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Correlation Between Electronic Structure, Mechanical Properties and Stability of TE-TL Metallic Glasses*

Ramir Ristić,^{a,**} Emil Babić,^b Mirko Stubičar,^b and Ahmed Kuršumović^c

^a*Department of Physics, Josip Juraj Strossmayer University of Osijek, Trg Ljudevita Gaja 6, HR-31000 Osijek, Croatia*

^b*Department of Physics, Faculty of Science, University of Zagreb, Bijenička cesta 32, HR-10000 Zagreb, Croatia*

^c*Department of Materials Science, Cambridge University, Pembroke Street, Cambridge CB2 3QZ, United Kingdom*

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Abstract. Mechanical properties (Young's modulus and hardness) of TE-Ni,Cu (TE = Ti, Zr and Hf) amorphous alloys increase approximately linearly with Ni, Cu fraction, x , over a wide composition range (fraction of Ni, Cu atoms $\leq 66\%$). This correlates with the observed increase of the Debye temperatures with x and shows that the strength of interatomic bonding increases with x in these alloys. The thermal stability (e.g. the crystallization temperatures) of these alloys also increases with x , thus with increasing interatomic bonding. Since in all these alloys the electronic density of states at the Fermi level, $N(E_F)$, decreases linearly with x within the same x -range, a very simple relationship exists between the electronic structure and mechanical and thermal properties. We also estimate the mechanical properties of amorphous TEs (by extrapolating the properties of alloys to $x = 0$) and compare them with those of crystalline TEs.

Keywords: metallic glasses, electronic structure, mechanical properties, thermal stability, transition metals

INTRODUCTION

Amorphous TE-TL alloys (TE = Ti, Zr, Hf and TL = Co, Ni, Cu) have been extensively studied¹ and interest in these alloys further increased upon the discovery of TE-TL based bulk metallic glasses (BMG).² These alloys show wide glass-forming range (fraction of TL atoms usually 20–70 %) which enables detailed study of the change in the electronic structure and properties on alloying, thus a thorough comparison between the model and experiment.³

In nonmagnetic amorphous TE-TL alloys the properties that are directly related to electronic density of states (DOS) show often linear variation with TL fraction.^{1,3–7} This finding correlates with Ultraviolet photoemission spectroscopy (UPS) results^{1,8} which showed that in these alloys DOS at the Fermi level (E_F), $N(E_F)$, is dominated by TE d-states and is reduced on increasing TL fraction. Therefore, the effect of alloying with TL can be approximated with the dilution of amorphous TE.⁶ Thus, one can extrapolate the data for glassy TE-TL alloys to zero TL fraction in order to deduce the properties of amorphous (a) TE. This method has been used in order to obtain the $N(E_F)$ and probable local atomic structure of a-Ti and a-Zr.³ More recently the same approach has been used in order to

obtain the atomic volumes of a-TE⁷ and magnetic and superconducting properties of a-Zr.⁹ The obtained parameters for a-TE agree well with those calculated for cubic crystalline phases of these metals¹⁰ and are quite different from those of stable crystalline phases (hcp) of TEs. This result is plausible since the amorphous (random) atomic arrangement is incompatible with a high anisotropy of the hcp lattice.

Very recently, a simple relationship between the electronic structure (DOS) and mechanical (Young's modulus and hardness) and thermal properties of glassy Zr-TL alloys has been found.¹¹ This finding is quite surprising since the observations of a direct relationship between the mechanical properties (especially hardness) of materials and their electronic band structure are very rare.^{12,13} Therefore, it is of interest to check whether similar relationships exist in other glassy TE-TL alloys, or not. Unfortunately, most of the results for TE-TL metallic glasses are obtained on Zr-TL alloys, so that a comparison between the alloy systems based on different TE is rarely possible.^{3,7,14,15} Here, we report new results for mechanical properties (Young's modulus and hardness) of several Ti-Cu and Hf-Cu glassy alloys and combine these results with the literature data for mechanical properties, Debye temperatures³ and thermal stability of Ti-TL and Hf-TL alloys in order to see how

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 Dedicated to Professor Boran Leontić on the occasion of his 80th birthday.

