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Metallic Glasses: A Revolution in Material Science

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Abstract

Metallic glasses represent one kind of advanced material, very popular in recent decades. These materials are very adaptable like plastics for their manufacturability in very complex shapes. TPF (Thermoplastic forming) based processes seem very good method to process them. These materials can compete with plastics but have metallic properties. They behave as magnetic materials with less hysteresis loss and less eddy current loss making them suitable for transformer and MEMS (Micro-Electromechanical System) applications. These materials exhibit good corrosion resistance, hardness and toughness. Based on the property and application, metallic glasses are good rivals to plastics, metals and ceramics. Chemical composition and kinetics of supercooling of these materials are the areas where young researchers can focus attention with a view to their improvement.

Keywords: metallic glass, crystalline and amorphous structure, supercooling, TPF-based processing

1. Introduction

In our day-to-day life, we use several types of products made from different materials. Based on the area of application, the desired properties of equipment for their production and processing can vary. According to this, material selection takes place in the way to supply the best possible outcomes compared to others. In recent decades, aluminum, steel and plastics have been the most commonly used materials. The aluminum is considered as the best choice for automobile and structural object for its low density and high specific strength, whereas, due to good strength and cost-effectiveness, steel is the most preferred material for structural applications like construction, railway industry etc. Likewise, plastics are used for various home and kitchen appliances and also for the interior design of buildings, vehicles etc. Plastics are very adaptable materials because of their easy processing, but characterized by low strength compared to metals. On the other hand, aluminum and steel lose the battle with plastics in area of their processing in order to produce very intricate shapes. Because of the high processing temperature, metal products may have more defects over plastics. In this sense, metallic glasses compete with both metals and plastics. These materials have good strength and toughness compared to plastics and can be formed in any desired intricate design compared to metals. Also they possess high corrosion and wear resistance. So, we can say that metallic glasses are the materials having the good properties of metals (like steel and aluminum) as

well as good adaptability like plastics. In metallic glasses, during deformation, dislocation movements occurs, shearing a localization of atoms, but in the case of crystalline materials several defects occur weakening them [1].

As promising materials for different applications, metallic glasses are preferred over metals, ceramics, magnetic and some other types of existing materials due to their enhanced properties. Some of the important reasons for which we consider these glasses for specific applications are discussed in the followings.

1. As discussed earlier metallic glasses have no long-range of ordering like crystalline materials. It develops more homogeneity inside the material because defects like point defects, dislocations and stacking faults are absent.
2. These materials possess very high strength in the elastic region. It can be declared as a good yielding strength of the material which is higher than steel.
3. Because of the good homogeneity of atoms in metallic glasses, very good corrosion resistance is achieved along with good wear resistance.
4. Ordinary silica glasses are brittle in nature unlike the metallic glasses which are very tough materials.
5. These materials have good luster and mirror effects but they are opaque.
6. The metallic glasses are very hard materials and their fracture resistance is much better compared to ceramics.
7. Because of the metallic atoms, these glasses possess significant magnetic effects. It helps to easily magnetize or demagnetize these materials. Metallic glasses with soft magnetism have very small hysteresis loop. Due to the narrow hysteresis, in these glasses hysteresis loss is minimized.
8. Because of the amorphous structure of metallic glasses, their electrical resistivity is higher resulting in less eddy current loss during its application.

The good adaptation of these materials for different industrial applications is a consequence of combinations of those properties. All those applications are discussed later.

The discovery of metallic glass in 1960 motivated scientists to research and manufacture this kind of materials [2]. They were first manufactured in California Institute of Technology, USA. The researchers got the non-crystalline structure in Au-Si alloys. Rapid quenching of those alloys from their liquid state was conducted by the gun technology. They formed a very thin layer of metallic glass over a cold copper substrate. The reason to take copper as the substrate is because of its good thermal conductivity. After that, people are continuously discovering various metallic glasses with different compositions of elements [2]. After 2000 AD, people are making varieties of metallic glasses and its demand is increasing for industrial applications. In the 1990s, the development of different BMGs (Bulk Metallic Glasses) based on late transition metals (LTM) started. A. Inoue et al. successfully developed the Fe-Al-Ga-P-C-B BMGs in 1995 [3]. Today the availability and cost-effectiveness are the two major factors in selecting and production of such materials.

For the stabilization of a supercooled metallic liquid the three rules were proposed [4]:

1. multicomponent BMG systems must consist of at least three or more elements;
2. atomic size of the constituent elements should have a significant size difference (greater than 12%);
3. heat of mixing of the elements should be negative.

