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Metallic glass/light alloy (MEGA) multimaterials elaborated by co-pressing at high temperature

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ABSTRACT

Metallic glass/light alloy (MEGA) multilayered materials are elaborated by co-pressing at high temperature. They constitute a layer of Zr-based bulk metallic glass inserted between two layers of either aluminium or magnesium alloys. The co-pressing tests are performed at various temperatures above the glass transition temperature of the glass under inert gas. The selected conditions for co-pressing are chosen from the knowledge of the rheological behaviour (deduced from strain rate jump tests) of both the metallic glass and the light alloys and also from information concerning the thermal stability of the glass. It is shown that the bonding between the metallic glass and the light alloys is improved by increasing the strain undergone by the glass during the process. Bonding is also improved by increasing co-pressing temperature, keeping in mind that the amorphous structure of the glass must also be preserved during the process.

1. Introduction

Bulk metallic glasses (BMGs) display very interesting mechanical properties at room temperature such as a large elastic domain and high fracture stresses [1] together with an excellent formability above the glass transition temperature [2]. In the recent past, there has been a growing use of BMGs in various industrial sectors. However, in order to extend the use of these materials, studies have been made to overcome their lack of plasticity [3] or to produce larger sample by using different welding techniques [4,5] as well as new shaping process [6]. It could also be of interest to associate BMGs with conventional metallic alloys in order to take advantage of the high strength of the glass and of the large ductility of the conventional crystalline alloy in a similar way as in the case of ceramic fibre reinforced alloys. In this framework, a first study has shown that it was possible to associate a bulk metallic glass to conventional light alloys by co-extrusion carried out at high temperature [7].

The aim of this paper is to investigate the feasibility of the elaboration of multilayered materials involving a BMG and a conventional light alloy (Al or Mg alloy) by co-pressing carried out in the supercooled liquid region (SLR) of the selected BMG and to report preliminary results concerning the mechanical strength of the bond between the two materials.

2. Experimental procedure

The $Zr_{52.5}Cu_{27}Al_{10}Ni_8Ti_{2.5}$ (at.%) metallic glass was selected for this investigation. Ingots were first prepared from elemental metals (purity of 99.99%) by arc melting under argon atmosphere and the melting was repeated several times to get a homogenous alloy. The alloys were then cast in a copper mould to produce rods of 3 mm and 5 mm in diameter. The amorphous state of the rod was confirmed by X-ray diffraction (XRD) with CuKα radiation. Thermal stability of the BMG was investigated by Differential Scanning Calorimetry (DSC) at 10 K/min. Details about both the mechanical behaviour and the thermal stability of this BMG have been reported elsewhere [8,9]. In particular, a glass transition temperature T_g^{onset} = 663 K and a temperature of the first crystallisation peak T_x = 759 K were measured in good agreement with previously reported values.

The two light alloys used in this work were an AZ31 (Mg–3Al–1Zn, wt.%) magnesium alloy in the form of 10 mm thick rolled plate and an Al-5056 (Al–5.0Mg–0.1Cu–0.1Mn, wt.%) aluminium alloy in the form of a 10 mm diameter extruded bar.

The rheological behaviour of the three alloys was determined by compression tests involving strain rate jumps. The compression samples were heated up to the testing temperature at 20 K/min and maintained 360 s in order to stabilize the temperature before starting the test. The multilayered material consisted of a circular glass sheet inserted between two circular sheets of light alloy. Before processing, the surfaces of the sheets were systematically polished with SiC 1200 paper. Co-pressing tests were carried out under argon atmosphere in order to limit oxidation. After co-pressing, the sections perpendicular to the interfaces were observed mainly by optical microscopy and the quality of the bonds was roughly estimated by specifically developed shear tests.

