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A Study of the Compressive Properties of Bulk Metallic Glass Composite

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Abstract

This study aims to investigates the plastic behavior and strength of tungsten wire-, molybdenum wire-, and tantalum wire-reinforced bulk metallic glass composites of $Cu_{50}Zr_{43}Al_{7}$, $Zr_{57}Nb_{5}Cu_{15}Al_{10}Ti_{12}$ and $Mg_{65}Cu_{25}Gd_{10}$. Bulk metallic glass materials are of brittle materials with low plasticity and high strength. Through reinforcing these materials with tungsten, molybdenum and tantalum wires, their plasticity and strength increases and decreases, respectively.The simulation of experiments using the Abaqus software in the present study showed that the reinforcement phase resulted in increased plastic deformation and decreased yield strength of bulk metallic glasses $Cu_{50}Zr_{43}Al_{7}$, $Zr_{57}Nb_{5}Cu_{15}Al_{10}Ti_{12}$ and $Mg_{65}Cu_{25}Gd_{10}$, among which the strength of those composites reinforced with tungsten wires, due to the high strength of tungsten, is very close to the strength of those in the matrix phase and a significant amount of plastic deformation is observed in them. The reinforcement phase of molybdenum increases the plastic deformation more than that of tungsten and tantalum. Tantalum, due to its low strength, reduces the total strength of the composite and, in turn, increases the plastic deformation of the bulk metallic glass.

Keywords: Tantalum, Molybdenum, Bulk Metallic Glass, Tungsten, Plastic Deformation

Introduction

Metal materials are generally recalled by a crystalline structure, i.e. the regular and symmetrical placement of atoms in a 3D sequence, but in 1691, Pol Duwez challenged this definition by the synthesis of alloys (au-25% atsi) in an amorphous state. Using an extremely rapid cooling technique (nearly a million degrees per second), he stopped the kinetics of crystallization and stabilized the melt in an amorphous form at ambient temperature. The amorphous phase inherits randomized placement of atoms from conventional glasses (oxide glasses) and non-directional metallic bonding from glassy metal (known as amorphous metal). Over the past five decades, a variety of glassy metals have been produced and investigated using a variety of techniques, indicating the attractiveness of this category of advanced materials to the researchers.Amorphous metals, due to good engineering properties, have been widely used in various fields such as sports equipment, precision gears, springs, fuel cells, electromechanical systems, and medical equipment. Favorable mechanical properties such as high strength, high elasticity, very high stiffness, very good resistance to corrosion and good magnetic properties have resulted in great interest to this newly developed material. Contrast these optimal properties, the dimensional limitations of amorphous metals, low fracture toughness and very low plasticity, as well as high cost of production, are of the challenges faced by the researchers.

Since the properties of glass matrix composites, as compared to single-phase metallic glass, have been upgraded using crystalline reinforcing phase and its fracture toughness has been enhanced, many researchers have been attracted to metallic glass composite. It has also been observed that using industrial raw materials, instead of high-purity raw materials, can effectively reduce the production cost without adversely affecting the glass-forming ability of the alloy. Glass matrix composites are classified into in situ composites and ex-situ composites (reinforced with external particles). In ex-situ composites, reinforcing particles, in the form of rods with a given diameter, are generally placed in the mold in parallel, and the glassy alloy is injected and frozen between these rods, but in-situ composites are created along with germination and crystalline phase inside the matrix alloy, which can take place before the freezing of glass matrix or after freezing and in separate processes.

Tungsten, molybdenum and tantalum wires have a good deformation and can increase the plasticity of composites by creating them with initial phase. Also, the amount or volume fraction of these reinforcing phases also greatly effective in increasing the plasticity of the bulk metallic glass. In present study, the volume fraction of these phases is assumed constant and equal to 40%.

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Bulk metallic glass

Glassy or amorphous or non-crystalline metal is referred to a metallic solid with no distinct atomic order and periodicity, and no single atomic structure can be considered for them. It should be noted that researchers distinguish between "glassy materials" and "amorphous materials", and define “glassy materials” as those "materials" frozen continuously from molten state to non-crystalline state, and “amorphous materials” as those materials reaching “non-crystalline state” using other methods such as vapor deposition or severe deformation.

Due to the need for a high cooling rate to prevent the crystallization kinetics, the glassy metals was first produced in the form of glass strips with a cross-sectional diameter of less than 1 mm, but when Chen reported that he froze the alloy in the form of long wires with 1 to 3 mm diameter and several centimeters length with glass phase by quenching in water, bulk metallic glass was first introduced in 1674. After him, Dorman and Kouee, in 1684, could increase the critical diameter to 11 mm for these alloys by removing the locations of heterogeneous germination. To do this, they etched the mold surface and fluxed it. In total, bulk metallic glasses are non-crystalline metallic solids that are continuously cooled from molten state to solid state and their cross-sectional diameter is at least a few millimeters.

