U.P.B. Sci. Bull., Series B, Vol. 68, No. 1, 2006

GLASS PARTICLES - A COMPLEMENTARY MATERIAL FOR METALLIC COMPOSITES

G. NEAGU, FL. ŞTEFĂNESCU*

În lucrare se prezintă o categorie de compozite metalice, puţin cunoscută, din sistemul aluminiu-particule de sticlă. Sunt analizate problemele teoretice şi practice legate de obţinerea prin turnare a compozitelor cu matrice metalică şi particule. De asemenea, sunt prezentate date experimentale privind rezistenţa la uzare a acestor materiale.

The papes refers to a less known category of metallic composites: aluminium-glass particles. Some theoretical and technological aspects concerning the production of cast composites with metallic matrix and particles are analyzed. Also, experiment data regarding the wear resistance of composites are presented.

Keywords: aluminum matrix, glass particles, casting, wear resistance.

Introduction

Many modern technologies require materials with special properties and low cost that cannot be met at classic materials. In consequence, it becomes necessary to produce new materials.

A composite material can be defined as a mixture of two or more different materials with a distinct separation surface. It results a material with superior properties to those of the individual component (mechanical, tribological, thermal, etc.).

In most cases composites include a base material, the matrix, in which there is dispersed a complementary material such as particles (more than 5% vol.) or fibres.

The matrix of composite materials must be constituted from a material able to include the complementary component without to destroy it by dissolution, melting, chemical reaction or mechanical action. However, it is necessary to specify that it is not always aimed the complete elimination of the chemical phenomenon from the interface. Sometimes, the reaction layer can create a strong contact between the components. For each combination of materials it exists a

---

* Prof., Prof., Dept. of Materials Processing and Ecometallurgy, University POLITEHNICA of Bucharest, Romania

distinct value of the time after which the layer resistance from the interface reaches a maximum value.

The current technologies include organic, ceramic or metallic matrix composites [1]. Metals and alloys present many properties to be utilized as matrix: good values of tensile strength or elastic modulus, wear resistance, acceptable geometrical stability, high thermal or electrical conductivity, good processing and low gases porosity.

The main alloys used as matrix are based on aluminium, titanium, magnesium, copper, nickel, and stainless steel processed in solid, liquid or semi-solid state.

The use of complementary materials as particles presents some important advantages: low cost in comparison with fibres; simple technologies for their introduction and dispersion into the matrix; possibility to obtain isotropic materials and to employ standard technologies in metals forming or casting.

The particles used to obtain metallic composites include ceramic materials (oxides, carbides and nitrides), graphite, steel or cast iron, lead, etc. in spherical, plate (platelets) or other shape (such as particulates). Generally, the particles increase the matrix modulus, assure a reduced density of products, and geometrical stability, improving the wear resistance and the capacity of vibration damping.

The wear resistance of metals and alloys is a relative property that expresses the material behaviour in certain friction conditions. By using a lubricating or a hard complementary material the intensity of the friction processes can be diminished.

The main advantage of metallic composites with particles results from the possibility to assure the two mentioned conditions. The graphite is the most used lubricant and can assure small friction coefficients ($\mu = 0.2$) [2]. A similar effect is achieved when hard materials like silicon carbide, alumina or *glass particles* are included into a soft metallic matrix.

The principal advantages of glass particles in comparison with classical materials used to improve the wear resistance are the low cost and high chemical resistance.

Experimental results and Discussion

Experimental works were made with the aim to investigate metallic composites in aluminium matrix systems. Aluminium and its alloys can be utilized as matrix due to their low cost and density, satisfactory mechanical properties, high thermal conductivity, good fluidity and processing. Also, aluminium is used for its good corrosion resistance. In order to obtain the composite materials, commercial Al, Al-12%Si, Al-5%Si-3%Cu, were melted in a graphite crucible.

Glass micro-balloons, with the diameter between 160 and 200 μm, were utilized as complementary material. Glass particles are recommended by the following characteristics: low cost, high hardness (which causes, unfortunately, rapid wear of cutting tools), high chemical resistance, small density. During the experiments the fraction of dispersed phase varied from 3% to 30% mass. The features of glass particles are presented in Table 1.