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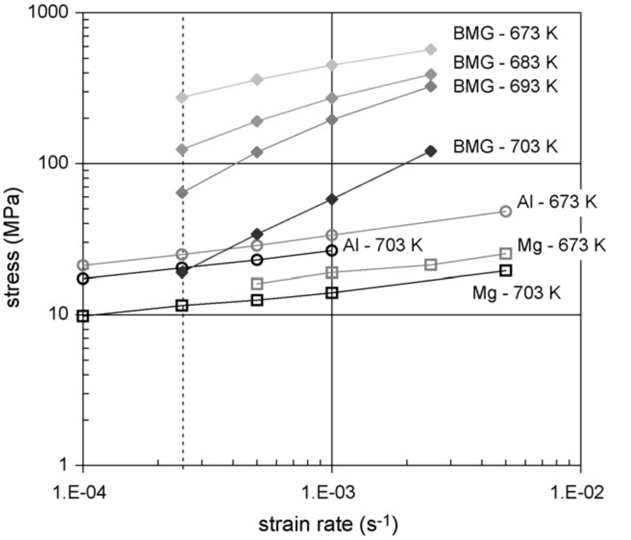

Fig. 1. Effect of temperature on the stress–strain rate curves for the three studied alloys.

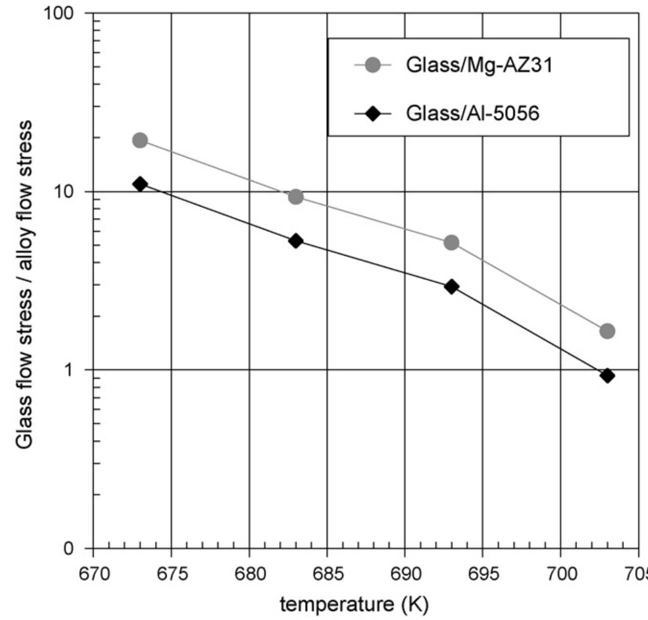

Fig. 2. Variation with temperature of the ratio between the flow stress of the glass and the flow stress of the light alloy for a strain rate equal to $2.5 \times 10^{-4}\,s^{-1}$.

3. Elaboration of the multimaterials

The co-pressing temperatures were selected in the SLR of the glass close to T_g in order to preserve its amorphous state during the process, i.e. between 673 K and 703 K. The compression behaviour of the three materials was determined in this range and Fig. 1 displays the effect of temperature on the resulting stress–strain rate curves. The mechanical behaviour of the alloys can be described by the conventional viscoplastic law $\sigma = K\dot{\varepsilon}^m$ where m is the strain rate sensitivity parameter and K a constant for a given value of m. In the studied experimental interval, the metallic glass exhibits the frequently reported transition between Newtonian ($m \approx 1$) and non-Newtonian behaviour ($m < 1$). As expected, the Newtonian behaviour is promoted by increasing temperature and/or decreasing strain rate. For the light alloys, values of m of about 0.2 are measured in the whole experimental interval. This value suggests that both alloys deform preferentially by dislocation creep as also expected for such experimental conditions [10–12].

For the co-pressing experiments, the total imposed strain is expected to be an important parameter. In this study, it was arbitrarily chosen equal to 0.5 since a preliminary work suggested that a lower strain was not sufficient to get satisfactory bonding. The strain rate was selected to avoid crystallisation of the glass during the process. Hence, an imposed macroscopic strain rate equal to $2.5 \times 10^{-4}\,s^{-1}$ leading to a co-pressing duration of 2000 s was systematically applied which allows keeping the amorphous structure whatever the selected temperature.