Bulk metallic glasses have four distinct and unique characteristics, as compared to other glassy metals:

1. Their alloy system consists of at least three elements, and usually, the number of elements is more than 3. Although some reports have been published on the production of 2-element bulk metallic glass, but their cross-sectional diameter ranges 1-2 mm and crystalline phases are observed in their glass matrix.
2. They can be produced at low cooling rates. Generally, 100 k/s or less is required to stabilize the glass phase, but in some cases, very low cooling rates have led to the stability of glass phase in some alloy systems.
3. The main characteristic of these materials is their cross-sectional diameter, which is at least 1 mm and the largest cross-sectional diameter of the alloy was reported 72 mm.
4. These materials have a large undercooled molten zone, i.e. the difference between the glass transition temperature and the crystallization temperature in these materials is great (several dozens of temperature), which creates a good thermal stability.

- *The concept of glass-forming*

Figure 1 shows variation of specific volume as a function of temperature. When the temperature of a molten metal is reduced to a melting temperature, there is a sudden drop in volume, indicating the formation of a crystalline phase. The further reduction of the specific volume with temperature is dependent on the thermal expansion coefficient of the substance and has a slight slope.

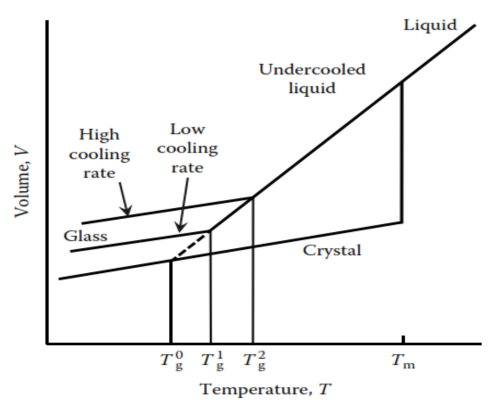

Fig. 1: Variation of specific volume against temperature for crystalline and glass freezing

Characterization of glassy alloys

When the alloy is frozen in glass form, it exhibits different characteristics than its crystalline state. Some of these features are:

The glossy and mirror-like appearance is the first and most distinctive feature of the non-crystalline structure. This glossy appearance is due to the lack of contraction of the material during freezing and the absence of formation of crystalline phases (Fig. 2). It should be noted that the transparency of metallic glass depends on their alloy system and is not the same for all bulk metallic glasses.

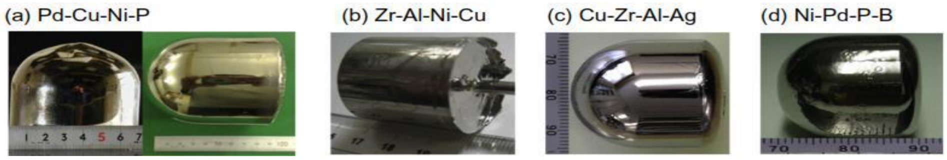

Fig. 2: The samples of bulk metallic glasses

The x-ray diffraction pattern of the amorphous phase looks like that of the molten metal pattern (Fig. 3). This pattern, which was first observed by Duwez, shows a wide peak that characterizes no particular crystalline phase and includes a range of angles. However, it should be kept in mind that the wide and short peak from the XRD of crust occurs in three different ways: 1) the glass phase; 2) the fine grain material; and 3) the glass phase with less than 5% crystalline phase inside it. However, in many reports, the glass phase is confirmed only by the XRD, but in case of higher sensitivity, other tests are also performed to confirm the glass phase.

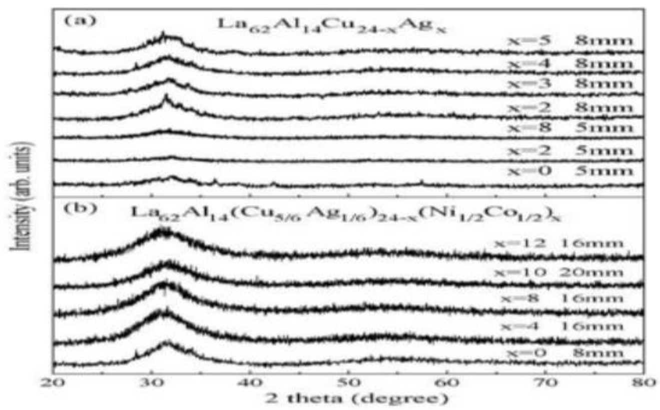

Fig. 3 - Different patterns for lanthanum-based bulk metallic glasses

Transmission Electron Microscopy (TEM) is an ideal tool for studying samples containing crystalline phase with very small dimensions, and it is possible to extract information such as the size, shape, and distribution of the crystalline phase through its image and the properties of its crystalline structure through its diffraction pattern. Also, thermal analysis is another method widely used for the characterization of bulk metallic glasses. DSC and DTA are the most widely used thermal analyses in the field of bulk metallic glasses, their main application is to determine the Tx, Tm, Tg temperatures of the amorphous alloys. Such measurements make it possible to calculate many parameters such as the ability of glass-forming and the thermal stability of the bulk metallic glass. It should be noted that the formation of amorphous phase is confirmed by comparing the graph of bulk metallic glass analysis and the graph of strip sample analysis. This is another application of the thermal analysis of the bulk metallic glasses.