Table 1

Characteristics of glass particles

Composition	71.35% SiO_2, 15% Na_2O, 10% CaO, 2% MgO, 1.5% Al_2O_3, 0.15% Fe_2O_3
Density	2520 kg/m^3
Diameter	< 180 μm
Softening point	595°C
Sintering temperature	820°C
Shape	Spherical
Colour	White-yellowish

Though to produce a metallic melt-solid particles mixture seems to be an easy problem, nevertheless there are many difficulties determined by wetting conditions in the system. The wetting degree of solid particles by a melt is estimated on the basis of the contact angle θ (included in Young's equation):

$$\cos\theta = \frac{\sigma_{pg} - \sigma_{pl}}{\sigma_{lg}}, \tag{1}$$

where: σ_{pg}, σ_{pl} and σ_{lg} are the interfacial tension particle-gas, particle-liquid and liquid-gas respectively.

In order to diminish the wetting angle for a better inclusion of the complementary material into the liquid matrix the following measures were taken:

- the overheating of the liquid matrix up to 810°C; the increase of temperature causes the diminution of the surface tension of the liquid phase and wetting angle between the matrix and the disperse material, reducing in this way the immersion energy.

Also, a high temperature for the aluminium melt favours the reduction of the dynamic viscosity. The temperature dependence of the aluminium viscosity can be described by the Arrhenius' equation:

$$\eta = A \exp^{\frac{E}{RT}}, \tag{2}$$

where: A is the pre-exponential viscosity, a constant for each element (for aluminium A = 0.257 mPa.s [3]);

E – the activation energy for viscous flow ($E = 13.08$ kJ mol^{-1} [3]);
T – the overheating temperature;
R – the gas constant (R = 8.3144 Jmol^{-1}K^{-1}).

The calculated data for the dynamic viscosity of pure aluminium are presented in Fig. 1.

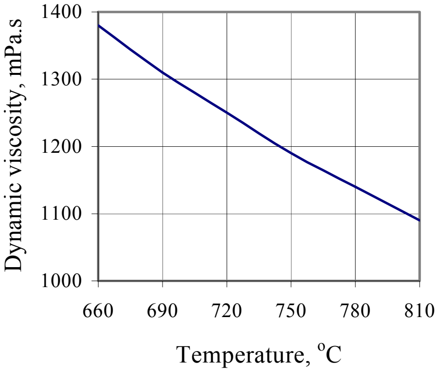

Fig. 1. The dependence between the dynamic viscosity and the temperature, in the case of an aluminium melt.

- the use of heat treated dispersed material; so the dispersed material was preheated at 300°C to remove the gaseous phase absorbed by the superficial layer. Preheating was realized at a temperature above a critical value, considered to be the softening point.
- the alloying of melt; magnesium introduced in the melt creates advantageous wetting conditions by diminution of surface tension of the liquid matrix. The effect of the magnesium addition can be analyzed on the basis on Lang's empiric relationship (valid for 0…1.5% alloying element [4]):

$$\sigma_{lg} = 0.851 - 0.0419[\%\mathrm{Mg}]. \tag{3}$$

The calculated values are presented in Fig. 2.

The viscosity of a binary Al-Mg system can be described by Moelwyn-Hughes model [3]:

$$\eta = \left(X_{\mathrm{Al}}\eta_{\mathrm{Al}} + X_{\mathrm{Mg}}\eta_{\mathrm{Mg}}\right)\left(1 - \frac{2X_{\mathrm{Al}}X_{\mathrm{Mg}}\Omega}{RT}\right) \tag{4}$$

where: η_{Al}, η_{Mg} are the viscosities of the two elements, X_{Al}, X_{Mg} – the mole fractions; Ω - the regular Al-Mg interaction parameter.

The composite materials analyzed were obtained by the introduction of secondary phase into a metallic bath using the Vortex method (based upon the mechanical stirring). The method was recommended due to its simplicity and the reduced costs. The stirring device contains an electric motor and a turning axle with two steel palettes.

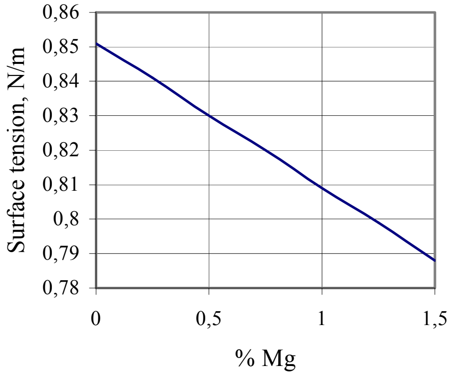

Fig. 2. Variation of the surface tension of aluminium melt with the alloying element content.

