Effect of reinforcement volume fraction on the density & elastic parameters of BMG's matrix composites

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ملخص
قمنا بدراسة تأثير تغير نسبة الألياف المقوية على الكثافة و ثوابت المرونة الطولية للخليط نو ماتريكس من الزجاج المعدني ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$) المدعمة بألياف من : glass E, Fe, Mo, Ni, Cr, Mn, Nb, Cd, Pt, U, Cu ا و الـ Zr. وجدنا أن ثوابت المرونة لبعض الخلائط تتحسن بإضافة بعض الأنواع من الألياف المدعمة مثل الـ Mo, Pt, Cr, Fe, U الخ. بينما تتناقص هذه الثوابت بإضافة أنواع أخرى من الألياف المقوية مثل الزجاج E و الـ Cd. كما وجدنا أيضا أن هذه المعاملات تتغير وفقا للنسبة بين ثوابت المادة الأم و الألياف المقوية لها. وقمنا باستخلاص علاقة هامة تمكننا من حساب الكثافة و ثوابت المرونة للزجاج المعدني بدلالة النسبة المئوية للألياف المقوية.

الكلمات المفتاحية : *الزجاج المعدني السميك - الخلائط، الدعامة - ثوابت المرونة.*

Résumé
L'influence de la fraction volumique de renfort sur les densités et les modules élastiques longitudinales de composite unidirectionnel à matrice verre métallique massif ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$, Vit.1) renforcée par les fibres : glass E, Fe, Mo, Ni, Cr, Mn, Nb, Cd, Pt, U, Cu ou Zr, a été étudié. Il a été montré que les modules élastiques des composites à grande fraction volumique de renfort sont améliorés en ajoutant certains types de fibres telles que le Mo, Pt, Cr, Fe, U etc. Par contre, ces modules élastiques diminuent avec d'autres fibres comme le glass E et le Cd. Nous avons également trouvé que les variations de ces constantes élastiques dépendent du rapport des paramètres des matrices et de ceux des fibres. Nous avons pu déduire une relation entre les densités, les constantes élastiques et la fraction volumique de renfort.

Mots clés : *Verres métalliques massifs - Composites - Renfort - Constantes élastiques.*

Abstract
The influence of reinforcement volume percentage on the densities and longitudinal elastic modulus of unidirectional $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$, Vit.1, Bulk metallic glass matrix composites, reinforced with glass E, Fe, Mo, Ni, Cr, Mn, Nb, Cd, Pt, U, Cu or Zr fibers, has been investigated. It was found the elastic moduli of high volume fraction composites are improved with the introduction of certain materials reinforcement such as Mo, Pt, Cr, Fe, U, etc. However, they decrease with some other materials reinforcements, as in the case of glass E and Cd. Moreover, we showed that the densities and elastic moduli variations of these materials depend on the ratio between matrix and fiber parameters. Finally, we were able to deduce a relation between densities, elastic moduli and reinforcement volume fraction.

Keywords: *Bulk metallic glass – Composite - Reinforcement - Elastic constants*

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1. INTRODUCTION

Bulk metallic glasses, (BMGs), represent a relatively new class of materials. Their unique properties place them among significant engineering materials: very high strength (1.9 GPa) and fracture toughness (40-55 $MPa.m^{1/2}$), excellent wear and corrosion resistance and a high elastic strain limit (up to 2%) [1-8].

These advantageous mechanical properties have opened up several attractive options for possible applications in aerospace, naval, sports equipment, luxury goods, armor and anti-armor systems, electronic packaging, and biomedical devices [9-13].

The alloy $Zr_{41.25}Ti_{13.75}Cu_{12.5}Ni_{10}Be_{22.5}$, commonly referred to as Vitreloy. 1 (Vit.1) [14], is the first commercial BMG and the most attractive among all BMGs due to its excellent forming ability and mechanical properties [14, 15]. However, they have the tendency to fail catastrophically, when unconstrained, by shear bending after very limited plastic deformation at room temperature [16]. To avoid this behavior and to obtain more tolerant damage BMGs, they are usually reinforced with fibers or particulates [17-19].