The glass-forming ability

In order to make bulk metallic glasses industrially, it is essential to measure the glass-forming ability of different alloys. For this reason, many criteria have been presented for calculating the glass-forming ability of alloys by various researchers. In following, some of them are briefly discussed.

1. Critical cooling rate: The formation of glass phase is possible only by stopping germination and the growth of crystals, and rapid cooling is the simplest method to prevent the crystallization kinetics. Thus, the critical cooling rate is the cooling rate that can stop the crystallization kinetics and it depends on the alloy system and its composition (Fig. 4)

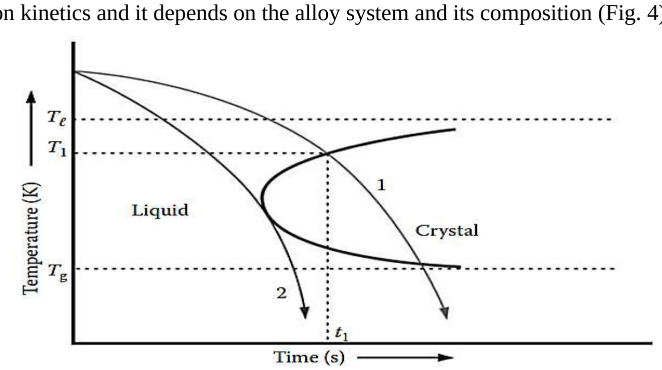

Fig. 4 - Schematic diagram for a hypothetical alloy, showing the critical cooling rate in a schematic form

2. Reduced glass transition temperature: To create a glass phase, the melt is cooled from the melting point to the glass transition temperature at high speeds. Based on Tranbol's suggestion, Tg to T1 ratio can be a good measure for calculating the glass-forming ability. This means that high Tg and low T1 indicate a high viscosity of the melt, and facilitate the quick reaching of the glass transition temperature. According to the Tranbol’s observations, there are alloys in which homogeneous germination of the crystalline phase is stopped, and in most alloys, at least Tg = 0.4 is necessary to create a glass phase.
3. Maximum diameter criterion: since it is required to cool all points of sample faster than the critical cooling rate to stabilize the glass phase, and the cooling rate is the function of the distance from the sample surface, the maximum diameter of the sample, which can be completely frozen in a glass form, can be a good criterion for investigating the glass-forming ability of different alloys.

But Inuoe’s criteria are considered to be the most comprehensive criteria. He has widely studied and investigated the glass-forming ability of different alloys and stated three general rules as the criteria for glass-forming ability:

- The alloy must be composed of at least three components and, increasing the number of elements must result in increased glass-forming ability.
- There should be a significant difference between the atomic sizes; it has been proposed that there must be at least 12% difference between the main components of the alloy in size.
- There must be is a negative mixing enthalpy between the main components of the alloy.

Production methods of bulk metallic glasses

The synthesis of the amorphous phase can take place in the vapor of metals, in the molten state and even in solid alloys, but metallic glass production generally takes place in the liquid phase. The synthesis of the metallic glasses is divided into two main stages of alloying and casting, each of which is briefly explained here.

- *alloying*

At this stage, the alloy is made of high-purity initial metals. Alloying is carried out to achieve a homogenous structure with a minimum amount of impurities and contamination that is suitable for re-melting and casting of metallic glass. To create a homogeneous structure, the melting and solidification processes of the main alloy are repeated 4 to 5 times in the furnace, and also to minimize impurities and contaminations, alloying must be performed according to the following steps with high-purity elements and using electric arc furnace equipped with a vacuum system or induction furnace equipped with a vacuum system.

1. To create a suitable vacuum in the furnace chamber. For this, two rotary and diffusion pumps are usually used.
2. To blow neutral gas with very high purity into the chamber. The presence of gas is required to establish electric arc, so, after creating vacuum, the argon gas with about 99.999 purity is usually blown in to the chamber.
3. To use proper de-oxygenator. Before melting the main alloy, elements such as titanium and zirconium, which have a high oxidation tendency, are molten in the furnace to absorb the remaining oxygen.