The diameter of the stirring device was 0.85% from the ladle diameter. The inclination angle is recommended to vary between 20 and 30°. The penetration depth of the turning axle was 0.8 *h*, where *h* is the height of the casting ladle. To avoid the agglomeration of the dispersed phase, the glass micro-balloons were directly let in the middle of the moving cone (eddy zone). Due to the impact with the steel blades, the particles were dispersed in the melt. The incorporation and dispersion degree of glass micro-balloons in the liquid matrix depends on the agitation time.

By a progressive introduction of the solid phase, the agitation intensity was reduced. Consequently, the rotation of the palettes was adjusted according to the proportion of complementary material ($n = 3…9$ rot/s).

It is well-known that the mechanical agitation method leads to satisfactory results regarding the incorporation of the complementary material but there are some disadvantages such as: the increase of the contact of the liquid alloy surface with the air; the possibility to appear a particle agglomeration during the incorporation process; the segregation of the dispersed phase.

After stirring, the composite material was cast in metallic and sand based moulds (T = 700°C). Gravitational casting is the simplest variant to obtain castings from composite materials.

Also, the presence of complementary material leads to the increase of the composite viscosity that allows one to consider the mixture just as multiphase fluids. Therefore, the liquid alloy Al-Mg can be investigated like a continuous medium in connection with the apparent viscosity. For the volumetric contents of complementary material larger than 5% the apparent dynamic viscosity is given by the relationship:

$$\eta_a = \eta_0\left(1 + aC_v + bC_v^2\right), \tag{5}$$

where: η_0 is the dynamic viscosity of the melt at the reference temperature (η_0 = 1.29 mPa.s); a – the hydrodynamic constant (for spherical particles a = 2.5); b = 2.5…14.1 (for aluminium composites b=10.05); C_v is the volumetric concentration.

If the volumetric concentrations is less than 5%, then b=0 (the Einstein's equation).

In Table 2 are presented the calculated values for the apparent dynamic viscosity, at 700°C, neglecting the effect of magnesium in the binary system.

Table 2

The values of apparent dynamic viscosity

C_v, %	5	10	15	20	25	30
η_a, mPa.s	1.484	1.742	2.065	2.454	2.907	3.424

Consequently, the addition of glass particles involves the drastic decrease of the fluidity for aluminium melts.

The glass micro-balloons incorporated in the liquid matrix followed a slight laying down trajectory due to the small difference between the densities of components. The density of the aluminium liquid at different overheating degrees can be estimated by the following equation:

$$\rho_{\mathrm{Al}} = 2.369 - 3.11\mathrm{x}10^{-4}\left(T - T_{melt}\right), \tag{6}$$

where $T - T_{melt}$ is the overheating degree. In consequence, at 810°C, the density of liquid aluminium is 2.322 g/cm^3 and at 700°C it is 2.356 g/cm^3. The value of aluminum density at the melting point calculated by the above equation (ρ_{Al} = 2.369 g/cm^3) is in agreement with data presented in the technical literature (ρ_{Al} = 2,380 g/cm^3 [3]).

In a transitory regime of acceleration ($\frac{\mathrm{d}v_p}{\mathrm{d}t} > 0$), the movement equation for the solid particle of spherical shape (ρ_p>ρ_l) into a stationary melt is:

$$m\frac{\mathrm{d}v_p}{\mathrm{d}t} = G - F_a - F_R, \tag{7}$$

where: $\frac{\mathrm{d}v_p}{\mathrm{d}t}$ is the particle acceleration; G - the particle weight; F_a - the Archimede's force; F_R - the advancement resistance force.

After a period, in the same time with the increase of particle velocity the advancement resistance force enhances too and it leads to $\frac{\mathrm{d}v_p}{\mathrm{d}t} = 0$. Consequently, the movement equation of the particle becomes:

$$F_R = G - F_a \tag{8}$$

or

$$\frac{1}{2} C_R \rho_l \pi r^2 v_p^2 = \frac{4\pi r^3}{3} \rho_p g - \frac{4\pi r^3}{3} \rho_l g , \tag{9}$$

where: C_R is the advancement resistance coefficient; g - the gravity acceleration.