** Authors to whom correspondence should be addressed. (E-mail: ramir.ristic@fizika.unios.hr)

a change of TE (Ti → Zr → Hf) affects these properties of glassy TE-TL alloys. We also estimate some parameters of hypothetical pure a-TE and compare them with those of crystalline TE.

EXPERIMENTAL

The $\text{Hf}_{100-x}\text{Cu}_x$ ($30 \leq x \leq 60$) and $\text{Ti}_{100-x}\text{Cu}_x$ ($x = 35$ and 50) glassy alloys were prepared in a form of ribbons by melt spinning of master alloys with a predetermined composition in either pure Ar¹⁶ or He¹⁷ atmosphere. Controlled casting conditions resulted in ribbons with nearly the same cross-sections ($\approx 2 \times 0.025 \text{ mm}^2$) and, thus, with probably the same degree of quenched in disorder. As-cast samples were used for actual measurements and their amorphousness was verified by X-ray diffraction.¹⁵

The microhardness, H_v , was measured at room temperature using a standard E. Leitz (Wetzlar, Germany) Miniload II apparatus supplied with a 136° diamond pyramid indenter.¹² A load of 0.981 N was used and the loading time was 10 s.¹¹ The average value of microhardness is obtained from ten or more indentations on each studied sample, and the standard deviation was about 5–10 % of the mean value. The measurements were performed on the shiny (upper) surface of the ribbons, but occasional checks on the lower surface (in contact with roller) gave the same H_v values. Altogether, six different alloys with $30 \leq x \leq 60$ were measured. The values of H_v measured in kgf/mm^2 were converted in Pa units by multiplying results in kgf/mm^2 with 9.81×10^6 .

The Young's modulus, E , was calculated from the relationship $E = \rho v_E^2$ where ρ is the density of the alloy and v_E the velocity of ultrasonic waves along the ribbon. v_E was measured with a pulse-echo technique at room temperature.¹¹ The density, ρ , of the samples was measured by the Archimedes method.⁶ The estimated error was around 2 %.

The experimental procedures used to obtain the literature data for mechanical properties and thermal stability can be found in original papers.

RESULTS AND DISCUSSION

As pointed out in the Introduction, the band structure of amorphous TE-TL alloys results in DOS with d-states of TE at E_F and those of TL well below E_F .^{1,8} Such DOS combined with weak tendency for the formation of chemical short range order (CSRO) at TL fraction $x \leq 65$ ^{3,7} yields approximately linear decrease of $N(E_F)$ with increasing x of TL.

In superconducting alloys the value of $N(E_F)$ can be obtained from the measurements of low temperature specific heat (LTSH) and superconducting transition

temperature, T_c .^{3,9,15} LTSH yields the Debye temperature, θ_D , and dressed DOS at E_F , $N_\gamma(E_F) = (1 + \lambda_{ep}) N(E_F)$ (with λ_{ep} the electron-phonon enhancement), while T_c via the McMillan equation¹⁸ enables the calculation of λ_{ep} . $N_\gamma(E_F)$ can also be calculated from the normal state resistivity, density, and the temperature dependence of the upper critical field, $H_{c2}(T)$.¹⁵ As shown in the upper part of Figure 1, $N_\gamma(E_F)$ ^{3,15} for all TE-TL glassy alloys decreases linearly with the increase of TL fraction. Since λ_{ep} also decreases approximately linearly³ with x the variation of $N(E_F)$ in TE-TL alloys is qualitatively the same as that of $N_\gamma(E_F)$ (Figure 1). The $N_\gamma(E_F)$ values in Figure 1 decrease in order Ti → Zr → Hf alloys which is mainly due to increasing bandwidth in the same order.¹⁰ At lower TL fractions (x) $N(E_F)$ values of Ti and Zr-base glassy alloys are sizably larger³ than those of pure crystalline (hcp) Ti and Zr.¹⁰ This enhancement of $N(E_F)$ in glassy state of TE is due to absence of local minimum in DOS at E_F which occurs in stable (hcp) phases of crystalline TE and is absent in cubic phases (fcc, bcc) of the same metals.¹⁰ Thus, the