- *Casting*

After alloying, the casting of glass samples is carried out. Casting techniques vary depending on the shape, the glass-forming ability of the alloy, the diameter of the sample, and so on. Three widely used casting techniques of bulk metallic glasses are, including:

1. Quenching in the water
2. Casting in copper mold
3. Suction casting

Mechanical properties of bulk metallic glasses

Mechanical properties play a very important role in the industrialization of materials, so, characterization of mechanical properties of the material has been of great importance from long time ago. The different mechanical properties of the bulk metallic glasses are the main attractiveness of these materials to the researchers (Fig. 8.2). they are due to uncommon deformation mechanisms, in which there are no dislocations (the main factor responsible for the flow of plastic in crystalline materials). The unique properties of bulk metallic glasses can be divided into four categories:

1. Very high compressive strength
2. Low Young’s modulus and high elasticity
3. Tensile underload, almost without tolerance and plasticity
4. High stored elastic energy

- *Deformation and fracture behaviors of bulk metallic glasses*

The deformation of the bulk metallic glass can be divided into homogeneous and heterogeneous categories. The heterogeneous deformation occurring at Tg less than about 0.5, is along with local deformation in the shear bands, which takes place in the direction of maximum shear stress and at an angle of 45° to loading direction, while in homogeneous deformation occurring at Tg higher than 0.5, the entire volume of the material experiences plastic deformation.

Glass-matrix composites

Due to high strength and wide elastic strain range, bulk metallic glasses are suitable for the metal-metal composites. Also, the limited plasticity of these materials has been somewhat met by creating composite microstructures including glass matrix with the second crystalline phase. Two types of composites have been produced and tested by the researchers: in-situ and ex-situ composites.

- *Composites with external reinforcement*

Ex-situ composites reinforced with reinforcing particles such as wires are compared with single-phase bulk metallic glasses.

Effects of reinforcement on mechanical properties of composites

The toughness of bulk metallic glasses can be increased by adding an external reinforcing particles with appropriate volume fraction and size. Moreover, the strong interface between the matrix and the reinforcing phase must be considered. On average, bulk metallic glasses exhibit only 0.5% plastic strain, while ex-situ composites can exhibit up to 16% plastic strain. A pressure test carried out on zirconium-base bulk metallic glass reinforced with external particles, shows the plastic strain.

- *Plasticity enhancement mechanisms in composites*

It has been found that the fracture mechanism is affected by the reinforcing particles; the single-phase bulk metallic glasses fractures after the growth of initial shear band, but with the presence of tungsten particles, composite has a fracture surface like what observed in ductile materials. As a result, a change in the fracture surface along with the increased plastic strain shows the increased toughness of these composites.

Method

Tensile and pressure testing is one of the ways to measure strength and plasticity of materials. The yield strength, ultimate strength, plastic strain and failure strain can be calculated by making standard samples (with given diameter and height) and using tensile testing device. Using Abaqus software, we modeled a sample using a Abaqus software. The sample is a cylinder of 100 diameter and 10 radius. It also includes the main bulk. Four cylinders with a 40% volume fraction were used inside the main bulk including the reinforcing phase (Figure 5).

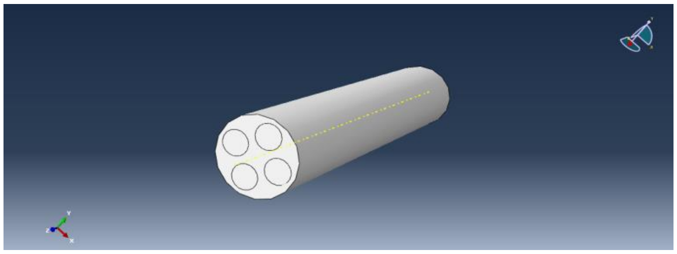

Fig. 5 - The prototype taken from the software

In this study, 3 materials of $Cu_{50}Zr_{43}Al_{7}$, $Zr_{57}Nb_{5}Cu_{15}Al_{10}Ti_{12}$ and $Mg_{65}Cu_{25}Gd_{10}$ were used as bulk metallic glasses and 3 materials of W, Mo and Ta as a reinforcing phase.

Boundary conditions

In the software, by applying the conditions, the force was applied only in one axis. Then, the sample was meshed to obtain the mechanical properties for each element (Fig. 6).

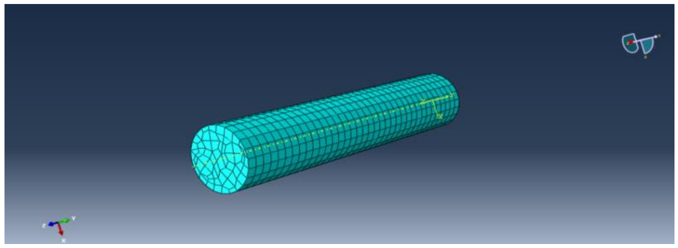

Fig. 6 - Taking the element from the sample

Finally, using the obtained results, the stress-strain graph was plotted and analyzed (Fig. 7).