Tensile ductility of Zr-based BMGs have been enhanced by including reinforcing fibers, ceramics or ductile particles such as of Mo, Nb, Pt or Ta to obtain a 17% improvement [17, 20, 21]. Fortunately, the development of Zr-based BMGs matrix composites opened new opportunities for metallic glass applications.

In this paper, we studied the effect of fibers volume fraction, Λ_f, effect on the densities and elastic constants of Vit.1 ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$) BMG's matrix composites with several ductile metallic elements Fe, Mo, Ni, Cr, Mn, Nb, Cd, Pt, U, Cu & Zr and the glass E, as unidirectional continuous fibers reinforcements. The aim is to investigate the elastic behavior, density and ductility of Vit.1 BMG's composites as function of wires reinforcement percentage.

2. THEORETICAL FORMULAE AND MATERIALS

In unidirectional composites, densities, ρ_c, longitudinal Young modulus, E_c, shearmodulus, G_c & Poisson ratio, Λ_c, can be written as a function of fibers volume percentage as follows[22]:

$$\rho_c = \frac{1}{\frac{\Lambda_f}{\rho_f} + \frac{(1-\Lambda_f)}{\rho_m}} \qquad (1)$$

$$E_c = E_f \Lambda_f + E_m (1-\Lambda_f) \qquad (2)$$

$$G_c = \frac{G_m G_f}{G_m \Lambda_f + G_f (1-\Lambda_f)} \qquad (3)$$

$$\nu_c = \nu_f \Lambda_f + \nu_m (1-\Lambda_f) \qquad (4)$$

with ρ_f, E_f, G_f and ν_f are the density, Young's and shear moduli, Poisson ratio of fiber; ρ_m, E_m, G_m and ν_m are those of matrix; Λ_f and ($1-\Lambda_m$) are the volume fraction of fiber and matrix composite, respectively. ρ, E and G of monolithic Vit.1, glass E, Fe, Mo, Ni, Cr, Mn, Nb, Cd, Pt, U, Cu, Zr at normal conditions of temperature and pressure are assembled in table 1.

Table 1. Densities and elastic moduli of Vit. 1 and various reinforcements at normal conditions of temperature and pressure.

Materials [Ref]	**ρ [kg/m^3]**	**E [GPa]**	**G [GPa]**	**ν**
Vit.1 [23]	6130	101.2	37.4	0.35
Zr [24]	6504	95.9	35.8	0.33
Cd [24]	8600	66.6	25.1	0.32
Pt [24]	21400	177	63.5	0.39
U [24]	19281	78.6	87.9	0.19
Ni [24]	8840	217	83.2	0.30
Cu [24]	8930	127	47.3	0.34
Cr [24]	7194	277	114	0.21
Nb [24]	8578	105	37.5	0.39
Mn [23]	7473	191	77.1	0.24
Mo [23]	10222	330	125.5	0.29
Fe [24]	7690	203	80	0.30
Glass E [22]	2600	74	30	0.25

3. RESULTS AND DISCUSSIONS

3.1 Individual Parameter Variations

The calculated elastic parameters of Vit.1 BMG's composites in various volume percentages of glass E, Fe, Mo, Ni, Cr, Mn, Nb, Cd, Pt, U, Cu & Zr (0%, 20%, 40%, 60%, 80% &100%), are used to plot in figure 1 their variations as a function of reinforcement volume fraction.