2. General description of metallic glasses

According to atomic arrangement, we can categorize the existing and man-made solid materials into two main groups: crystalline and amorphous. When there is a proper ordered arrangement of atoms then we say it is a crystalline material. If there is a random arrangement of atoms, then the material is called amorphous. The atomic arrangements of crystalline and amorphous materials are shown in **Figure 1**. To get such randomness, the sizes of the atoms are very important. Much difference in the atomic radius of the components leads to more randomness in the atomic arrangement. Glass forming is majorly concerned with the study of crystallization of materials in order to avoid crystallization. When metallic alloys are cooled at a very fast rate, possibilities of getting an ordered arrangement are poor [5].

The glass transition temperature (Generally denoted as " T_g ") characterizes amorphous/glass nature of materials. This is more easily understood in the case of a polymer. If we cool a polymer from its liquid state, initially it undergoes cooling and it gets a rubbery state and then after crossing the T_g , it becomes brittle. This kind of phenomenon occurs in amorphous metals too. In case of metallic glasses, we can say that T_g is the temperature at which material gets soft from hard upon heating or get hard upon cooling. This definition for polymers and metals looks similar but it is restricted to amorphous and semicrystalline metals only. The best way to explain the process of getting an amorphous metal or metallic glass is by supercooling the metal from its liquid state. In **Figure 2** T_f is the freezing temperature. During cooling, the liquid goes beyond the freezing point and is known as supercooled metal which can have an amorphous structure [6]. In this way, we can get a metallic glass. In the absence of supercooling, the liquid has a tendency to crystallize [6].

During the formation of glass, the material should avoid the route of crystallization. Crystallization happens during the cooling of material below its liquidus temperature. The difference in Gibbs free energy between liquid and crystalline state is an important factor for the ability of a metal to crystallize or to become amorphous. Whenever there is a transformation between liquid to the solid-state of a material,

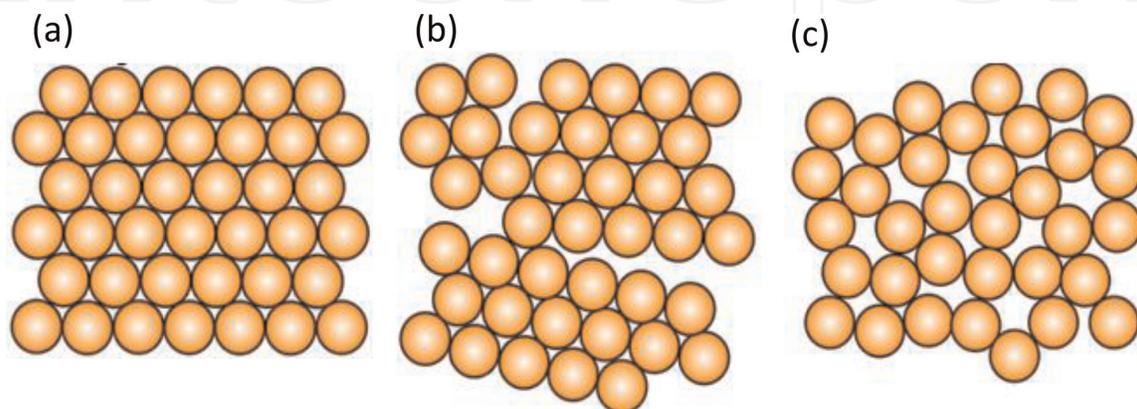


Figure 1.
The atomic arrangement of (a) single crystal, (b) polycrystal and (c) amorphous structures.