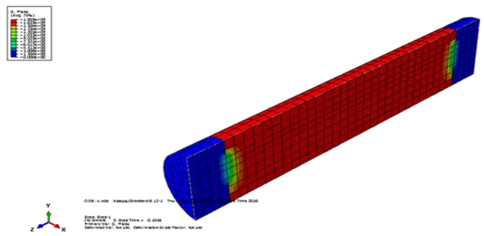

Fig. 7 – Output of the sample

Result Analysis

Investigation of pressure stress-strain curve

As stated above, bulk metallic glasses are brittle materials of bulk materials and have very little plastic deformation (about 2%). Through compositing and combining them with metals such as tungsten, molybdenum and tantalum, their plastic deformation can be increased.

Table 1 - Physical and mechanical properties of primary and reinforcing phrases

Elastic modulus (E) (GPa)		Poisson's ratio (ν)	Yield strength, (σY) (MPa)
Vit106 85		0.38	1800
Cu50Zr43Al7 122		0.36	2000
Mg65Cu25Gd1050.6		0.36	2000
Mo	330	0.38	400
Ta	186	0.35	350
W	410	0.28	

Bulk metallic glass vit106 ($Zr_{57}Nb_5Cu_{15}Al_{10}Ti_{12}$)

Vit106 has a chemical composition of $Zr_{57}Nb_5Cu_{15}Al_{10}Ti_{12}$. The physical properties of this BMG and the reinforcing phases are listed in Table 2. Through pressure testing, the stress-strain curves for Vit106, Mo, Ta, W are shown in Fig. 8.

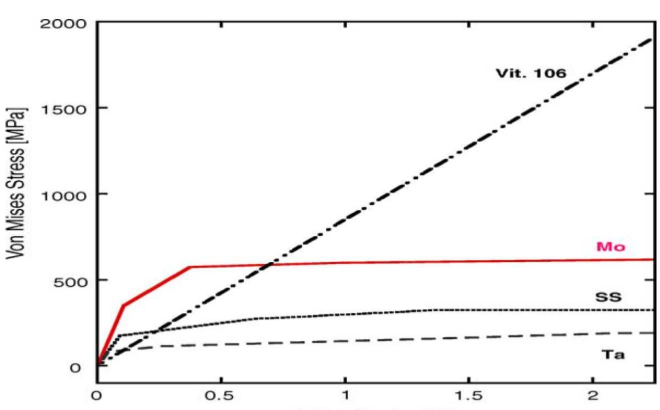

Fig. 8 - True stress-strain curves for Vit106, Mo, Ta, SS

By making the composite and adding reinforcing phases with a 40% volume fraction and performing stress-strain test, new stress-strain curves were obtained (Figure 9).

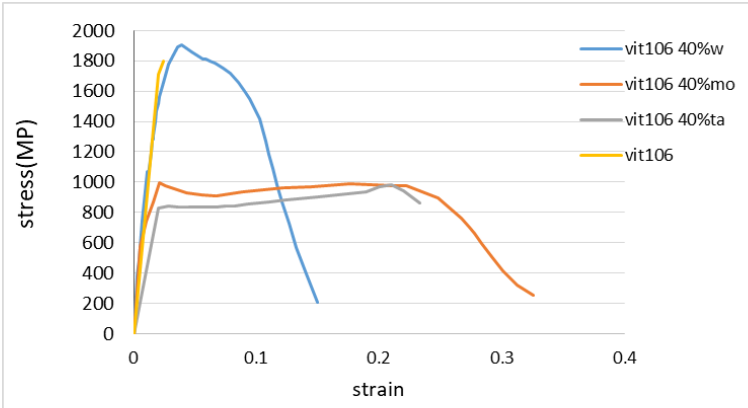

Fig. 9 - Stress-strain curves for Vit106, Vit106 40% W, Vit106 40% Mo, Vit106 40% Ta

Also, by calculating the yield point and the plastic strain of each composite, figures 10 and 11 were obtained.