From the equilibrium condition of forces that act on the particle, it appears the expression of the particle velocity:

$$v_p = \sqrt{\frac{8}{3} \frac{r_p}{C_R} \frac{\rho_p - \rho_l}{\rho_l} g} , \tag{10}$$

where the advancement resistance coefficient is determined on the basis of Oseen's relationship:

$$C_R = \left(1 + \frac{3}{16} \mathrm{Re}_p\right) \frac{24}{\mathrm{Re}_p} . \tag{11}$$

Taking into account a laminar movement for solid micro-balloons ($\mathrm{Re}_p<1$), the calculated values of the particle velocity for different values of Reynolds number [5] are presented in Fig. 3.

The analysis of the solidification process was achieved in the case of ϕ10x200 mm, ϕ20x200 mm, and ϕ 30x300 mm samples cast in metallic, dry sand, and green sand moulds. To calculate approximately the solidification duration t_s, the following relationship can be used:

$$t_s = \frac{R_{eq}^2}{K^2} , \tag{12}$$

where: R_{eq} is the equivalent radius or solidification modulus: K – the solidification coefficient.

The equivalent radius is defined by the ratio between the volume and the area of the sample.

The following values were adopted for the solidification coefficient: $k = 3.10^{-3}$m.s$^{-1/2}$ for metallic mould (cast iron); $k = 1.3.10^{-3}$ m.s$^{-1/2}$ for green sand mould; $k = 1.1.10^{-3}$ m.s$^{-1/2}$ for dry sand mould.

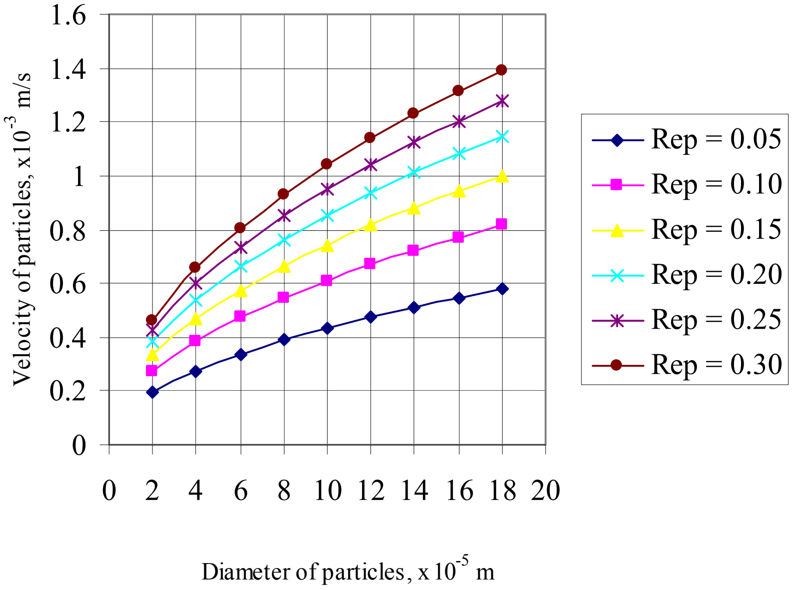

Fig. 3. The velocity of the particle for different values of Reynolds' number.

The calculated values are presented in Table 3.

Table 3

The solidification duration for different moulds

Diameter of sample, m	Equivalent radius, m	Volume of sample, m^3	Time of solidification, s		
			K=3.10^{-3}m.s$^{-1/2}$	*K*=1.3.10^{-3} m.s$^{-1/2}$	*K*=1.1.10^{-3} m.s$^{-1/2}$
1.10^{-3}	2.44.10^{-3}	15.7.10^{-4}	0.662	3.523	4.92
2.10^{-3}	4.76.10^{-3}	62.8.10^{-4}	2.518	13.40	18.725
3.10^{-3}	6.98.10^{-3}	141.3.$^{10-3}$	5.413	28.8	40.265

When casting is made in metallic moulds, the segregation phenomenon is diminished due to the high rate of solidification. Consequently, it is possible to obtain a better distribution of glass particles in the matrix.

Subsequently, cast specimens were processed for metallographic analysis, Brinell hardness and wear resistance testing.

No reaction zone was observed at the matrix–glass particles interface. At a searching analysis of the included particles, the presence of an exterior layer was observed, with a different aspect in comparison with the remainder of material (Fig. 4). This is probably due to a superficial melt of the particles in contact with the liquid alloy.