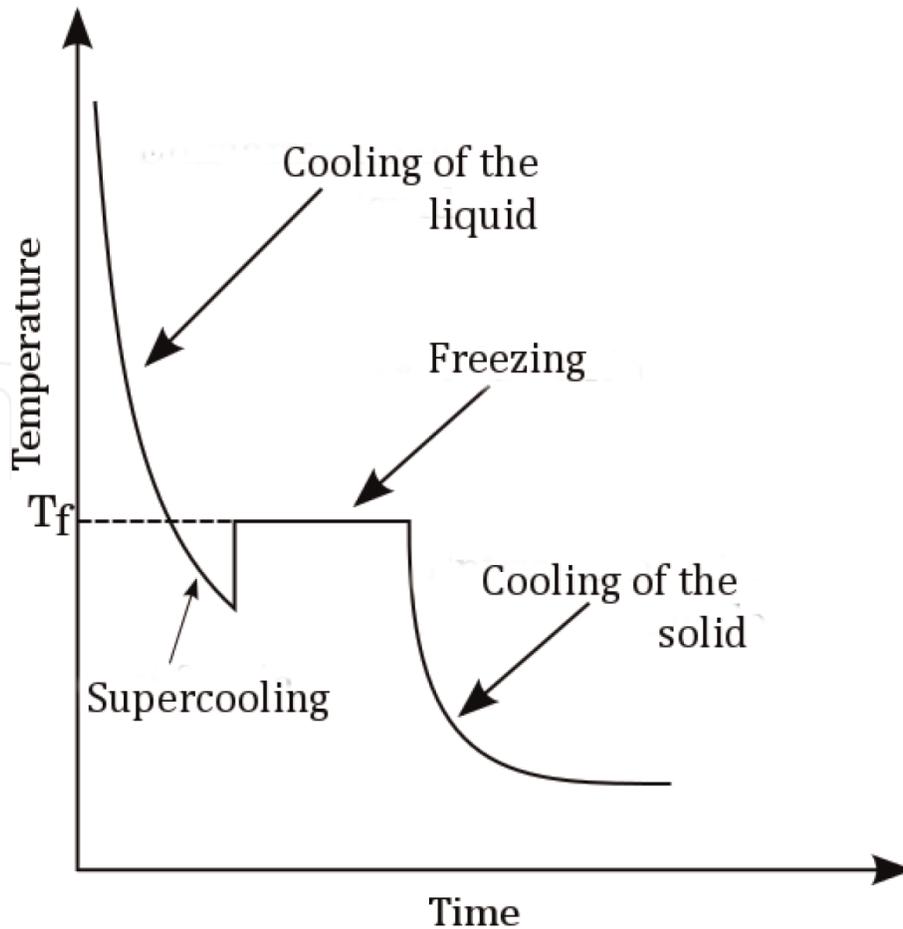


Figure 2.
Principle of supercooling.

the phase transformation at constant enthalpy gives a crystalline material. If enthalpy varies in that process, then the material escapes from the crystalline route and becomes amorphous. As we can see in **Figure 2**, a supercooling does not exist in the case of getting a crystalline material. On the other hand, in the case of supercooling, the enthalpy of transformation changes gradually. Therefore, during the manufacturing of the metallic glasses kinetics of the supercooling has a great impact on the quality of the glasses.

Considering the periodic table, the metallic glasses are mainly divided into two categories: metal–metal and metal–metalloid. In **Figure 3** elements which are metals and metalloids are shown in different colors for the better understanding of their selection for making metallic glasses.

As shown in the periodic table, metals are the elements starting from Lithium with atomic number 3. These are placed from group IA to VIA and shown by the yellow color in **Figure 3**. Some of the examples of metal–metal type metallic glasses are Ni–Nb, Mg–Zn, Hf–V, Cu–Zr, etc. Metals can be alkali, alkaline and rare earth metals etc. In metal–metal type, the atomic percentage of individual constituents can be up to 50%.

In the case of metal–metalloid type, one constituent is a metal and the other one is a metalloid. Metalloids like B, Si, Ge can be mixed with metals like Fe, Ni, Co etc. Metalloids in the periodic table are positioned in a step-like manner and colored with peach color. Metalloids have properties that are intermediate to both metals and non-metals. The composition percentage of metalloids in this category is lower than percentage of metals. After the discovery, the compositions of metalloids in the glasses were generally up to 20% but gradually people work on that problem and

successfully decreased its percentage beyond 20% [7]. As described earlier atomic size, the heat of mixing is also considered for making metallic glasses and used for classification of these materials. The properties based on base metal and categories of metallic glasses are provided in **Tables 1** and **2**, respectively. This type of glasses is more often used in commercial applications.

