Poisson Ratio of BMGs Zr-Ti-Al-Ni-Cu-Be with Variation in Fraction of Constituent Elements.

Patel ram Suthar[1,2]

[1]Associate Professor, Physics, Dr. Bhimrao Ambedkar Govt. College, Sriganganagar, Rajasthan, India.
[2]Research Scholar, Department of Physics, Shri JJTUniversity, Jhunjhunu, Rajasthan, India.
Email: prsuthara@gmail.com

Abstract

Bulk metallic glasses are unique materials that exhibit an amorphous glassy atomic structure, which is different from their counterparts' crystalline materials with well-ordered atomic arrangements. The amorphous structure of BMGs can give them distinct mechanical properties compared to crystalline materials. The Poisson ratio (ν) can vary widely depending on various factors of the BMG alloy, such as composition, cooling rate, alloying element and their fraction, atomic structure, processing condition, etc. The high strength of BMGs is associated with their frangibility. Most BMGs fail shortly after yielding, even though some of them show a small amount of plastic deformation. Numerically, Poisson's ratio is usually in the range of 0 to 0.5 in crystalline materials. Most common crystalline metals have a Poisson ratio of around 0.3. However, in BMGs, lack of regular crystal planes and due to their amorphous nature, the atomic arrangement can facilitate a greater degree of isotropic deformation behaviour, resulting in a leading Poisson ratio. The higher value of the Poisson ratio close to 0.5 is not abnormal for BMGs. A direct relationship appears between higher Poisson ratios and increased plasticity (ductility). In the case of BMGs, the pivotal ν value marking the transition between ductile and brittle behaviour falls within the range of 0.32 to 0.33.

Keywords: BMGs, amorphous alloys, Poisson ratio, ductility.

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Introduction

Bulk metallic glasses (BMGs) exhibit a remarkable array of advantageous physical and chemical attributes. Notably, they display exceptional wear resistance and heightened corrosion resistance. At elevated temperatures, their inherent flexibility comes to the forefront, while at lower temperatures, their strength reaches extraordinary levels. Intriguingly, akin to certain polymer materials, BMGs demonstrate a substantial elastic limit, nearly 2% of strain, setting them apart. In a striking contrast with their crystalline

counterparts, BMGs boast approximately 2 to 3 times the strength, an impressive enhancement.

These distinctive properties hold immense appeal for real-world applications, establishing BMGs as an innovative category of materials with both functional and structural utility. These characteristics, which are seldom found in crystalline materials, underscore the significance of understanding the complex phenomena and formation of glass in bulk metallic glasses. The amalgamation of scientific intrigue and technological promise has sparked widespread interest in BMG research endeavours.

The journey into BMGs began with their discovery in 1969 by Chen and Turnbull [2], with ternary Pd-Cu-Si alloys leading the way as the pioneering "bulk" amorphous alloy. These alloys showcased an exceptional critical cooling rate of approximately 102 K s-1. The momentum surged in the scientific community, particularly following the emergence of novel multi-component BMGs such as $La_{55}Al_{25}Ni_{20}$ (unveiled in 1989 by Inoue et al.) [3] and $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ (introduced in 1993 by Peker and Johnson) [4]. Notably, these alloys could be crafted at lower cooling rates through direct casting from molten liquids, marking a significant advancement in BMG fabrication techniques.

Poisson's Ratio

Poisson's ratio (ν) is the ratio of the transverse compressive stress (negative) and the longitudinal extensional stress (positive) when a material is subjected to an axial load. The deformation behaviour of bulk metallic glasses (BMGs) depends on Poisson's ratio; therefore, Poisson's ratio is an important mechanical property in BMGs. Bulk metallic glasses are unique materials that exhibit an amorphous glassy atomic structure, which is different from their counterparts' crystalline materials with well-ordered atomic arrangements. The amorphous structure of BMGs can give them distinct mechanical properties compared to crystalline materials. The Poisson ratio is one notable aspect of BMGs can be close to 0.5, which is remarkably higher than the typical values for crystalline materials.

Numerically, Poisson's ratio is usually in the range of 0 to 0.5 in crystalline materials. Most common crystalline metals have a Poisson ratio of around 0.3. However, in BMGs, lack of regular crystal planes and due to their amorphous nature, the atomic arrangement can facilitate a greater degree of isotropic deformation behaviour, resulting in a leading Poisson ratio. The higher value of the Poisson ratio close to 0.5 is not abnormal for BMGs.

