) alloys indicates that the amorphization of the present binary alloys cannot be explained only by the empirical concept of the volume mismatch caused by the difference in atomic size.

It has previously been pointed out⁽¹⁴⁾ that the attractive interaction between Al and transition metal must be taken into consideration in the glass formation of Al-early transition metal (ETM)-late transition metal (LTM) such as Al-Ni-Zr, Al-Ni-Nb and Al-Ni-Hf etc., because the empirical critical size ratio of 0.8 is not satisfied in many Al-rich amorphous alloys belonging to the Al-ETM-LTM system. It is reasonable to consider that the concept of the attractive interaction can be also applied to the interpretation of the glass formation of the present Al-Ln alloys. Although there are no measured data of the mixing energies between Al and Ln metals, the enthalpy of mixing for Al-Y and Al-La alloys has theoretically been predicted⁽¹⁵⁾ to be -131 and -109 kJ/g-atom, respectively. In addition, the equilibrium phase diagrams⁽¹⁰⁾ of the Al-Ln binary alloys indicate that intermetallic compounds with high melting temperatures exist at Al-rich compositions and the solute concentrations and melting temperature, which reflect the magnitude of attractive interaction (mixing energy) between Al and Ln metals, are in the range of 21.4 to 25 at% and 1253 to 1723 K, respectively. The minimum solute concentrations (21.4 to 25 at%) for formation of the Al-rich compounds are equal to or lower than that of Al-TM binary alloys, and the melting temperatures of $\text{Al}_{11}\text{Ln}_3$ and Al_3Ln compounds are higher than those (973 to 1217 K) of Al-rich compounds (Al_4Ca and Al_9Co_2) with solute concentrations below 25 at%. These data allow us to conclude that the magnitude of attractive interaction between Al and Ln metals is larger for Ln metals than for the other solute metals. Accordingly, it may be concluded that the glass formation in Al-Ln binary alloys by

liquid quenching results mainly from a strong attractive interaction of the constituent elements rather than the volume mismatch by the difference in the atomic size ratio. The enhancement of the glass-forming capacity by the increase of the attractive interaction of the constituent atoms is probably because the diffusivity of the constituent atoms in the supercooled liquid region becomes difficult and the temperature dependence of viscosity becomes steep for the supercooled liquid.

2. Compositional dependence of mechanical strengths

It was described in section IV.1 that the attractive bonding nature between Al and Ln metals plays an important role in the glass formation of Al-Ln binary alloys. Furthermore, the structural data⁽¹⁶⁾ obtained by the anomalous X-ray diffraction method indicate that the coordination number of Al atoms around Y atoms is about 1.4 times as large as the average value expected from the atomic concentration of the alloy and hence the short-range ordering of Al-Y pairs develops homogeneously in the Al-Y amorphous phase. The formation of the short-range ordered structure is probably due to the strongly attractive interaction between Al and Y atoms. It is therefore reasonable to consider that the number of the attractive Al-Y pair in the Al-Y amorphous alloys increases linearly with increasing Y content, leading to the linear increase of mechanical strengths. This presumption is consistent with the result in which the σ_f and H_v values of the Al-Ln amorphous alloys increase almost linearly with increasing Ln content and are independent of the atomic number of the Ln metals. In other words, the high strengths of the Al-Ln binary amorphous alloys seem to result from the Al-Ln bonding with strong attractive interaction and the remarkable increase of σ_f and H_v is due to the increase of the number of the Al-Ln bonding. Furthermore, the attractive bonding nature between Al and Ln metals is probably due to a high hybridization tendency between s-p electrons in Al and f-d electrons in Ln metals.