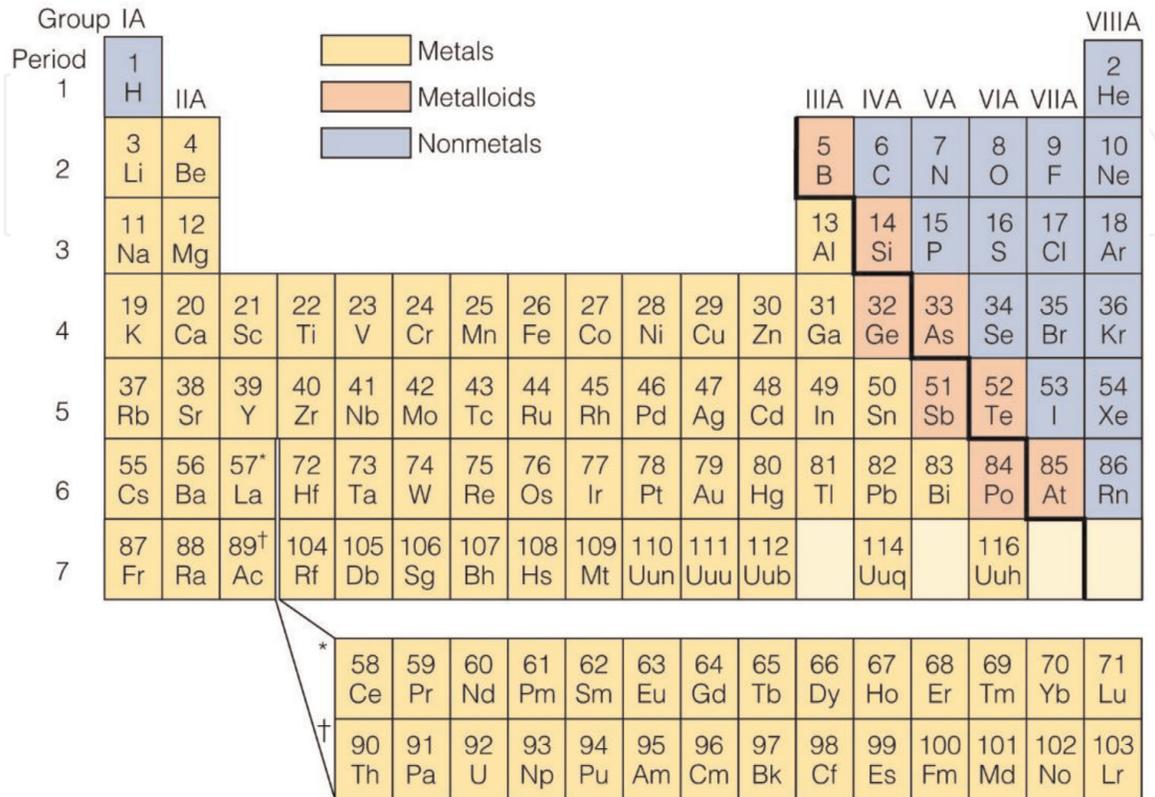


Figure 3.
 Periodic table showing metals, metalloids and non-metals.

Base metal	Properties based on the base metal
Fe-based	Soft magnetism (glass, nanocrystal) Hard magnetism (nanocrystal) High corrosion resistance High endurance against cycled impact deformation
Co-based	Soft magnetism (glass, nanocrystal) Hard magnetism (nanocrystal) High corrosion resistance High endurance against cycled impact deformation
Ni-based	High strength, high ductility High corrosion resistance High hydrogen permeation
Cu-based	High strength, high ductility (glass, nanocrystal) High fracture toughness, high fatigue strength High corrosion resistance
Pd-based	High strength High fatigue strength, high fracture toughness High corrosion resistance

Table 1.
 The basic properties of different LTM-based BMGs [8].

Base metal	Metal–Metalloids	Metal–Metal
Fe-based	Fe–(Al,Ga)–(P,C,B,Si) Fe–Ga–(P,C,B,Si) Fe–Ga–(Nb,Cr,Mo)–(P,C,B) (Los Alamos) Fe–(Cr,Mo)–(B,C) Fe–(Zr,Hf,Nb,Ta)–B Fe–(B,Si)–Nb	Fe–Nd–Al
Co-based	Co–Ga–(Cr,Mo)–(P,C,B) Co–(Zr,Hf,Nb,Ta)–B Co–Ln–B	Co–Nd–Al Co–Sm–Al
Ni-based	Ni–(NbCr,Mo)–(P,B) Ni–(Ta,Cr,Mo)–(P,B) Ni–Zr–Ti–Sn–Si (Yonsei University) Ni–Pd–P	Ni–Nb–Ti Ni–Nb–Zr Ni–Nb–Hf Ni–Nb–Zr–Ti Ni–Nb–Zr–Ti–M (M = Fe, Co, Cu) Ni–Nb–Hf–Ti Ni–Nb–Hf–Ti–M Ni–Nb–Sn (Cal Tech)
Cu-based	Cu–Pd–P Cu–Ni–Pd–P	Cu–Zr–Ti Cu–Hf–Ti Cu–Zr–Ti–Ni Cu–Hf–Ti–Ni Cu–Zr–Ti–Y Cu–Hf–Ti–Y Cu–Zr–Ti–Be Cu–Hf–Ti–Be Cu–Zr–Al Cu–Hf–Al Cu–Zr–Al–M Cu–Hf–Al–M (M = Ni, Co, Pd, Ag) Cu–Zr–Ga Cu–Hf–Ga Cu–Zr–Ga–M Cu–Hf–Ga–M Cu–Zr–Al–Y (Cal Tech)
Pd-based	Pt–Cu–P Pt–Cu–Co–P (Cal Tech) Pt–Pd–Cu–P	