This unique behaviour (higher value of Poisson ratio) has practical implications. For example, under the applied loads, materials possessing higher Poisson ratios exhibit reduced susceptibility to stress concentrations. This unique quality can prove advantageous by

minimizing the risk of crack initiation. Under some conditions, compared to crystalline materials, BMGs exhibit improved toughness and ductility with their high Poisson ratios. The Poisson ratio can vary widely depending on various factors of the BMG alloy, such as composition, cooling rate, alloying element and their fraction, atomic structure, processing condition, etc.

The high strength of BMGs is associated with their frangibility. Most BMGs fail shortly after yielding, even though some of them show a small amount of plastic deformation. Bulk metallic glasses do not normally show work-hardening characteristics, so they generally fail in ductility. Flexibility is not a clearly defined term. Resilience is sometimes described as beauty, and it is not absolute.

It is difficult to describe, as it has no scale and as the saying goes, “beauty is in the eye of the beholder”. However, if we define ductility to failure in terms of plastic stress, bulk metallic glasses are very ductile, regardless of their distribution. This is because most of the deformation is concentrated in the shear bands, where the plastic strain can be ≫100%. Despite all this, BMGs do not show any remarkable macroscopic flexibility. Thus, in the shear band, bulk metallic glasses showing flexibility cannot be called appropriately ductile over a wide range, under tension, there is no plastic strain leading to failure so, bulk metallic glasses are very brittle.

Bulk metallic glasses before failure in plasticity exhibit some amount of plastic strain, it seems to when they are tested in compression. Differences can be found in the way the shear bands are nucleated and propagated, which may account for this difference. During tensile testing, soon after the formation of the first shear band, crack initiation occurs and as a result, bulk metallic glasses show essentially zero plastic strain before failure and after that fail catastrophically. However, specimens loaded under constrained geometry (for example, compression) show some plastic strain before failure but quickly fail in an elastic, fully plastic manner by the formation and propagation of multiple shear bands.

The role of Poisson's ratio in ductility has been first described by Chen et al. [5] “It is the high Poisson's ratio (ν) that is responsible for the ductile behaviour of many metallic glasses”. The brittle behaviour and fracture strength of Fe-based bulk metallic glasses increase rapidly when ν decreases with decreasing temperature, as well as due to the relatively low ν (<0.40). Schroers and Johnson [6] followed Chen in noting that the plasticity of BMGs can be related to Poisson's ratio, based on observing their $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$ BMGs at 20% compressive plastic strain before fracture. This leads to the formation of

multiple shear bands and the improvement of global ductility and very high fracture toughness. Gu et al. [7] also noted that large plastic strain (of about 0.8%) was obtained in their $Fe_{65-x}Mo_{14}C_{15}B_6Er_x$ (x = 0–2 at.%) BMG alloys only when x = 0 and 0.15. The flexibility in other BMGs of this group was limited and was almost zero when there was a high amount of Er (with at% Er at x = 1-2). Thus, lower resilience was related to a smaller value of Poisson's ratio at higher values of x. In Zr–Cu–Ni–Al BMGs this relationship was again successfully confirmed by Liu et al. [8]. By compositional changes, these authors produced these BMG alloys with a high Poisson's ratio. These authors produced these BMG alloys, by compositional changes, with high Poisson ratios. It is shown that superplastic elongation under compression tests, of up to 160% could be accomplished at room temperature. Yu and Bai [9] synthesized a group of bulk metallic glass alloys with x = 0 and 4 to 10 of $(Cu_{50}Zr_{50})_{100-x}Al_x$ and evaluated their plastic elongation and elastic moduli. Of all these alloys tested, only the alloy $(Cu_{50}Zr_{50})_{95}Al_5$ (for x = 5) exhibited a maximum plastic strain of 16%. Other alloys in this group showed much less plastic strain towards failure. A similar trend was observed in the variation of Poisson's ratio with Al content in these BMG alloys towards higher and lower values of the maximum value (Table 1). Therfore, a very good correlation can be established in these bulk metallic amorphous alloys between plastic strain to failure and the Poisson ratio.