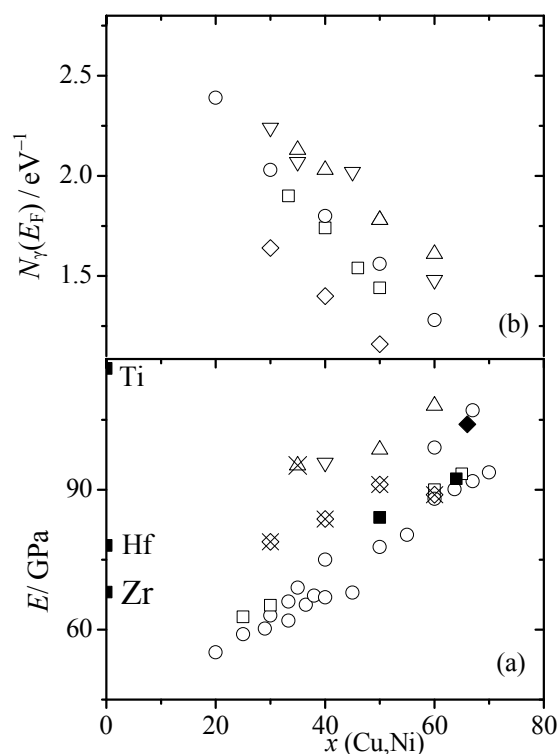


Figure 1. a) Young's modulus E of $\text{Zr}_{100-x}\text{Cu}_x$ (\square),¹¹ $\text{Zr}_{100-x}\text{Ni}_x$ (\circ),¹¹ $\text{Ti}_{100-x}\text{Cu}_x$ (\triangle),¹⁹ $\text{Ti}_{60}\text{Ni}_{40}$ (∇),²⁰ $\text{Hf}_{100-x}\text{Cu}_x$ (\otimes), $\text{Ti}_{65}\text{Cu}_{35}$ (\otimes) and (full symbols) of bulk metallic glasses $\text{Hf}_{34}\text{Cu}_{66}$ (\blacklozenge),²¹ $\text{Zr}_{50}\text{Cu}_{50}$ (\blacksquare)²² and $\text{Zr}_{36}\text{Cu}_{64}$ (\blacksquare)²³ alloys vs. fraction of Cu, Ni atoms, x . E of pure (a) Ti, Zr and Hf are also shown. b) Dressed densities of states $N_\gamma(E_F)$ per atom of $\text{Zr}_{100-x}\text{Cu}_x$ (\square),³ $\text{Zr}_{100-x}\text{Ni}_x$ (\circ),³ $\text{Ti}_{100-x}\text{Cu}_x$ (\triangle),³ $\text{Ti}_{100-x}\text{Ni}_x$ (∇)³ and $\text{Hf}_{100-x}\text{Cu}_x$ (\diamond)¹⁵ alloys vs. fraction of Cu or Ni atoms, x , expressed in percents.

absence of a hcp atomic arrangements in a-TEs and their alloys has profound influence on their DOS and interatomic bonding, hence affects all properties of glassy TE-TL alloys.^{3,7,9,11,15}

In Figure 1 we compare the variations of the Young's modulus, E , in TE-TL glassy alloys with TL fractions x (lower part of the Figure)^{11,19–23} with those of $N_V(E_F)$ with x (upper part). In all alloy systems a linear decrease of $N_V(E_F)$ (also $N(E_F)$ at least in Zr and Ti-base alloys)^{3,11} is accompanied with approximately linear increase of E with x . Thus, a simple relationship between DOS at E_F and stiffness (E) first observed in Zr-TL metallic glasses¹¹ seems to hold also for other TE-TL glassy alloys.