Table 2. *Composition and properties of different types of BMGs (composition of base metal is greater than 50 at. %) [8].*

3. Structure, properties and applications

Structure of material defines its property. BMGs do not exhibit a long-range order structure, as they solidify from liquid without reaching the crystalline ground state. However, short to medium-range structural order does develop to a considerable extent under the given kinetic constraints. This happens because the atoms strive to find comfortable configurations to lower their energy. The structure of the bulk metallic liquids was first observed by Bernal [9] and it was described as dense random packing. Structural features of metallic glasses are discussed by Michael et al. where the concept of efficient filling of space is supported [10]. The rationalization of the good glass forming compositions can be possible by the analysis of dense packing. An example of simple binary metallic glass is shown in **Figure 4** [10].

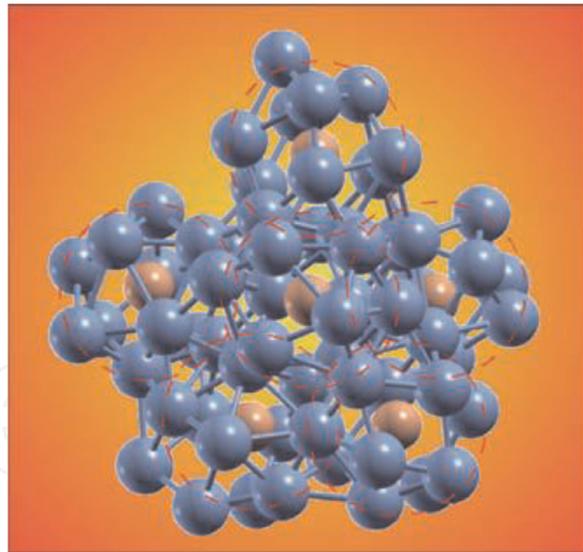


Figure 4.

Model of a simple binary metallic glass: Interpenetrating quasi-equivalent clusters sharing faces, edges, or vertices in the atomic packing configuration of a Zr-Pt metallic glass. The blue balls represent the solvent Zr atoms centered around Pt solute atoms [10].

These structural motifs arise from the strong tendency to form as many bonds as possible between unlike species because of the large negative heat of mixing which is usual in good glass formers. The size of the cluster and its type depend on the relative size of the solvent and the solute. The replacement of Pt solute in **Figure 4** by much smaller Be reduce the number of Zr neighbors which can be accommodated around the solute, and the solute concentration in the alloy would be correspondingly much higher. The medium-range order and dense packing in three-dimensional space can be possible by the overlapping of the cluster via various solvent-atom sharing schemes [11].

The adaptability of the metallic glass in the real world applications is spread in various fields, such as striking face plate in golf clubs, frame in tennis rackets, various shapes of optical mirrors, casing in cellular phones, casing in electromagnetic instruments, connecting part of optical fibers, shot penning balls, electromagnetic shielding plates, soft magnetic choke coils, soft magnetic high frequency power coils, high torque geared motor parts, high corrosion resistance coating plates, vessels for lead-free soldering, colliori type liquid flow meter, spring, in-printing plate, high frequency type antenna material, biomedical instruments such as endoscope parts etc. [12]. Metallic glasses are very strong compared to other conventional materials and that makes it a very good candidate for military applications like armor (Bulletproof vest) piercing bullets, anti-tank projectiles etc. Those metallic glasses, which are stronger than titanium, are also tried for aerospace application. Recently researchers from NASA and different research centers and organizations of China, Britain and Japan are doing several tests to get such ultimate material. This type of materials can give relatively double the performance compared to that of a titanium product in the space application. The major problem lies in the BMGs are the very quick aging of these materials. They become fragile after exposed to external physical stress conditions. The non-uniformity or not enough randomness inside the glass structure leads to the quick aging of these BMGs. So reliability cannot be achieved for the space applications. To remove such weakness of these materials, Wei-Hua Wang from China did experiments like the operation of a blacksmith. Cryogenic treatment of melted metal was done by using liquid nitrogen and maintained to room temperature after solidification. Again the material was melted and the process was repeated for several cycles. The purpose of

choosing the repeated cryo-treatment is to reduce the instability of the material by increasing the randomness inside the material. This type of processing technique enhances the life of a BMG and increases its reliability.