Large amounts of ductility have also been reported in other BMGs, like $Ni_{60}Pd_{20}P_{17}B_3$ by Kawashima et al. [10] Flexible BMGs with high mechanical strength are being actively explored. This is through the control of alloy components, alloy compositions, thermal treatment, mechanical treatment, preparation technique, irradiation, in-situ and ex-situ composite, and surface coating. Several factors have been reported to influence the ductility of BMGs, and these factors can be categorized into extrinsic, intrinsic, and semi-intrinsic types. The Poisson's ratio (ν = B/G, B, G are coefficient of elasticity) stands out as the most important intrinsic parameter.

A direct relationship appears between higher Poisson ratios and increased plasticity (ductility). In the case of BMGs, the pivotal ν value marking the transition between ductile and brittle behaviour falls within the range of 0.32 to 0.33. Moreover, a consistent pattern reveals enhanced ductility as ν values above 0.33 [11]. Several bulk metallic glasses (BMGs) and their constituent elements such as Pd, Pt, La, Zr, and Ti demonstrate notable ν values above 0.33. However, BMGs based on Co, Fe, and Mg lack distinct plasticity/ductility and have low ν values lower than 0.32 [11]. Yokoyama et al. [12] have presented a comprehensive

investigation into the relationship between the ν value and plasticity, encompassing significant Zr–Al–Ni–Cu quaternary BMGs.

Analysis of Various Zr-Ti-Al-Ni-Cu-Be BMGs

The crystal structure of constituent elements Zr, Ti, Al, Ni, Cu, and Be is hcp, hcp, fcc, fcc, fcc, and hcp respectively.

The coefficient of thermal expansion (α) for Zr, Ti, Al, Ni, Cu, and Be is 5.7, 8.6, 23.1, 13.4, 16.5, and 11.3 µm/(m.K) respectively. The thermal conductivity (W) for Zr, Ti, Al, Ni, Cu, and Be is 22.6, 21.9, 237, 90.9, 401, and 200 W/(m.K) respectively. The covalent radius (R) is 148, 136, 118, 121, 138, and 90 pm respectively.

The following formula is used to calculate the quantitative value of thermal expansion coefficient, thermal conductivity and covalent radius-

$$x = \sum_i f_i \, x_i$$

Where: x stands for coefficient of thermal expansion or thermal conductivity or covalent radius. $f_i$means the quantitative fraction of the quantity x_i in BMG.

The Poisson ratio of various BMGs with different compositions is shown in table 1.

S. No.	**BMG**	**ν**	**α** {µm/(m·K)}	**W** {W/(m·K)}	**R** {pm}	**reference**
1.	Zr	0.34	5.7	22.6	148	20
2.	Ti	0.32	8.6	21.9	136	20
3.	Cu	0.34	16.5	401	138	20
4.	Ni	0.31	13.4	90.9	121	20
5.	Al	0.35	23.1	237	118	20
6.	Be	0.032	11.3	200	90	20
7.	$(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{98}Al_2$	0.347	9.34007	109.147	127.418	14
8.	$(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{95}Al_5$	0.343	9.76129	113.06	127.13	14
9.	$(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{92}Al_8$	0.342	10.1825	116.974	126.841	14
10.	$(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{90}Al_{10}$	0.327	10.4633	119.584	126.649	14
11.	$(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{88}Al_{12}$	0.326	10.7441	122.193	126.457	14
12.	$(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{85}Al_{15}$	0.318	11.1654	126.107	126.169	14

Table 1 : Value of the α, W, and R for various Zr-Ti-Al-Ni-Cu-Be BMGs.

The fraction of the base element Zr is in a decreasing order in the BMGs with serial numbers 7 and 8, namely $(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{98}Al_2$ and $(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{95}Al_5$. The fraction of crystal structure similar to base element is decreasing and crystal structure other

than base element (fcc) is increasing. The value of the coefficient of thermal expansion (α) and thermal conductance (W) are in increasing order, while the covalent radius (R) is in decreasing order. The value of Poisson ratio of BMG $(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{95}Al_5$ is less than that of BMG $(Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5})_{98}Al_2$. So, for the base element of the BMG or for elements those have the same crystal structure as the base element, the quantitively fraction of such elements decreases. Meanwhile, the quantitively fraction of other elements with a crystal structure other than that of the base element increases. In this case, the change in Poisson ratio is inversely related to any two identical changes in the coefficient of thermal expansion, thermal conductivity, and covalent radius. This is also applicable for the BMGs of serial numbers 8 to 12 of table 1.