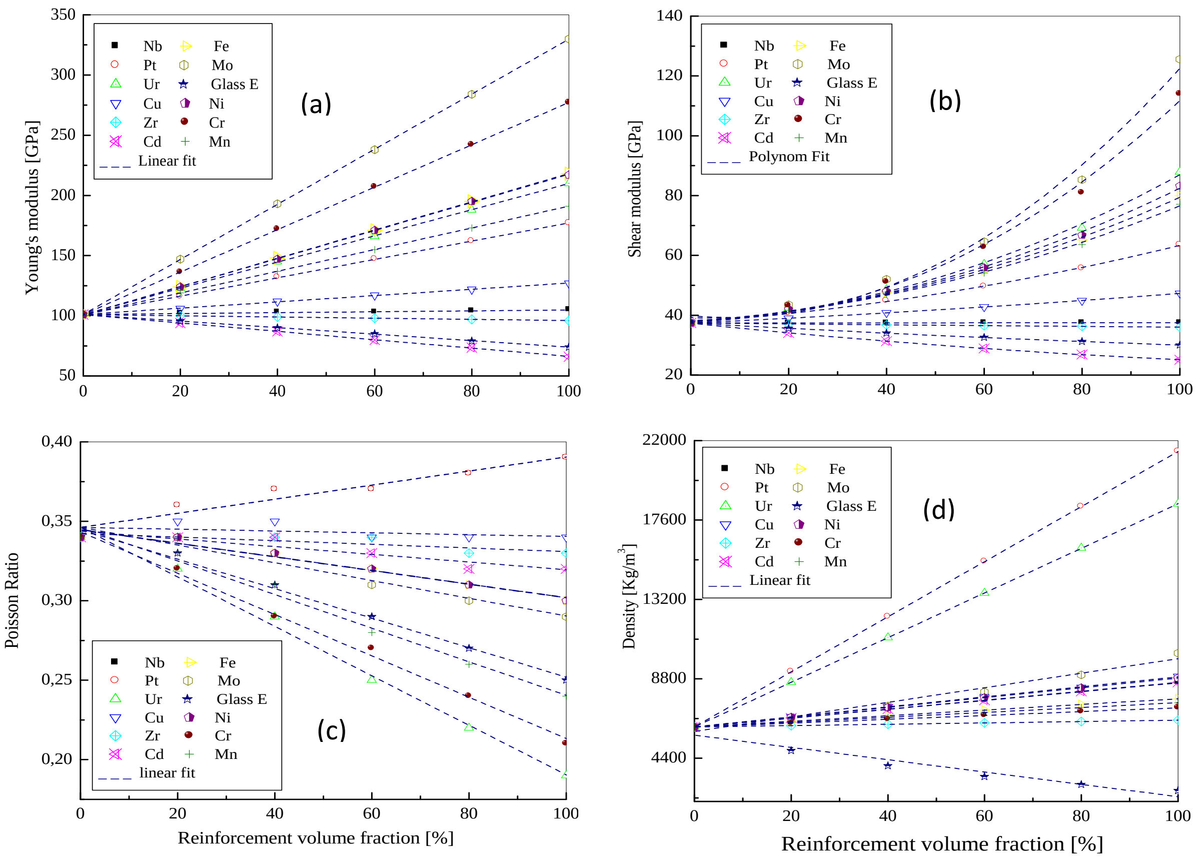

Figure 1. Vit.1 matrix composites density & elastic constants as function of reinforcement volume fraction of , glass E, Fe, Mo, Ni, Cr, Mn, Nb, Cd, Pt, U, Zr: (a) Young's modulus, (b) shear modulus, (c) Poisson ratio & (d) density.

In figure 1(a), it is clear that longitudinal composite's Young's modulus, E_c varies linearly with reinforcement volume fraction growing. This linear form is often obtained for different fiber-matrix combination [22]. The following remarks can be made:

- E_c decreases for Cd, Glass E and Zr fibers.
- E_c increases with Nb, Pt, U, Cu, Fe, Mo, Ni, Cr and Mn fibers.
- The Mo/Vit.1 composite takes the highest values of E_c, which increase from 101.2 GPa to 330 GPa.
- E_c of Cd/Vit.1 composite takes the lowest values down to 66.6 GPa.
- E_c of Composites with Zr and Cu reinforcement, with high percentages of matrix components, changed slightly with Λ_f.