Firstly China and secondly the United States are the major producers of BMGs. Currently, most applications are focused on electric based products like transformer core. Because of the good conductivity properties of BMGs, it dominates in that sector. Other applications like high-temperature applications, aerospace applications and military applications have a long way to go for becoming a better replacement for the recently used materials.

4. Processing of metallic glasses

The flow chart for the processing of metallic glasses in order to obtain different products is presented in **Figure 5**.

4.1 Direct casting

For the net shape fabrication of BMG, two types of casting processes (suction and die casting) have been adopted. Among the two methods the suction casting method develops a product with higher quality and lower porosity than die casting method. BMGs having low melting temperature are beneficial because this process reduces tool cost and wear, lowers energy consumption and shortens cycle time. The TTT diagram and the critical cooling rate (T_c) are highly affected by the maximum temperature prior to cooling rate [13]. The glass forming ability of the BMGs can be diminished if the maximum temperature prior to cooling does not exceed a threshold temperature (TOH). The overheating is rapid and it affects the viscosity in some type of BMGs [14]. This viscosity effect can be broadly explained by the phenomenon including the melting of an oxide phase, ordering phenomenon and a chemical decomposition process [15, 16]. During the casting processes (both die casting and suction casting) shrinkage has to be taken in to consideration. The shrinkage phenomenon is absent in the BMGs formers due to the absence of a first order phase transition during solidification. Cooling process in the casting plays a vital role. Low solidification shrinkage in BMGs develops a gap between the mold and the BMG during the cooling process [19, 20]. The heat transfer through the gap is different for the presence of atmosphere and vacuum, can become the rate

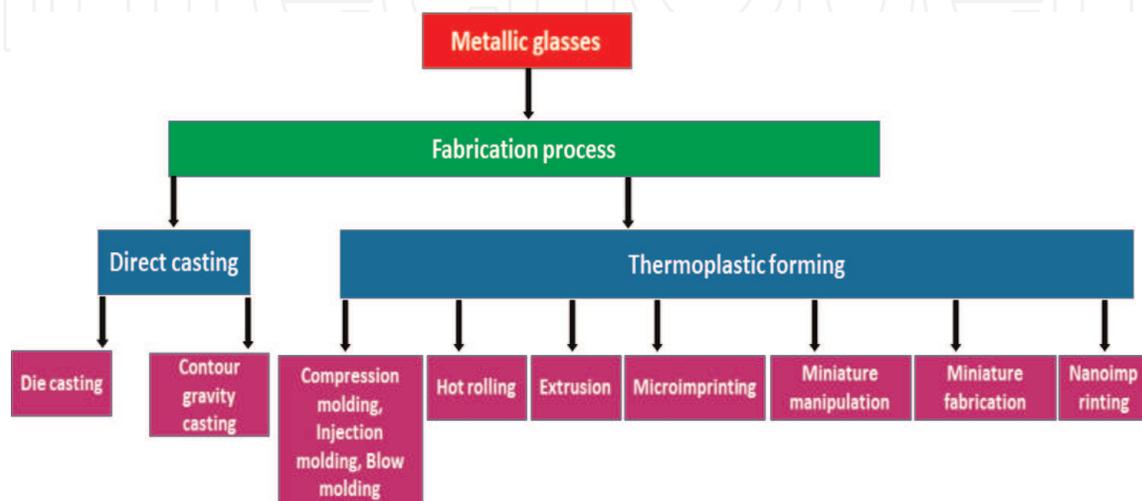


Figure 5.
Flow chart of the processing of metallic glasses.

limiting factor and leads to affect the cooling rate. In the process of direct casting of BMG formers, fast cooling and forming have to be done simultaneously due to the crystallization mechanism and the crystallization kinetics [7, 8]. Direct casting needs care during the mold filling and at the same time to avoid crystallization during solidification. This process is very difficult while making an intricate part which has attractive properties. **Figure 6** represents some application of BMGs fabricated by direct casting. The advantages and disadvantages of the direct casting BMG process are given in **Table 3**.

4.2 Thermoplastic forming

Thermoplastic forming (TPF) is the alternative process to direct casting for developing BMGs. This process has different nomenclature such as hot forming, hot pressing, superplastic forming, viscous flow working and viscous flow forming. The preferable condition for TPF is the drastic softening of the BMG former upon heating above T_g and its thermal stability. The measure of the ability of BMG formers to adopt an amorphous structure by heating above its glass transition temperature is known as thermal stability and it can be quantified by the width of the supercooled liquid region (SCLR). For $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$ a very long processing time is available at low temperature by a high viscosity [17]. Furthermore, the viscosity is significantly reduced at a high temperature which leads to the reduction in processing time. Low viscosity and the long processing time are the



Figure 6.
BMG articles fabricated by direct casting method.