In binary amorphous metal-metal alloys such as the TE-TL glassy alloys the dense random packing of constituent atoms^{3,7,9} suggests direct link between their Young's modulus and strength of interatomic bonding.^{11,24} Thus, approximately linear increase of E with x implies an increase in the strength of bonding which correlates with the simultaneous decrease of DOS at E_F ($N_V(E_F)$, $N(E_F)$) with x (Figure 1). Accordingly, the enhanced DOS at E_F in a-TE yields Young's modulus which is strongly reduced with respect to that of corresponding crystalline TE (Figure 1). Therefore, in contrast to recent claim,²⁵ the values of E for TE-TL metallic glasses cannot be calculated as a suitable average of those for pure crystalline TE and TL. The estimated values for E of a-TEs (Figure 1) are some 50 % lower than those of corresponding crystalline TEs.

Since the amorphous alloys are approximately isostructural (there are no qualitative differences between different random atomic arrangements) their vibrational spectra should be quite similar,²⁴ thus their Debye temperatures, θ_D , should be directly related to the strength of interatomic bonding. Indeed as illustrated in Figure 2 the variations of θ_D for Zr-TL and Ti-TL glassy alloys with TL fraction are qualitatively the same as that of E with x (Figure 1) for the same alloy systems. The extrapolated values of θ_D for a-Zr and a-Ti are much lower than those of crystalline Zr and Ti (Figure 2) which demonstrates weak interatomic bonding in a-TEs. The rate of increase of θ_D with x in Ti-TL alloys is lower than that in Zr-TL alloys. This is due to increase of the average ionic mass with x in the former system.

The absence of extended crystal defects (thus, the absence of crystal-slip mechanisms)²⁶ in metallic glasses leads to very large hardness, H_V , and also to direct relationship between H_V and the strength of interatomic bonding. Thus, H_V of TE-TL glasses should increase with the increase of TL fraction, x , within the explored range. In spite of considerable scatter of the data (inherent to measurements of H_V) Figure 3 shows clearly that H_V increases with x (TL) for all TEs.^{11,20,21,23,27} Furthermore, for the same TL=Cu

fraction, the magnitudes of H_V of Ti-Cu and Hf-Cu glassy alloys are larger than those of corresponding Zr-Cu alloys which is consistent with higher E of the former alloys (Figure 1). Since in metallic glasses both Young's modulus and hardness reflect the strength of interatomic bonding there should be proportionality between H_V and E in these materials.^{11,20} Indeed, as shown in the inset to Figure 3 there is an approximate proportionality between H_V and E in TE-TL glassy alloys.^{11,19–21,23}

Since the amorphous alloys are macroscopically homogenous and isotropic systems, their elastic constants are correlated. Thus, the shear modulus, G , and the bulk modulus, B , can be calculated from known E and Poisson ratio σ .^{9,11} Furthermore, very little variation of σ in alloys²⁰ leads to proportionality between the elastic constants E , G and B in glassy alloys.^{9,11} Therefore,

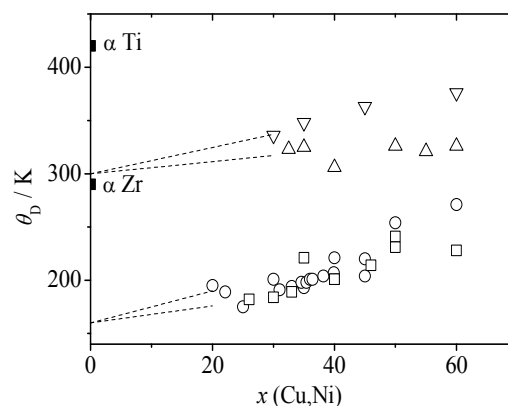


Figure 2. Debye temperatures of $Zr_{100-x}Cu_x$ (\square),³ $Zr_{100-x}Ni_x$ (\circ),³ $Ti_{100-x}Cu_x$ (\triangle)³ and $Ti_{100-x}Ni_x$ (∇)³ alloys vs. fraction of Cu or Ni atoms, x , expressed in percents. θ_D of pure (α) Ti and Zr are also shown.