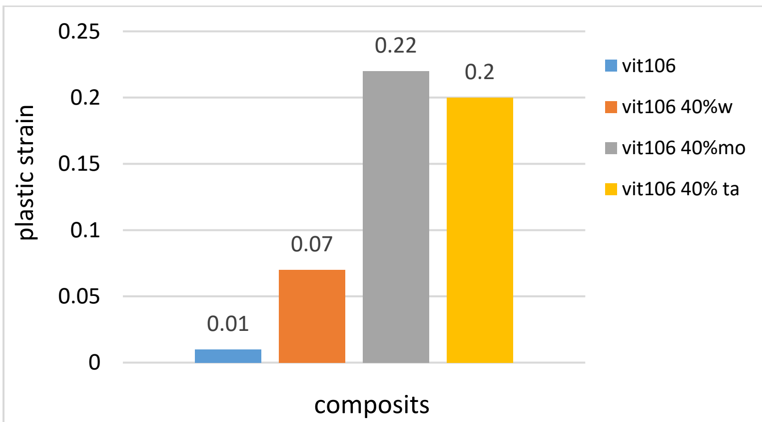

Fig. 10 - Plastic Strain diagram for Vit106, Vit106 40% W, Vit106 40% Mo, Vit106 40% Ta

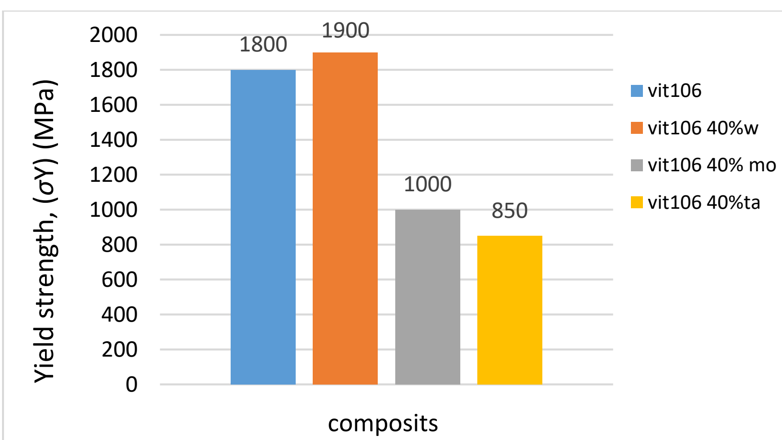

Fig. 11 - Yield stress diagram for Vit106, Vit106 40% W, Vit106 40% Mo, Vit106 40% Ta

According to above results and curves, it is found that with a 40% volume fraction, the reinforcing phase increases the plastic deformation of vit106. On the other hand, due to low strength of Mo and Ta and elasticity of these metals, the strength of Vit106 decreases and its plasticity increases significantly. Tungsten, due to its high strength, not only does not reduce the strength of the composite, but also increases it somewhat, but due to its low plasticity, it cannot increase the overall deformation of the composite.

Bulk metallic glass ($Cu_{50}Zr_{43}Al_7$)

The alloy has a chemical composition of $Cu_{50}Zr_{43}Al_7$, and the physical and mechanical properties of this BMG and its reinforcing phases are listed in Table 1. By conducting the pressure test, the stress-strain curve of this alloy is obtained, as shown in Fig. 12.

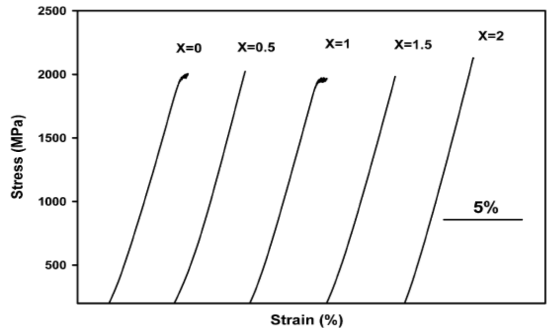

Fig. 12 - Stress-strain curve for $Cu_{50}Zr_{43}Al_7$

By making the composite and adding reinforcing phases with a 40% volume fraction and conducting stress-strain test, new stress-strain curves are obtained. (Figure 13)

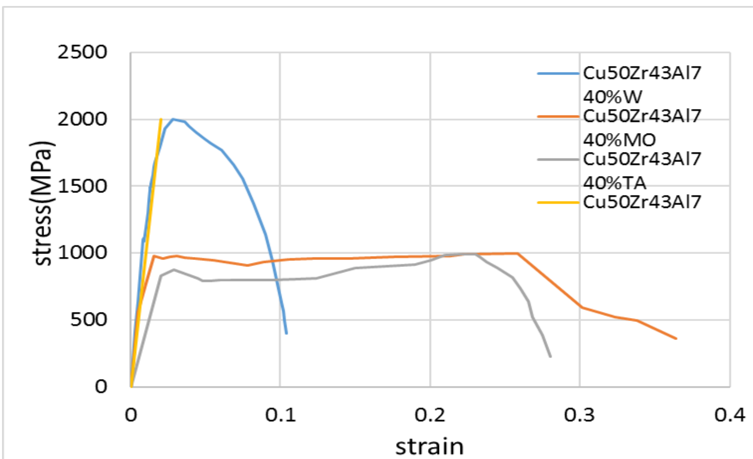

Fig. 13 - Stress-strain curves for $Cu_{50}Zr_{43}Al_7$, $Cu_{50}Zr_{43}Al_7$40% W, $Cu_{50}Zr_{43}Al_7$40% Mo, $Cu_{50}Zr_{43}Al_7$40% Ta

Also, by calculating the yield point and the plastic strain of each composite, figures 14 and 15 were obtained.