Advantages	Disadvantages
<ul style="list-style-type: none"> • Low melting temperature 	<ul style="list-style-type: none"> • Cooling and forming are coupled
<ul style="list-style-type: none"> • Low shrinkage 	<ul style="list-style-type: none"> • Processing environment can influence crystallization kinetics
<ul style="list-style-type: none"> • One step process 	<ul style="list-style-type: none"> • High viscosity
<ul style="list-style-type: none"> • Homogeneous microstructure 	<ul style="list-style-type: none"> • Internal stresses
<ul style="list-style-type: none"> • Mechanical properties are already matured in the as-cast state 	<ul style="list-style-type: none"> • BMGs contaminate during processing

Table 3.
Advantages and disadvantages of the direct casting process.

optimum parameters for the highest formability of BMG former in its SCLR. According to the data reported in the open literature, the thermos physical properties that are detecting good formability of a BMG former are fragile liquid behavior, large poison ratio and low glass transition. Some TPF based BMG formers the glass transition temperature and the softening in the SCLR permits the use of processing temperature and pressure [18]. The capacity to plastically form metallic glasses in the zone of T_g was recognized by researchers. The enhanced formability of BMGs, which is developed by a wide range of processing methods based on the thermo-plastic forming, attracts a wide range of researchers. In all the above-said processes, one thing is common that is the “feedstock”. The shape and size of the feedstock are different for powder, over rods, disks, plates and films. These TPF based processing methods have been described in the below sections.

4.2.1 TPF based compression and injection molding

Compression molding was an adopted form of plastic processing. In this process, the feedstock material is placed in a mold and is given temperature into SCLR and pressure must exceed the flow stress of the BMG to achieve the required strain prior to crystallization setting. Fast cooling is not required for the forming. **Figure 7** represents the schematic diagram of the process equipped with examples. Under low applied pressure in the compression molding Pd-Ni-Cu-P alloy can be formed as a gear-shaped structure. It can be noticed that a dense compact part with mechanical properties close to those of bulk material and an outstanding surface finish can be obtained [11, 12]. **Figure 7** indicates compression molding of BM6 former, having different feedstock shapes such as pellets, plates and rods. The needed molding pressure not only depends on the formability of the BMG at the processing temperature but also on the shape of the final product. Injection molding is also a TPF based molding process. The only difference between the injection molding and compression molding is that in injection molding, feedstock material is rendered into the mold cavity which has the benefits for the development of the commercial fabrication processes with minimized cycle time.

4.2.2 Miniature fabrication

The development in technologies like micro-electromechanical systems (MEMS), electronics devices, and medical devices have created a rising demand for miniature products and parts. The miniature formation is done by different processes like German method LIGA (lithography, electroplating and molding), UV-LIGA etc. Due to the drawbacks of the LIGA i.e. cost, UV-LIGA is developed giving the similar products. Both processes can be used as the surface patterning