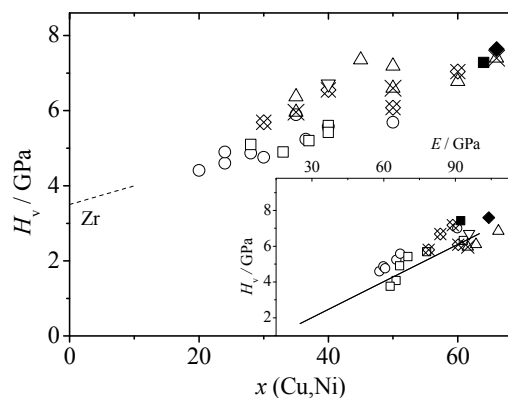


Figure 3. Microhardness H_V of $Zr_{100-x}Cu_x$ (\square),¹¹ $Zr_{100-x}Ni_x$ (\circ),¹¹ $Ti_{100-x}Cu_x$ (\triangle),²⁷ $Ti_{60}Ni_{40}$ (∇),²⁰ $Zr_{36}Cu_{64}$ (\blacksquare),²³ $Hf_{34}Cu_{66}$ (\blacklozenge),²¹ $Ti_{100-x}Cu_x$ (\otimes) and $Hf_{100-x}Cu_x$ (\otimes) alloys vs. fraction of Cu or Ni atoms, x , expressed in percents. In the inset microhardness H_V of $Zr_{100-x}Cu_x$ (\square),¹¹ $Zr_{100-x}Ni_x$ (\circ),¹¹ $Ti_{100-x}Cu_x$ (\triangle),¹⁹ $Ti_{60}Ni_{40}$ (∇),²⁰ $Hf_{100-x}Cu_x$ (\otimes), $Ti_{65}Cu_{35}$ (\otimes) alloys and (full symbols) of bulk metallic glasses $Hf_{34}Cu_{66}$ (\blacklozenge),²¹ and $Zr_{36}Cu_{64}$ (\blacksquare)²³ vs. E .

in TE-TL alloys H_v is proportional to all elastic constants and simultaneously all elastic constants and H_v show simple relationship with the electronic structure represented by DOS at E_F . This finding differs markedly from the situation encountered in TE carbonitrides¹³ where H_v is proportional to G only. As discussed elsewhere, this difference is associated with a more complex electronic structure of TE carbonitrides.¹¹ Taken together, all mechanical properties and Debye temperatures of TE-TL glassy alloys indicate large reduction in the strength of interatomic bonding on approaching pure a-TEs.

We expect that this progressive weakening of the interatomic bonding in TE-TL alloys on decreasing TL fraction (x) affects their thermal stabilities and glass-forming abilities.²⁸ Indeed, as shown in Figure 4 the variation of the first crystallization temperatures T_x of all glassy TE-TL alloys with x support this prediction. In particular, the available literature data^{11,27-31} for T_x decrease rapidly with decreasing the fraction of TL. The progressive decrease of thermal stability as $x \rightarrow 0$ (Figure 4) is accompanied with rapid decrease of glass-forming ability (GFA)^{11,20} with result that TE-TL metallic glasses with $x < 18$ have not been prepared so far.^{3,7,26} The findings from Figure 4 extend the simple relationship between the electronic structure (Figure 1) and mechanical properties also to thermal stability and GFA of TE-TL glassy alloys.

CONCLUSION

Recent analysis¹¹ of the relationship between the electronic structure and mechanical and thermal properties of $Zr_{100-x}TL_x$ glassy alloys ($20 \leq x \leq 70$) has been extended to include corresponding Ti-TL and Hf-TL alloys. New results for mechanical properties of these alloys have been combined with literature data in order to obtain an insight into the electronic structure/mechanical property relationship for all TE-TL metallic glasses.