Figure 1 (b) shows the effects of reinforcement volume fraction on shear modulus. An increasing behavior is obtained for Mo/Vit.1, Cr/Vit.1, U/Vit.1, Ni/Vit.1, Fe/Vit.1, Mn/Vit.1, Pt/Vit.1, Nb/Vit.1 & Cu/Vit.1 composites. Curve fitting gave a polynomial dependence of G_c as function of Λ_f. Composites with Mo fibers showed the highest values that approach 130 GPa. However, G_c of Zr/Vit.1, glass E/Vit.1 and Cd/Vit.1 composites decrease with Λ_f. Whereas, the Cd/Vit.1 composite, takes the lowest value.
We can conclude that composites with Mo, Cr, Ni, Fe, U, Mn or Pt fibers with high Young's and shear modulus have much higher tensile and yield strength in comparison with other studied composites due to the fact that high elastic moduli possess high tensile strength [25]. These results are in good agreement with experimental results [20], [26-28] which showed that Young's and shear modulus of Zr-based metallic glass matrix composites increase as function of wires reinforcement percentage such that the Mo, Pt, Cu or Nb. The published experimental value of Young's moduli (265 GPa) of Mo/Vit.1 composites (80% of Mo) [29] is not far from that presently calculated (170 GPa); The measured shear moduli (78 GPa) is comparable to the calculated value (82 GPa). The calculated Young's modulus for a 60 vol.% copper-fibers is 116 GPa, this value is close to that measured from the compression tests which revealed a Young's modulus of 110±1GPa [30].

Furthermore, published experimental studies [29] proved that elastic modili of Mo wires reinforced Zr-based BMG composites are higher than those of other wires reinforcement such that Nb, Pt or Cu, in agreement with our results. Moreover, any increase in elastic moduli leads to an increase in potential applications of BMG matrix composites as structural materials. For example, Zr/Vit.1, Nb/Vit.1, Cu/Vit.1 & glass E/Vit.1 with low value of Young's & shear modulus (>90 GPa & >35 Gpa) are good candidates for biomedical applications because of their elastic modulus which is close to that of human bone (~ 25 GPa & ~4 GPa) [31] and inferior than those of both monolithic BMG (~100 GPa & ~40 GPa) and conventional metallic biomaterials (~200 GPa & ~80GPa) [24].
This allows a good biocompatibility and good transfer of stress between bone and material.

To confirm such investigations, we plot in figure 1(c) Poisson ratio variations as function of Λ_f. High ductility is corresponding to low values of G/B (B is the bulk modulus) and so high values of Poisson's ratio [32]. In this curve, we observe a little change in Poisson ratio and so a little change in the ductility of 20% Cu, Nb or Mo containing Vit.1 composites. This observation was also reported in the literature [20, 33, 34]. The same observation was obtained for Zr/Vit.1, Pt/Vit.1 and Ni/Vit.1. Moreover, Pt/Vit.1 and Nb/Vit.1 composites Poisson ratio increases significantly with reinforcement volume fraction and their value varied from 0.34 to 0.39. However, ν_c of other composites studied decreases when Λ_f increases. This implied that the interface Pt-matrix & Nb-matrix is stronger than that of other fiber-matrix interface studied.

Figure 1(d) illustrates clearly that Vit.1 composites densities increase linearly with all metallic fibers when reinforcement volume fraction increase, whereas, it decreases with glass E fiber. The lowest value of ρ_c were obtained for the Pt/Vit.1 composite whose density attains 21000 kg/m^3.
Densities composites with glass E fibers decrease to very low value (~ 2000 kg/m^3). The increase of BMGs composites densities enhances their typical strength and hardness [35].

3.2. Relative Parameter Variations

We note that Vit.1 composites densities and elastic modulus studied depend on E, G & ρ matrix parameters and those of reinforcement. Therefore, we can represent the ratio between these parameters by a unique ψ_i parameter such that:

$$\psi_i = \frac{i_{matrix}}{i_{fiber}} \quad (\text{were i} = \text{E, G or } \rho) \qquad (5)$$

Hence, we obtain (i) for $\psi_i < 1$: composite properties values increase as function of reinforcement volume fraction and (ii) the inverse behavior for $\psi_i > 1$. We plot in figure 2 E, G & ρ composites variations as function of ψ_i ratio in the volume percentage range from 20% to 100 %. It can be noticed that in figure 2(a) composites Young's modulus decreases exponentially as the ψ_E ratio increases.