One may notice also a good adhesion between the matrix and the glass particles, in fact, a good inclusion of the particles in the metallic material. The distribution in a cross section is good enough, although for 20% particles an agglomeration process was noticed. Also, a sintering process was observed, due to the high temperature of the melt (Fig.5).

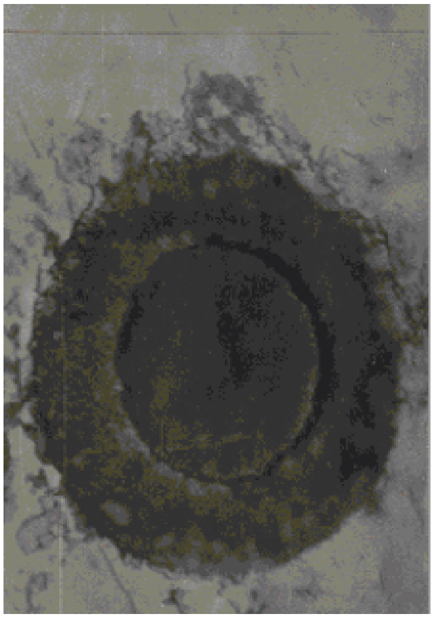

Fig. 4. Glass particle with a visible external layer (x400).

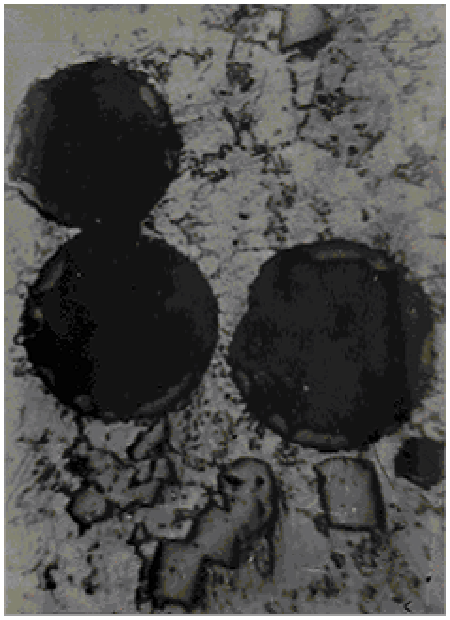

Fig. 5. The sintering process of glass particles (x200).

The solidification structure of glass particles composites is strongly influenced by the presence of the complementary phase. The process can be easily analyzed because the glass micro-balloons are inert in contact with the metallic matrix.

Generally, when the solidification front meets an insoluble particle, two situations can appear:

- the solidified material retains the particles, which remain inside the crystalline grains;
- the solidified material does not include the particles from the beginning, they being pushed into the liquid phase.

Analyzing the solidification structure in the case of Al-12%Si/glass particles, one can observe that the micro-balloons are pushed by the primary dendrites to the eutectic liquid, which is the last to solidify. The reject tendency of particles can be explained by their large size in comparison with the distance between the dendritic arms (smaller than 20 μm). A similar situation is met in the case of the aluminum matrix.

The observed data are in agreement with the theoretical model proposed by Zubkov et al., based on the thermal conductivities ratio ($K = \lambda_p / \lambda_l$). If $K > 1$, the capture takes place. If $K < 1$, a particle pushing occurs. In the aluminum glass system $K = 0.0285$ [5], and the complementary material is pushed as individual or as agglomerated particles.

Generally, the structure of unmodified Al-Si alloys is characterized by the presence of a coarse eutectic. The silicon crystallites are fragile, hard and have a powerful notch effect, which is responsible for the low values of the mechanical properties. Metallographic examination of cast specimens (ϕ20x200 mm) indicates a finer structure of the eutectic alloy at the particle matrix interface. This structure is different from that on the outside of the contact zone. Also, the dispersed phase promotes the heterogenous nucleation of silicon primary crystals. These prove that the glass particles encourage the heterogenous nucleation.

The aspect of the fracture surface of the specimens was analyzed by means of an electron microscope. It was observed that the samples do not crack through the glass particles, which remain on one or another side of the breaking.

In order to establish the influence of the glass particles content on the Brinell hardness of the composite materials a ball with the diameter of 10 mm was used. The force applied was of 500 N. The experimental data indicate an increase up to 90-95 HB for the cast specimens with 15% complementary material.