Analysis of Various Zr-Al-Ni-Cu-Be BMGs

S. No.	BMG	ν	α {μm/(m·K)}	W {W/(m·K)}	R {pm}	reference
1	$Zr_{60}Al_{10}Ni_{10}Cu_{15}Be_5$	0.381	10.11	116.5	137.9	14
2	$Zr_{55}Al_{10}Ni_{10}Cu_{15}Be_{10}$	0.36	10.39	125.37	135	14
3	$Zr_{52.5}Al_{10}Ni_{10}Cu_{15}Be_{12.5}$	0.355	10.53	129.805	133.55	14
4	$Zr_{50}Al_{10}Ni_{10}Cu_{15}Be_{15}$	0.343	10.67	134.24	132.1	14

Table 2 : Value of the α, W, and R for various Zr-Al-Ni-Cu-Be BMGs.

The fraction of the base element Zr is in a decreasing order in the BMGs with serial numbers 1 to 4 in table 2. The fraction of crystal structure similar to base element is remains unchanged. The value of the α and W are in increasing order, while the value of R is in decreasing order. So, if the quantitative fraction of the base element decreases, but the crystal structure remains unchanged in fraction. In this case, the change in the Poisson ratio is reversed to any two identical changes in the coefficient of thermal expansion, thermal conductivity, and covalent radius.

Analysis of Various Zr-Ti-Cu-Be-Al BMGs

S. No.	BMG	ν	α {μm/(m·K)}	W {W/(m·K)}	R {pm}	reference
1.	$Zr_{35}Ti_{30}Cu_{8.25}Be_{26.75}$	0.37	8.959	101.063	128.06	14
2.	$Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$	0.34	8.92	99.555	127.7	14
3.	$Zr_{33}Ti_{30}Cu_{7.5}Be_{27.5}Al_2$	0.332	9.268	103.843	127.1	14
4.	$Zr_{30}Ti_{30}Cu_{7.5}Be_{27.5}Al_5$	0.322	9.79	110.275	126.2	14

5.	$Zr_{25}Ti_{30}Cu_{7.5}Be_{27.5}Al_{10}$	0.312	10.66	120.995	124.7	14

Table 3 : Value of the α, W, and R for various Zr-Ti-Cu-Be-Al BMGs

The fraction of the base element Zr is remains unchanged in BMG of serial numbers 1 and 2 in table 3. The fraction of crystal structure similar to base element is increasing. So, if there is no change in the quantitative fraction of the base element, then the quantitative fraction of elements with the same crystal structure as the base element increases in the BMG. Meanwhile, the quantitative fraction of elements with other structures decreases. In this case, the change in the Poisson ratio corresponds to all identical changes in the coefficient of thermal expansion, thermal conductivity, and covalent radius.

The fraction of the base element Zr as well as the fraction of crystal structure similar to the base element is also decreasing in order in BMG of serial numbers 2 to 5 in table 3. So, if the quantitative fraction of the base element as well as the fraction of crystal structure similar to the base element is also decreasing in order. In this case, the change in the Poisson ratio is reversed to any two identical changes in the coefficient of thermal expansion, thermal conductivity, and covalent radius.

Analysis of Various Zr-Ti-Ni-Cu-Be BMGs

S. No.	**BMG**	**ν**	**α** {μm/(m·K)}	**W** {W/(m·K)}	**R** {pm}	**reference**
1.	$Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$	0.35	9.05925	106.537	127.61	13
2.	$Zr_{45.4}Ti_{9.6}Cu_{10.15}Ni_{8.6}Be_{26.25}$	0.35	9.2068	113.382	128.286	13
3.	$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$	0.352	9.486	116.547	129.32	13
4.	$Ti_{40}Zr_{25}Ni_{3}Cu_{12}Be_{20}$	0.354	9.507	105.257	129.59	14
5.	$Ti_{45}Zr_{20}Be_{35}$	0.36	8.965	84.375	122.3	14

Table 4 : Value of the α, W, and R for various Zr-Ti-Ni-Cu-Be BMGs.