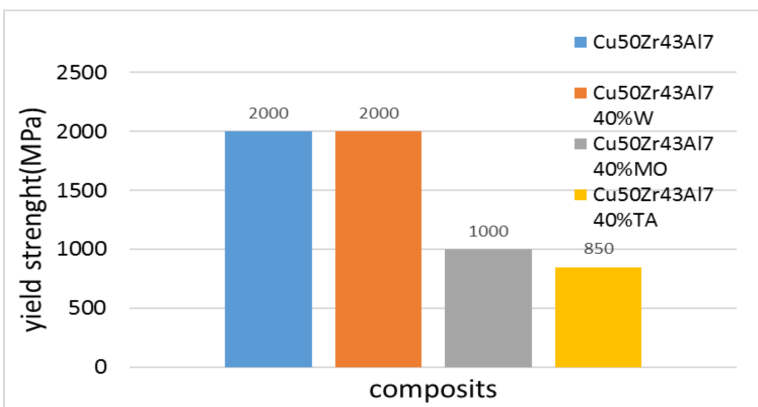

Fig. 14. Yield stress diagram for $Cu_{50}Zr_{43}Al_7$, $Cu_{50}Zr_{43}Al_7$40% W, $Cu_{50}Zr_{43}Al_7$40% Mo, $Cu_{50}Zr_{43}Al_7$40% Ta

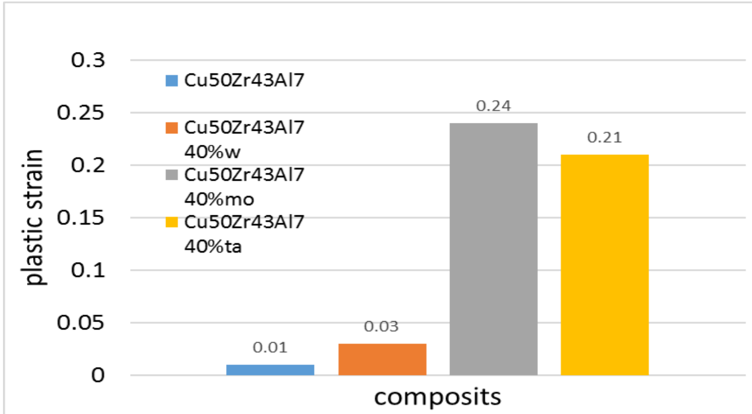

Fig. 15. Plastic strain diagram for $Cu_{50}Zr_{43}Al_{7}$, $Cu_{50}Zr_{43}Al_{7}$40% W, $Cu_{50}Zr_{43}Al_{7}$40% Mo, $Cu_{50}Zr_{43}Al_{7}$40% Ta

Using above results and curves, it was found that with a 40% volume fraction, the reinforcing phase increases the plastic deformation of $Cu_{50}Zr_{43}Al_{7}$. On the other hand, due to the low strength of Mo and Ta and the elasticity of these metals, the strength of $Cu_{50}Zr_{43}Al_{7}$ decreases and its plasticity increases significantly.

Bulk metallic glass ($Mg_{65}Cu_{25}Gd_{10}$)

This alloy has chemical composition of $Mg_{65}Cu_{25}Gd_{10}$. The physical properties of this BMG and its reinforcing phases are listed in Table 1. By making the composite and adding reinforcing phases with a 40% volume fraction and performing stress-strain test, new stress-strain curves were obtained (Fig. 16).

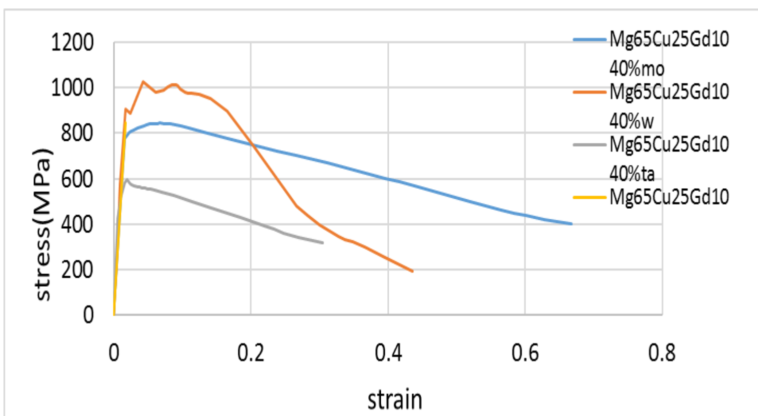

Fig. 16 - Stress-strain curves for $Mg_{65}Cu_{25}Gd_{10}$, $Mg_{65}Cu_{25}Gd_{10}$40% W, $Mg_{65}Cu_{25}Gd_{10}$40% Mo, $Mg_{65}Cu_{25}Gd_{10}$40% Ta

By calculating the yield point and the plastic strain of each composite, figures 17 and 18 were obtained.