Fig. 2 displays the variation with temperature of the ratio between the flow stress of the glass and the flow stress of the light alloy for a strain rate of $2.5 \times 10^{-4}\,s^{-1}$. This ratio is higher than 10 when $T = 673$ K and continuously decreases when temperature increases. The only tested condition where the flow stress of the glass is lower than the flow stress of the light alloy corresponds to the multilayer with the aluminium alloy at the highest temperature ($T = 703$ K).

In this work, all the samples were co-pressed at four temperatures (673 K, 683 K, 693 K and 703 K) up to a macroscopic strain of about 0.5. After co-pressing at the lower temperature (673 K), the samples split off in three pieces just after removal from the furnace indicating that no bonding occurred during the process whatever the nature of the light alloy. At a temperature of 683 K only the Al/BMG/Al is bonded and for the higher temperatures the quality of the interfaces appears quite satisfactory as illustrated by the observations of the multilayer cross sections shown in Fig. 3. This figure shows also that the relative strains of the light alloy and the metallic glass are strongly affected by the temperature of co-pressing. Table 1 gives the strain values experienced by the glass as a function of temperature for both multimaterials. At 683 K, the mean strain in the metallic glass is very limited whereas it increases significantly with increasing co-pressing temperature.

These results are in agreement with the rheological behaviours shown in Figs. 1 and 2: at 683 K, the metallic glass is much harder than the aluminium or magnesium alloy whereas at 703 K, the flow stresses of the three materials are in the same range (if the comparison is performed at the macroscopic applied strain rate). In fact the difference between experimental values of the strain experienced by the glass and predictions, assuming isostress conditions of deformation (i.e. equal stresses in each constituent) and viscoplastic behaviour for the various materials (i.e. $\sigma = K\dot{\varepsilon}^m$), is less than 0.01 for both multilayers.

These results suggest that the bonding between the glass and the light alloy is improved when the glass is significantly deformed during the process.

4. Characterisation of the multilayer materials

In order to evaluate, even roughly, the mechanical strength of the bonding at room temperature, a “shear out” test has been developed. As shown in Fig. 4, the test consists in shearing the bonded specimen along the interfaces which allows calculating a “shear out strength” from the maximum force required for breaking the interfaces. For all the tests, the cross head speed was 0.01 mm/min. The measured values are reported in Table 2, confirming that the bonding strength increases with increasing co-pressing temperature.

Table 1
Experimental values of the strain undergone by the glass for the Al/BMG/Al and Mg/BMG/Mg multilayers at the various temperatures of co-pressing.

Co-pressing temperature (K)	673	683	693	703
ε_{exp} of the glass (Al/BMG/Al)	0.04	0.1	0.34	0.90
ε_{exp} of the glass (Mg/BMG/Mg)	0	0.02	0.16	0.51