Cast samples were processed for the abrasion testing. A non-conventional wear resistance testing machine was used in which the specimen pressed against the rolling disc of manganese steel, turns itself and simultaneously describes a semicircular movement.

Experimental data, expressed by mass losses, show that there is a large variation of the wear resistance (Fig. 6) with the glass particles content.

Because the thermal conductivity of glass micro-balloons is less than the same property of the matrix, the heat generated in the friction process will promote an overheating of the material around the particles from the active surface. So, under the action of the pressure the particles will be continuously pushed in the matrix and the active surface will remain smooth all the time.

It can be observed that for an addition higher than 15% glass particles, the mass losses are about constant.

Because metallic composites with particles are heterogeneous media, they can have geometrical and physical defects. For aluminium – glass particles, cast samples, the main following defects can appear:

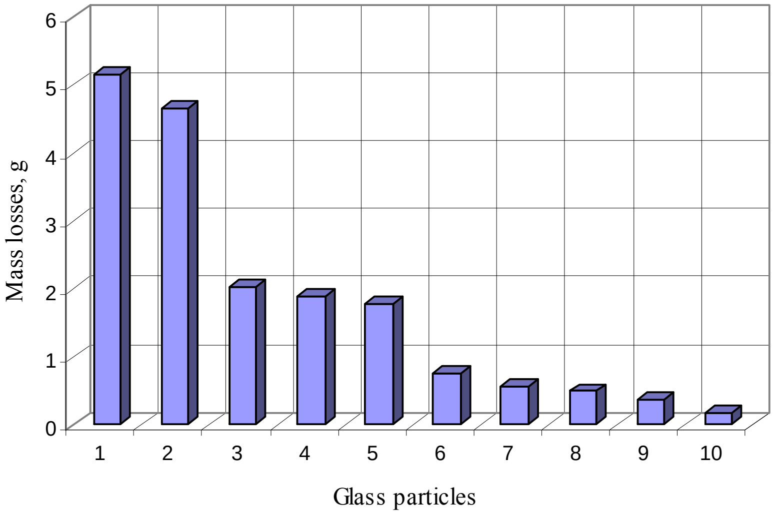

Fig. 6. Mass losses of Al-12% Si/glass particles cast specimens after wear testing for different content of glass particles: 1 – 0%; 2 – 2%; 3 – 4%; 4 – 6%; 5 – 8%; 6 – 10%; 7 – 15%; 8 – 20%; 9 – 25%; 10 – 30% glass particles.

- draws, because aluminium alloys have a coefficient of contraction at solidification of 4…5% and the presence of particles makes difficult the compensation shrinkage cavities with the liquid from the adjacent zones;
- occluded gases, because the surrounding air is partially included in the matrix during the mechanical mixing of the components;
- damage of the complementary material by superficial melting.

Conclusions

Glass particles can be considered an available dispersed material in producing composites with soft matrix and hard surface used as wear resistant material in many possible applications in the field of mechanical installations. In addition, the glass micro–balloons are not expensive.

To obtain these materials a well knowledge of the behaviour of the complementary material in each stage of the production is required, the stages being: introduction into the melt, movement into the liquid alloy, casting (viscosity of the mixture), chemical interaction between components, solidification of the matrix, etc.

R E F E R E N C E S

1. *D. Miracle and S. Donaldson*, “Introduction to Composites”, in ASM Handbook, **vol. 21**, Composites, ASM International, Material Park OH, 2001, pp. 1029-1032.
2. *P.K. Rohatgi, S. Ray and Y. Liu*, “Tribological properties of metal matrix-graphite particle composites” in International Materials Reviews, **vol. 37**, no. 3, 1992, pp. 129-149.
3. *A.T. Dinsdale and P.N. Quested*, The viscosity of aluminium and its alloys-a review of data and models, 1st Fe-Al Meeting, Boulder, 22 June 2003.
4. *F. Frisvold*, Filtration of Aluminium, Theory, Mechanism and Experiments. PhD Thesis, Metalurgisk Institut, Trondheim, 1990.
5. *B.N. Keshavaram, P.K. Rohatgi, R. Asthana and K.G. Sathyanarayana*, “Solidification of Al-glass particulate composite”, in Solidification of Metal Matrix Composites, The Minerals, Metals &Materials Society, 1990, pp. 151-169.