For ψ_E<1, This decrease of composites with 20% volume fraction is lower followed by 40%, 60%, 80% & 100% respectively. This arrangement will be inversed with ψ_E>1. We show in figure 2(b) composites shear modulus variations as function of ψ_G ratio. This elastic modulus decreases exponentially as ψ_G increase. This decrease is more important when fiber volume percentage increases. Moreover, we can deduce that Mo/Vit.1 composite's yield strength increases very significantly with Λ_f followed by Cr, Fe, Ni, U or Mn containingVit.1 composites. Whereas, composites with Pt, Cu, Nb, Glass E, or Cd show a slight change. The same observations were obtained for the Poisson's ratio in figure 2(c). Moreover, BMG's composites densities exhibit in figure 2(d) an exponential decrease with ψ_ρ ratio increasing. As shown, curve's fitting prove that elastic modulus and density varied exponentially with fiber volume percentage of the form:

$$Y = Y_0 + Ae^{-x/t} \qquad (6)$$

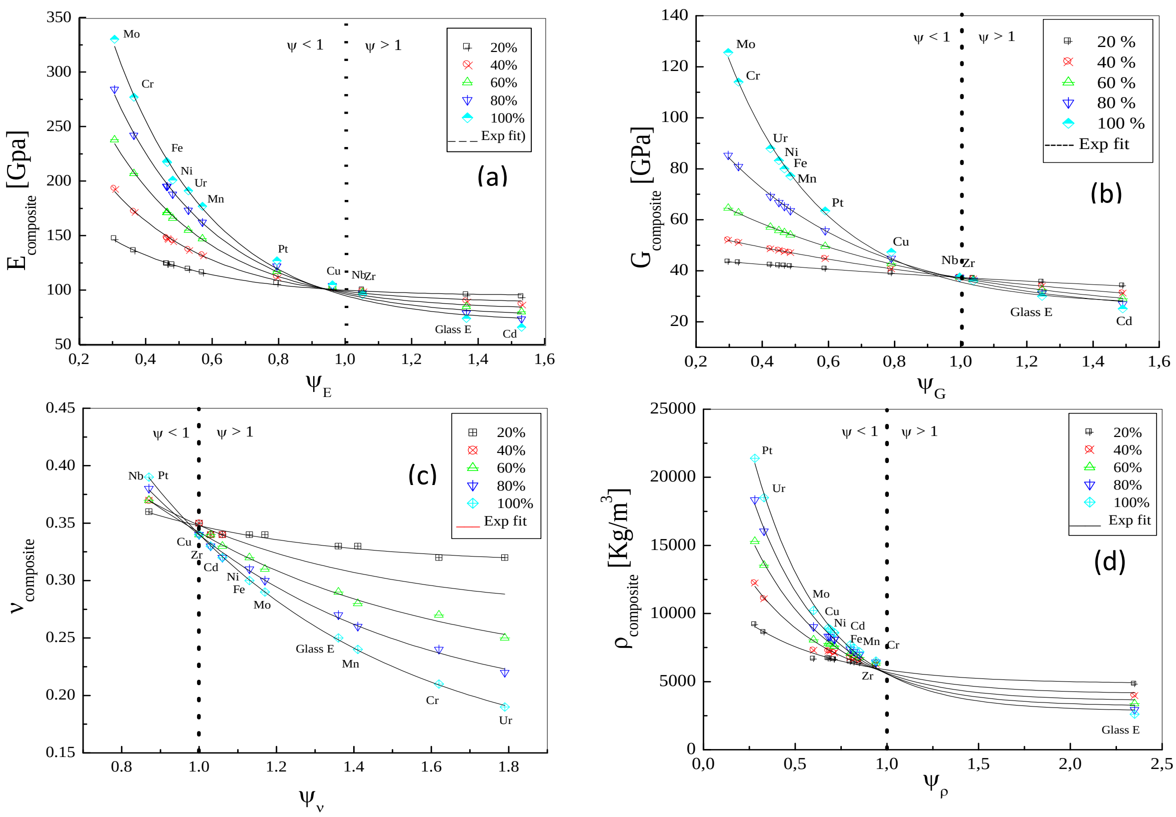

Figure 2. Vit.1 matrix composites mechanical properties variations as function of ψ ratio for various reinforcement volume fractions.(a) Young's modulus, (b) shear modulus, (c) Poisson ratio & (d) density.

3.3 General formulation of parameter variations

To obtain a general form of densities elastic modulus variations for Vit.1 matrix composites; we had to transform the exponential form to linear form. After neglecting Y_0, the exponential expression can be written as:

$$ln\,Y = ln\,A - \frac{x}{t} \qquad (7)$$

Afterwards, we plot in figure 3, the slopes $(\frac{Ln\ i_c}{\psi_i}\ where\ i = \rho, E\ or\ G)$ of these linear curves as function of reinforcement volume fraction.

Figure 3(a) illustrates clearly that for Young's modulus, the slopes curves $\frac{E_c}{\psi_E}$ are constants as reinforcement volume fraction increase according to the form:

$$\frac{ln\ E_c}{\psi_E} = -3.33\ \Lambda_f \qquad (8)$$

Figure 3(b) shows the variations of shear modulus slopes as function of reinforcement volume fraction. The dependence which is polynomial, takes the form:

$$\frac{lnG_c}{\psi_G} = -0.3 - 9.6\ \Lambda_f - 3.14\ \Lambda_f^2 \qquad (9)$$

Figure 3(c) illustrates polynomial variations of Poisson ratio with Λ_f according to the form:

$$\frac{ln\,\nu_c}{\psi_\nu} = -2.83 - 0.03\ \Lambda_f - 2.21\ \Lambda_f^2 \qquad (10)$$