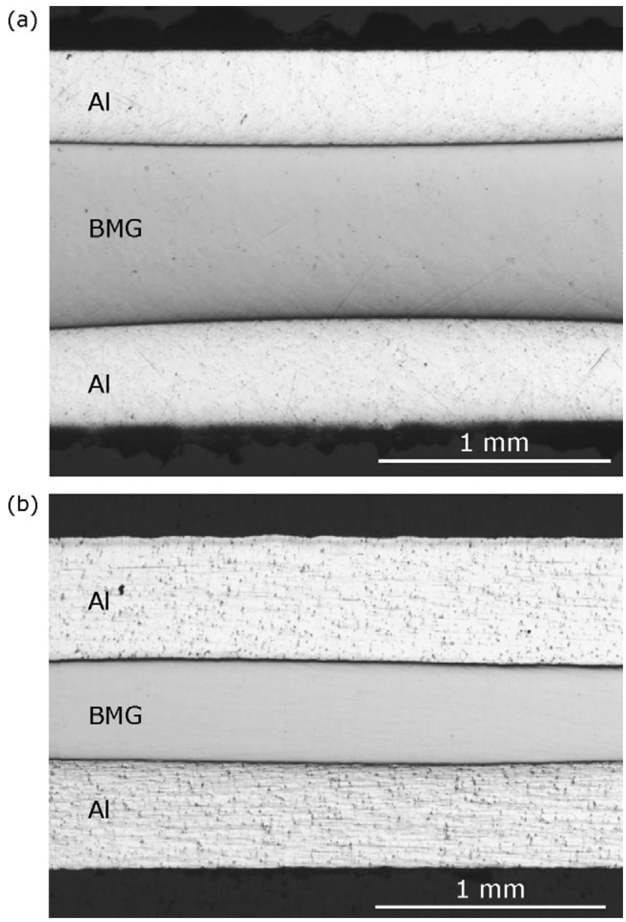

Fig. 3. Optical micrograph of the cross section of co-pressed Al/BMG/Al samples. (a) Co-pressing at low temperature (683 K) leading to no significant deformation of the glass. (b) Co-pressing at high temperature (703 K) leading to significant deformation of the glass.

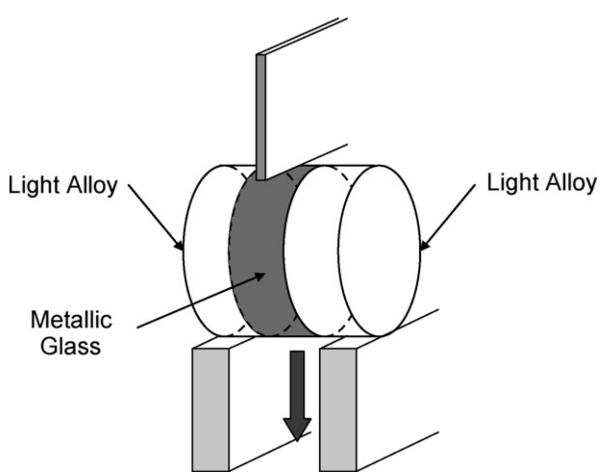

Fig. 4. Scheme of the shear out test.

Table 2
Shear out strength as a function of temperature of co-pressing.

Co-pressing temperature (K)	Shear out strength (MPa)	
	Mg/BMG/Mg	Al/BMG/Al
673	No adhesion	No adhesion
683	No adhesion	2.5
693	2	14
703	17	16

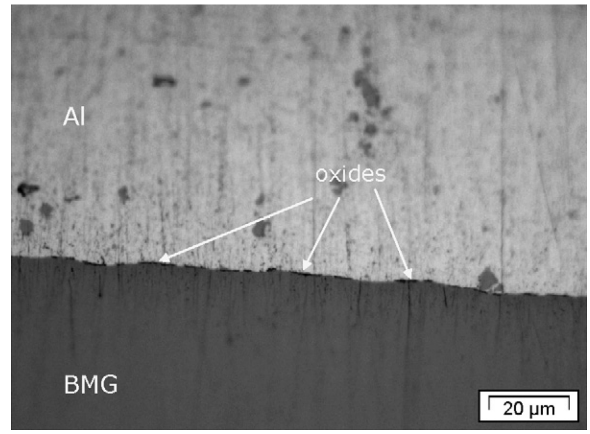

Fig. 5. Optical micrograph of the Al/BMG interface after co-pressing performed at 703 K.

Nevertheless, the interpretation of these values remains quite delicate since it requires a more detailed investigation of the interfaces. In particular, despite the argon atmosphere used during processing, some oxide products can be detected along the interfaces between the glass and the light alloy. These products were preferentially attributed to the oxidation of the glass. The oxidation of various Zr-based BMGs has been quite extensively investigated in the recent past [13–15]. In particular, it was shown that in the case of a ZrAlCuNi metallic glass, the oxidation rate significantly increased between 623 K and 673 K and the main oxide formed was ZrO_2 which is generally considered as a non-protective oxide layer [15]. Roughly similar conclusions were also drawn for a ZrAlNi composition [13]. Fig. 5 displays an optical micrograph of the Al/BMG interface after co-pressing performed at 703 K. A discontinuous oxide layer can be observed with a thickness of about 1–2 μm. Additional tests have shown that this layer is significantly thicker if co-pressing is carried out in air instead of argon, which confirms the beneficial role played by the inert gas. The interesting point is related to the discontinuity of the oxide film. Such a discontinuity supports the idea that oxidation occurred preferentially during heating rather than during the co-pressing itself. In other words, a relatively continuous thin oxide layer probably grew at the glass interface during heating and this layer was progressively broken as the glass deformed. Even if additional characterisation is required before any definite conclusion, it seems the fracture of the oxide layer leads to a direct contact between the light alloy and the glass and thus to the formation of the bonding. This supports the idea of a beneficial effect of the glass deformation on the quality of the bonding.