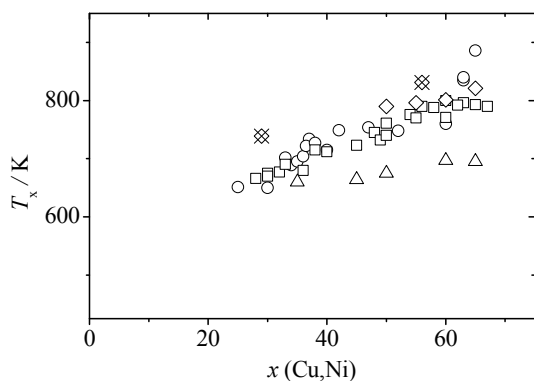


Figure 4. The first crystallization temperature T_x of $Zr_{100-x}Cu_x$ (□),^{11,30} $Zr_{100-x}Ni_x$ (○),^{11,31} $Ti_{100-x}Cu_x$ (△),²⁷ $Hf_{100-x}Cu_x$ (⊗)²⁹ and $Hf_{100-x}Cu_x$ (◇)²⁸ alloys vs. fraction of Cu or Ni atoms, x , expressed in percents.

Both Young's modulus and hardness (Figures 1 and 3) and the Debye temperatures (Figure 2) of TE-TL glassy alloys increase with the increase of TL fraction (x). Due to amorphous atomic structure and the absence of crystal slip all these properties reflect the strength of interatomic bonding. Thus, low extrapolated elastic moduli and Debye temperatures of amorphous (a) TEs compared to those of their stable crystalline modifications indicate very weak interatomic bonding in a-TEs, atypical of transition metals. This softening of the interatomic bonding on decreasing x probably affects the thermal stability (Figure 4) and the glass forming ability of TE-TL alloys which rapidly decrease as $x \rightarrow 0$.

Rather simple electronic structure of TE-TL metallic glasses^{1,3,8} results in simple variations of their several properties with the composition. This enables one to predict the properties of as yet unprepared amorphous alloys, including those of pure amorphous TEs. Since many bulk metallic glasses (BMG) are based on TE and TL components the results of systematic studies of binary TE-TL metallic glasses may be useful for the understanding and development of such BMGs.

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SAŽETAK

Međuovisnost elektronske strukture, mehaničkih svojstava i stabilnosti amorfnih legura ranih i kasnih prijelaznih metala

Ramir Ristić,^a Emil Babić,^b Mirko Stubičar^b i Ahmed Kuršumović^c

^a*Odjel za fiziku, Sveučilište Josipa Jurja Strossmayera Osijek, Trg Ljudevita Gaja 6, HR-31000 Osijek, Croatia*

^b*Fizički odsjek, Prirodoslovno-matematički fakultet, Sveučilište u Zagrebu, Bijenička c. 32, HR-10000 Zagreb, Hrvatska*

^c*Department of Materials Science, Cambridge University, Pembroke Street, Cambridge CB2 3QZ, United Kingdom*

Elastičnost i tvrdoća amorfnih slitina tipa TE-Ni,Cu (TE = Ti, Zr i Hf) rastu približno linearno s udjelom (x) Ni i Cu u širokom kompozicijskom području (udjeli Ni,Cu atoma, $x \leq 66\%$). Taj porast povezan je s porastom Debye-vih temperatura istih slitina s x te pokazuje da jakost međuatomske veze raste s x . Termička stabilnost tih slitina (npr. temperature kristalizacije) također raste s x dakle s međuatomskim vezanjem. Budući da u svim tim slitinama elektronska gustoća stanja na Fermijevoj razini $N(E_F)$ opada linearno s x , postoji vrlo jednostavna veza elektronske strukture s mehaničkim svojstvima i stabilnosti amorfnih slitina TE-Ni,Cu. Ocijenili smo i mehanička svojstva amorfnih TE (ekstrapolacijom svojstava slitina u $x = 0$) te ih usporedili s onima kristalnih TE.