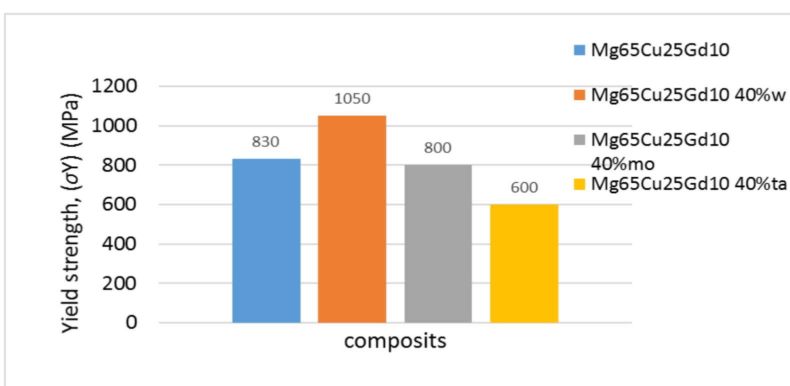

Fig. 17. Yield stress diagram for $Mg_{65}Cu_{25}Gd_{10}$, $Mg_{65}Cu_{25}Gd_{10}$40% W, $Mg_{65}Cu_{25}Gd_{10}$40% Mo, $Mg_{65}Cu_{25}Gd_{10}$40% Ta

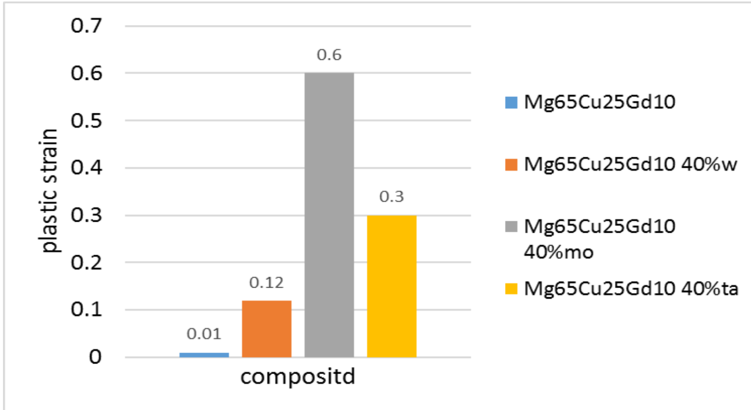

Fig. 18 - Plastic strain diagram for $Mg_{65}Cu_{25}Gd_{10}$, $Mg_{65}Cu_{25}Gd_{10}$40% W, $Mg_{65}Cu_{25}Gd_{10}$40% Mo, $Mg_{65}Cu_{25}Gd_{10}$40% Ta

According to above results, it was found that with a 40% volume fraction, reinforcing phase increases the plastic deformation of $Mg_{65}Cu_{25}Gd_{10}$. On the other hand, due to the low strength of Mo and Ta and the elasticity of these metals, the strength of $Mg_{65}Cu_{25}Gd_{10}$ decreases and its plasticity increases significantly. Also in this alloy, tungsten has increased the plastic deformation of the composites by 12%.

Conclusion

In this study, the plastic behavior and strength of $Cu_{50}Zr_{43}A_{l7}$, $Zr_{57}Nb_5Cu_{15}Al_{10}Ti_{12}$, $Mg_{65}Cu_{25}Gd_{10}$ bulk metallic glass composites reinforced with tungsten, molybdenum and tantalum wires were investigated using Abaqus software. After modeling in Abaqus software, following results were obtained:

- Bulk metallic glasses are brittle material with very low plastic deformation.
- Bulk metallic glasses have high strength, low density and low Young's modulus.
- Through reinforcing bulk metallic glasses with metals such as tungsten, molybdenum and tantalum, their plasticity can be enhanced.
- Tungsten, due to its high strength, not only does not reduce the composite strength, but in alloys with low strength, it can increase the total strength of the composite. On the other hand, due to their low plasticity, the plasticity of the composite increases negligibly.
- Molybdenum, due to its high plasticity, increases the deformation of the composite but, due to its low strength, reduces the strength of the composite.
- Tantalum, due to its favorable plasticity, increases the deformation of the composite but, due to its low strength, reduces the entire strength of the composite.
- Molybdenum and tantalum have similarities in terms of physical and mechanical properties, but in fact, molybdenum increases the deformation of bulk metallic glass further.

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