The fraction of the base element Zr is in a decreasing order in the BMGs with serial numbers 1 and 2 in table 4. The fraction of crystal structure similar to base element is decreasing and crystal structure other than base element (fcc) is increasing. The value of the α, W, and R all are in increasing order. The value of the Poisson ratio of BMGs remains unchanged. But for BMGs with serial numbers 2 and 3, the fraction of crystal structure similar to the base element is increasing. Therefore, For the base element of the BMG or for elements that have the same crystal structure as the base element, the quantitively fraction of such elements increases. Meanwhile, the quantitively fraction of other elements with a crystal structure

other than that of the base element decreases. In this case, changes in the Poisson ratio correspond to similar changes in all three thermal expansion coefficients, thermal conductivity, and covalent radius. The same situation is obtained in s. no. 3 and 4, 5 and 2, and 5 and 3.

Analysis of Various Zr-Cu-Ni-Al BMGs

S. No.	BMG	ν	α {μm/(m·K)}	W {W/(m·K)}	R {pm}	reference
1.	$Zr_{65.025}Cu_{14.85}Ni_{10.125}Al_{10}$	0.375	9.82343	107.148	140.781	15
2.	$Zr_{62.325}Cu_{17.55}$ $Ni_{10.125}Al_{10}$	0.375	10.115	117.365	140.511	15
3.	$Zr_{62}Al_{10}Ni_{12.6}Cu_{15.4}$	0.378	10.0734	110.919	140.058	15
4.	$Zr_{61.88}Al_{10}Ni_{10.12}Cu_{18}$	0.375	10.1632	119.064	140.468	15
5.	$Zr_{61}Cu_{18.3}Ni_{12.8}Al_{7.9}$	0.37	10.0366	117.527	140.344	15
6.	$Zr_{60.525}Cu_{19.35}Ni_{10.125}Al_{10}$	0.374	10.3094	124.176	140.331	15

Table 5 : Value of the α, W, and R for various Zr-Ni-Cu-Al BMGs.

The fraction of the base element Zr is in decreasing order in the BMGs with serial numbers 1 and 2 in table 5. The fraction of crystal structure similar to base element is decreasing and crystal structure other than base element (fcc) is increasing. The value of α and W are in increasing order, while the value of R is in decreasing order, so as per BMGs of serial numbers 7 and 8 in table 1 the value of ν should increase. But, the value of ν is unchanged. The value of ν increases in BMGs with serial numbers 2 to 3 and the values of α, W, and R all are in decreasing order. So, the change in the ν reversed to all identical changes in the coefficient of α, E, and R. BMGs with serial numbers (3, 4), (4, 5), and (5, 6) do not comply with the conditions mentioned in the above cases. This may be because the crystal structure of all three elements except Zr is similar, and some part of the quantitative fraction is changing into these. If Cu is taken as the base element with a higher fraction of these, then the above conditions are followed.

Conclusion

Various Zr-Ti-Al-Ni-Cu-Be BMGs whose variation in ν with partial changes in a fraction of constituent elements can be determined using the following point.

(i) For the base element of the BMG or for elements that have the same crystal structure as the base element, the quantitively fraction of such elements decreases or remains unchanged. Meanwhile, the quantitively fraction of other elements with a crystal structure other than that of the base element increases. In this case, the

change in the Poisson ratio is inversely related to any two or all identical changes in the coefficient of thermal expansion, thermal conductivity, and covalent radius.

(ii) If there is no change in the quantitative fraction of the base element, then the quantitative fraction of elements with the same crystal structure as the base element increases in the BMG. Meanwhile, the quantitative fraction of elements with other structures decreases. In this case, the change in Poisson ratio is reversed to any two identical changes in the coefficient of thermal expansion, thermal conductivity, and covalent radius.

(iii) In BMG, that element is taken as the base element, whose quantitative fraction in BMG is maximum. But if there is no change in the base element, that element is taken as the base element from the rest of the other elements whose quantitative fraction is maximum in that BMG.

(iv)The points described above can also be applied to the case when there is a change in the fraction of the elements, but there is no consequential change in the crystal structure.

(v) Elements should be replaced partially by others.

Some BMGs get partially correct results. A mathematical relation can also be found for how much change will happen in value of ν with partial changes in a fraction of constituent elements, and these points can also be used to estimate the elasticity coefficients and crystallization temperature and glass transition temperature. Partially correct results are obtained in some BMGs, the estimation of the theoretical result can be improved by using Lennard-Jones potential, etc.

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