It was shown that all the properties of T_x , H_v , σ_f and ρ_{RT} for the Al-Ln amorphous alloys

were independent of the atomic number of the Ln metals. The atomic size of the Ln metals varies systematically with the atomic number and hence the atomic size factor also seems to have little effect on the above-mentioned properties. On the other hand, it is generally known that the inherent chemical nature of the Ln metals results from 4f electrons which lie at the inner side in their atoms. Although the number of the 4f electrons varies systematically with the atomic number, the electrons are screened by 5s² and 5p⁶ electrons which lie at the outer side of the atoms, resulting in a similarity in chemical properties of the Ln metals. Accordingly, it may reasonably be assumed that the independence of the properties of the Al-Ln amorphous alloys as a function of atomic number is due to the unique electronic structure in which the 4f electrons are screened by 5s and 5p electrons.

Similarly, the disappearance of the primary Al phase with increasing Ln content in the crystallization process of the Al-Ln amorphous alloys is explained on the basis of the decrease of the number of Al atoms which do not bond with Ln metals. The suppression of the primary Al phase gave rise to a rapid increase of T_x . However, the increase of the number of the Al-Ln pairs seems to bring about an ease of the precipitation of the Al-Ln compounds, resulting in almost saturated T_x values at high Ln concentrations in the Al-Ln amorphous alloys shown in Fig. 3.

3. Compositional dependence of electrical resistivity

As described in section III, the electrical resistivity at room temperature for the Al-Ln amorphous alloys increases significantly from 48 to 109 $\mu\Omega\text{cm}$ with increasing Ln content from 8 to 12 at%. Furthermore, it has previously been reported⁽¹⁷⁾ that the Hall coefficient (R_H) at room temperature for the Al-Y amorphous alloys is negative and the magnitude increases from 22×10^{-12} to $32 \times 10^{-12} \text{ m}^3/\text{As}$ with increasing Y content. From the increase in the magnitude of the negative Hall coefficient, it is interpreted that the number of the free electrons contributing to electrical conductivity decreases with increasing Y content, if one

assumes the relation of $R_H = -1/(Nne)$ which can be derived from the nearly free electron model. Here, N is the atomic density and n is the number of electrons per atom. The strongly attractive interaction between Al and Ln atoms suggests that the s and p electrons in Al metal hybridize with f- and d-electrons in Ln metals, leading to the decrease of the free electrons which are attributed to electrical conductivity. As a result, the increase of the number of the Al-Ln pairs in Al-Ln amorphous alloys with increasing Ln content gives the significant increase in electrical resistivity and in magnitude of the negative Hall coefficient.

As described in this section, the glass formation of the Al-Ln binary alloys and the compositional dependences of σ_f , H_v , ρ_{RT} and R_H are explained by the common concept that the Al-Ln pairs with attractively bonding nature are formed preferentially in the Al-Ln amorphous alloys and the s and p electrons in Al hybridize tightly with the 4f and 5d electrons in the Ln metals.

V. Summary

Melt spinning of Al-rich alloys gives an amorphous phase in binary Al-Ln (Ln=Y, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er or Yb) systems. The glass formation extends from about 7 to 16 at% Ln for the Al-Ln alloys. All the amorphous alloys exhibit good ductility which is shown by bending through 180 degrees without fracture. Crystallization temperature increases rapidly with an increase of Ln content from 7 to 10 at% because of the disappearance of primary Al phase and becomes nearly constant in the higher Ln content range. Tensile strength (σ_f) and Vickers hardness (H_v) increase almost linearly from 360 to 870 MPa and 150 to 270, respectively, with increasing Ln content from 7 to 16 at%. The highest values of σ_f and H_v are considerably higher than those (530 MPa and 180) for conventional Al-base crystalline alloys. Similarly, the electrical resistivity also increases from 48 to 109 $\mu\Omega\text{cm}$ with an increase of 8 to 12%Ln. The glass formation and the compositional dependences of T_x , σ_f , H_v and ρ_{RT} are explained by the concept that the number of the attrac-

tive bonding between Al and Ln metals increases with an increase of Ln content, resulting in the decrease in diffusivity of the constituent atoms and the increases in both the temperature dependence of viscosity for the supercooled liquid and the hybridization between s-p electrons in Al and f-d electrons in Ln metals.

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