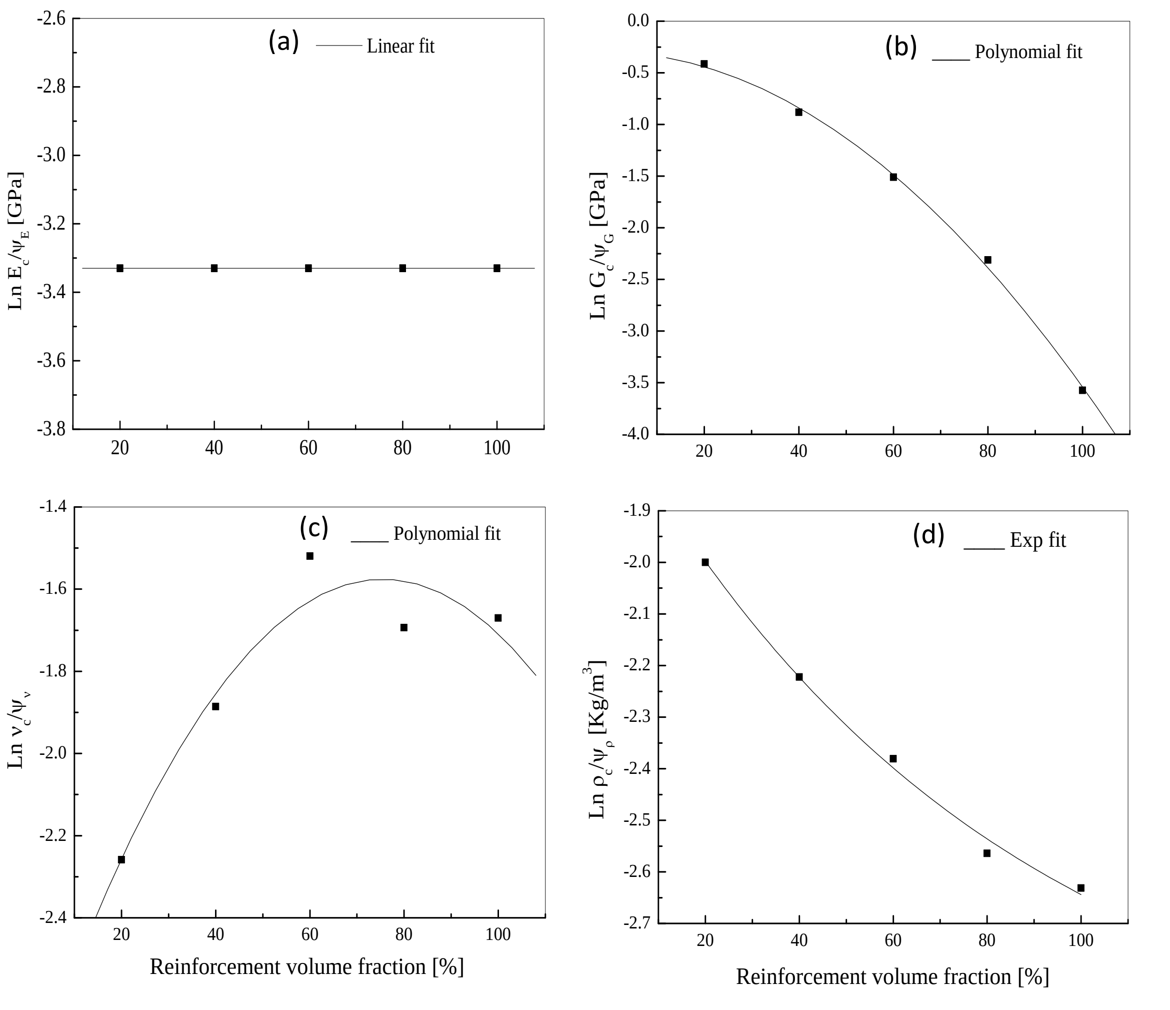

Figure 3. Vit.1 matrix composites slopes curves of fig. 2 variations as function of reinforcement volume fraction.

From figure 3(d), it is obvious that composites densities decrease exponentially with the of the increase of the reinforcement volume fraction. Their variation obeys the form:

$$\frac{ln\ \rho_c}{\psi_\rho} = -3.03 + 1.32\ e^{-\frac{\Lambda_f}{81.59}} \qquad (11)$$

From these relations, we can deduce the density; and elastic modulus of unidirectional $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ matrix composites for each Λ_f.

4. CONCLUSION

Densities, Young's, shear modulus and Poison's ratio of Vit.1/Zr, Vit.1/Cd, Vit.1/Cu, Vit.1/Ni, Vit.1/Ti, Vit.1/Mo, Vit.1/Mn, Vit.1/U, Vit.1/Pt, Vit.1/Cr, Vit.1/Cd and Vit.1/glass E BMGs matrix composites have been studied. It was found that the reinforcement of $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ BMG can improve the tensile strength (i.e., Mo, Cr and fibers Ni), the ductility and the toughness (i.e., Pt & Nb fibers). These behaviors are in agreement with that published for experimental results which revealed that Zr-based BMGs matrix composites are improved when reinforced by several fibrous reinforcement such that Mo, Pt, Nb or Cr. We were able to determine a ψ parameter which presents a ratio between matrix and fiber parameters. It was found that for ψ inferior than 1, densities and elastic constants increase as a function of reinforcement volume fraction. While, they decrease when the ratio ψ is superior to unity. Moreover, empirical relations were deduced, this relationship gives composites mechanical parameters as function of ψ ratio and fibers volume percentage.
The present result is very important to determine BMGs matrix composites behavior in high and low value of reinforcement volume fraction. Furthermore, this result can be used in the selection of BMGs composites for different applications.

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Nomenclature

Symbol	**Parameter**	**unit**
ρ_c	Composite density	Kg/m^3
E_c	Composite Young's moduli	GPa
G_c	Composite shear moduli	GPa
ν_c	Poisson ratio	/
Λ_f	Reinforcement volume fraction	%