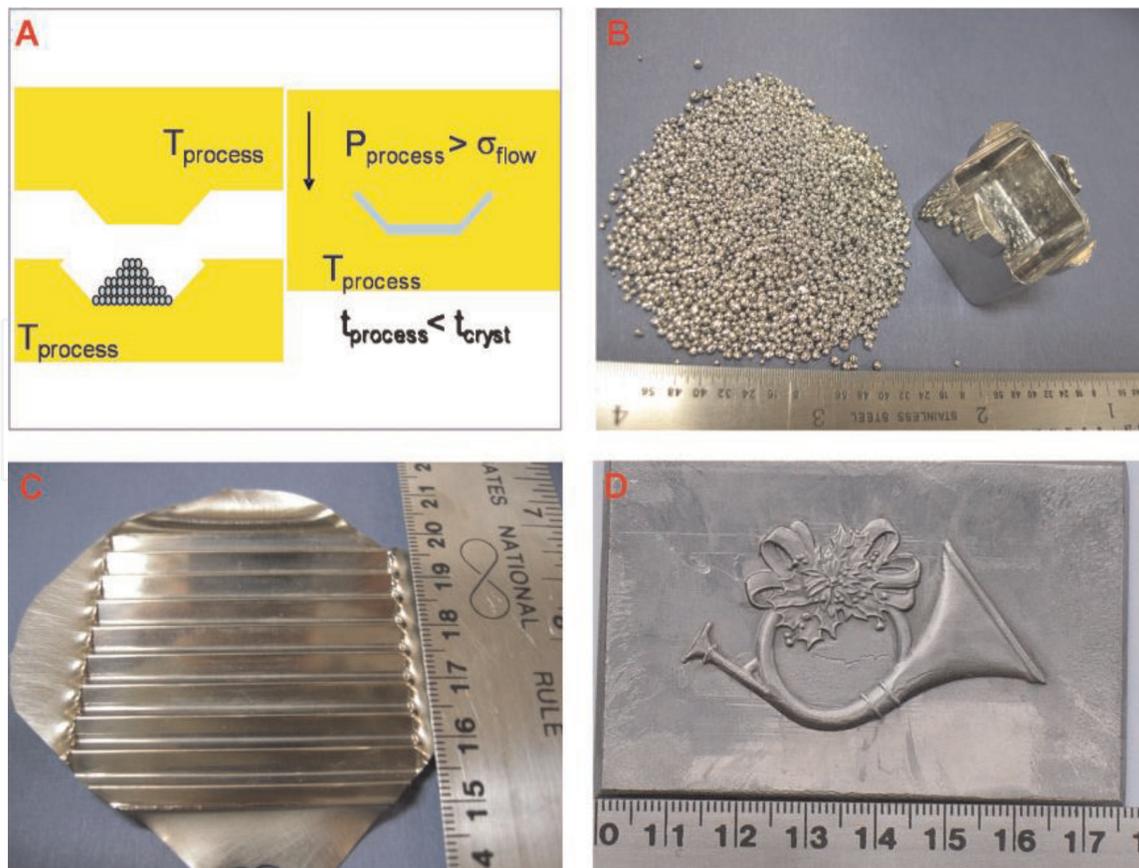


Figure 7. TPF based compression molding with BMG. (A) Schematic diagram of the compression molding with BMGs. (B) Pellets used as feedstock material to compression mold $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$ ($Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$) formed from a flat plate into a corrugated structure. (D) $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$ formed from a flat plate to create an embossing mold.

techniques for creating high aspect ratio structures. Due to the homogeneous and isotropic structure of BMGs in atomic scale and their superior properties over conventional materials used for miniature applications and the capability to produce stress-free parts, these methods attracted a lot of attention.

4.2.3 Nano forming

BMGs basically formed by top-down nanofabrication. The combination of different properties like high strength at room temperature, the ability to imprint a nanometer-sized parallel print process, the non-linear softening of BMGs when reaching their glass transition and the ability to repeatedly write and erase facility on the BMG surface recommends a wide range of application. Nanoimprinting on BMG permits to directly write, such as with atomic force microscopy (AFM) tip, as in a scanning probe lithography process. The capability of BMGs for direct nanoimprinting can be applied with a combination of surface smoothing method and used as a rewritable high-density data storage. Several materials have been developed for the mold formation and imprint for nanofforming such as silicon, quartz, and alumina.

4.2.4 Rolling

Rolling of metallic glasses can be categorized into two processes; one is based on liquid processing and the other is on thermoplastic forming. The example of the former is the melt spinning. In the melt spinning, the liquid sample is quenched by

Advantages	Disadvantages
<ul style="list-style-type: none"> • Forming and fast cooling decoupled 	<ul style="list-style-type: none"> • Two or more step process
<ul style="list-style-type: none"> • Highest dimensional accuracy 	
<ul style="list-style-type: none"> • Insensitive to heterogeneous influence 	<ul style="list-style-type: none"> • Novel and unique process
<ul style="list-style-type: none"> • Novel and unique process “green” process 	
<ul style="list-style-type: none"> • Low capital investment 	

Table 4.
Advantage and disadvantages of TPF based BMG processing.

injecting on a single, fast-spinning copper roll. Another process is cold rolling where the BMGs are rolled at the room temperature.

4.2.5 Extrusion

The thermoplastic formability of BMG formers can also be used for extrusion. The major advantage of extrusion is that it produces the highest aspect ratio shapes, 100+, of uniform cross-section [21]. During extrusion after the outlet of the effective die length, the swelling of the material is a common phenomenon (**Table 4**).

5. Conclusions

BMGs are now available in many different chemical compositions. Although various routes of their processing were adopted, still there is much scope for inventions targeting the development of new processing techniques and new types of glasses. TPF-based processing technique is accepted as a more suitable manufacturing process for obtaining glassy products. This process is competing with the processing of plastics. Very intricate shapes of different geometry, almost unachievable for metals, can be easily achieved with BMGs using TPF-based processing.

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