5. Conclusion

Multilayered materials consisting of a layer of Zr-based BMG and two layers of a light alloy have been successfully elaborated by co-pressing at various temperatures close to the glass transition temperature T_g of the glass. The conditions for co-pressing were the total strain imposed to the multilayered sample (i.e. 0.5) and the strain rate ($2.5 \times 10^{-4}\,s^{-1}$). These first experiments demonstrated that a poor bonding (or even no bonding) was achieved when the glass is not sufficiently deformed and that the adhesion was improved when both the glass and the alloy, whatever it is Al or Mg, deformed. This result suggests that a viscosity ratio close to 1 between the glass and the alloy could be a necessary condition for good bonding between the constituents of the multilayer. Even if co-pressing was carried out under argon atmosphere, an oxide layer was formed on the glass surface during heating. This oxide partially broke up during deformation of the glass which led to the direct contact between the materials and thus to the formation of the bond. Additional work is required to characterise in depth the interfaces between the various constituents of the multilayered material. Moreover, this work was carried out in conditions where the flow stress of the glass is higher than that of the alloys. Experiments will be also performed by selecting an alloy with a larger flow stress that the glass in order to study the influence of this inverse situation on the bonding strength.

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References

[1] J.F. Löffler, Intermetallics 11 (2003) 529.
[2] Y. Kawamura, T. Nakamura, H. Kato, H. Mano, A. Inoue, Mater. Sci. Eng. A A304–306 (2001) 674.
[3] G. Xie, D.V. Louzguine-Luzgin, F. Wakai, H. Kimura, A. Inoue, Mater. Sci. Eng. B 77 (2008) 148.
[4] D.V. Louzguine-Luzgin, G.Q. Xie, T. Tsumura, H. Fukuda, K. Nakata, H.M. Kimura, A. Inoue, Mater. Sci. Eng. B 88 (2008) 148.
[5] T. Shoji, Y. Kawamura, Y. Ohno, Mater. Trans. 44 (2003) 1809.
[6] M. de Oliveira, W.J.F. Botta, A.R. Yavari, Mater. Trans. JIM 41 (2000) 1501.
[7] S. Gravier, S. Puech, J.-J. Blandin, M. Suéry, Adv. Eng. Mater. 8 (2006) 948.
[8] M. Bletry, PhD Thesis, INP Grenoble (F), 2004.
[9] M. Bletry, P. Guyot, Y. Brechet, J.J. Blandin, J.L. Soubeyroux, Intermetallics 12 (2004) 1051.
[10] R. Kaibyshev, F. Musin, E. Avtokratova, Y. Motohashi, Mater. Sci. Eng. A A392 (2005) 373.
[11] J.A. Del Valle, M.T. Perez-Prado, O.A. Ruano, Metall. Mater. Trans. 36A (2005) 1427.
[12] K. Ishikawa, H. Watanabe, T. Mukai, J. Mater. Sci. 40 (2005) 1577.
[13] K. Mondal, U.K. Chatterjee, B.S. Murty, J. Alloys Compd. 433 (2007) 162.
[14] W. Kai, H.H. Hsieh, Y.R. Chen, Y.F. Wang, C. Dong, Intermetallics 15 (2007) 1459.
[15] C.W. Kim, H.G. Jeong, D.B. Lee, Mater. Lett. 62 (2008) 584.