SOME THEORETICAL AND PRACTICAL ASPECTS IN PRODUCING CAST ALUMINIUM - SILICON CARBIDE PARTICLES COMPOSITES

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The paper presents some theoretical and practical aspects regarding the cast aluminium - silicon carbide particles composites. They are analyzed the main characteristics of the matrix and complementary material, the wetting conditions and the improvement methods, segregation processes of particles in the metallic bath under a laminar or turbulent movement regime, the flow capacity of mixtures and the influence of silicon carbide on the solidification process parameters..

Keywords: aluminium, silicon carbide particles, wetting conditions, segregation, fluidity, solidification process

1. Introduction

Generally, a composite can be defined as a combination of two or more components, whereby the properties complement each other, in order to obtain a complex material with some properties superior to those of components.

The combination of relatively low cost, reduced density (2.705 g/cm^3 at 20°C for 99.5% pure aluminium), enough raised fluidity, good resistance to the environment action (as a result of an alumina layer, compact and impermeable to gases), acceptable mechanical properties (35 MPa yield strength, 70 GPa modulus of elasticity), good thermal conductivity (λ = 237-250 W/(m·K)), easy workability, and recycling possibilities, recommends *aluminium and its alloys* as convenient materials for matrix [1] .

Particles of *silicon carbide* are widely used as complementary material, although their density is slightly higher than that of aluminium, because they are cheap and easy to be produced.

Silicon carbide is part of technical ceramics group. The structure of this material is composed of SiC_4 or CSi_4 tetrahedral flats. A cubic (αSiC) or hexagonal elementary cell type (βSiC) is obtained, depending on the arrangement of tetrahedral flats. In producing metallic composites it's frequently used αSiC.

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Silicon carbide presents the following characteristics: low density (ρ = 3.1...3.21 g/cm^3); large tensile strength, stable until high temperatures; high hardness (9.2...9.3 on the Mohs' scale); high values for the modulus of elasticity (E = 400 GPa); low coefficient of thermal expansion (α = 4·10^{-6} K^{-1}); high thermal conductivity (λ = 360...490 W/m·K); high resistance to thermal shock, [1,2,3].

Composites based on aluminium and silicon carbide particles have low weight, good thermal conductivity, high resistance to abrasive wear by friction and may be designed for a low coefficient of thermal expansion (α = 15·10^{-6} K^{-1} for 20% SiC$_p$) [4].

Wetting conditions are usually difficult to be achieved in the system aluminium - silicon carbide. Experimental data obtained by sessile drop technique indicated non-wetting conditions [5, 6, 7].

The high values calculated for the critical acceleration of particles required to penetrate the melt, impose to improve the wetting conditions [8].

2. Casting and solidification

The metallic matrix composites were produced by casting, using the method based on the mechanical mixing of components.

Gravitational casting is the simplest method of composites processing. Casting is made at low speed, avoiding the turbulences which lead to the adsorption of environmental gases. Gases removal is difficult due to the high viscosity of the multiphase melt.

Commercial aluminium has been used to obtain metallic composites (Table 1).

Table 1

Chemical composition of commercial aluminium

Matrix	Al	Fe	Cu	Mn	Mg	Si	Zn	Other elements
%	97.950	0.574	0.163	0.107	0.212	0.731	0.187	0.076

The aluminium was melt in a flame furnace.

Liquid matrix was strongly stirred with a shaft with two metallic blades, in connection with an electrical engine with variable rotation. The particles were introduced in the vortex zone. Rotation speed of the impeller was correlated with the proportion of particles introduced in the melt. The diameter of the stirring device was 0.85% from the casting ladle diameter. The angle between the longitudinal axis of the shaft and the vertical direction varied between 20…30°. Incorporation and dispersion degree of the silicon particles in the metallic bath depends on the agitation time. Initially, the complementary material penetrates the melt as clusters of particles and subsequently dispersion occurs [9].

Mechanical stirring was continued for 60 seconds after the introduction of complementary material.

Some methods to improve wetting condition were applied:

- *alloying of the melt*. In order, to reduce melt surface tension, 1.5% magnesium was introduced into the metallic bath. Magnesium also can react with the oxygen presents at the surface of silicon carbide particles favouring direct contact of the components.

- *overheating of the liquid matrix*. The temperature of the melt was 868°C.

- *heat treatment* of the particles at 500…550°C to remove gas film on the surface.

After embedding, the forces acting on solid particles can be classified in mass and surface forces. Steady state of these forces leads to particle movement or repose in the metallic melt.

The expression of the settling velocity results from the equilibrium condition:

$$v_p = \sqrt{\frac{8}{3}\frac{r_p}{C_R}\frac{\rho_p - \rho_l}{\rho_l}g} \ , \tag{1}$$

where: r_p is the particle radius; C_R - the drag coefficient; ρ_p - the particle density; ρ_l - the density of the liquid aluminium; g – gravitational acceleration.

The drag coefficient is dependent on Reynolds number attached to the particle and its shape.

For a spherical particle the drag coefficient can be determined by using Oseen's relationship:

$$C_R = \left(1 + \frac{3}{16}\mathrm{Re}_p\right)\frac{24}{\mathrm{Re}_p} . \tag{2}$$

In Table 2 the settling velocity values of a spherical silicon carbide particle in aluminium melt are presented. The calculations were carried out for a laminar and turbulent movement respectively of particle, in a temperature range between 700…900°C, for different radius of complementary material. Luca's relationship was used to determine the density of the metallic bath [10]:

$$\rho_{Al} = 2.369 - 3.11 \cdot 10^{-4}(T - T_{melt.}) . \tag{3}$$

Table 2

The values of settling velocity

Re_p	C_R	$T,$ °C	$v_p \cdot 10^{-3}$, m/s				
			$r_p{=}2\cdot10^{-5}$m	$r_p{=}4\cdot10^{-5}$m	$r_p{=}6\cdot10^{-5}$m	$r_p{=}8\cdot10^{-5}$m	$r_p{=}\cdot10^{-4}$m
0.75	36.5	700	2.264	3.202	3.922	4.528	5.063
		800	2.323	3.285	4.023	4.645	5.194
		900	2.379	3.365	4.121	4.759	5.320

		700	2.810	3.974	4.867	5.620	6.283
1.25	23.7	800	2.882	4.076	4.992	5.765	6.445
		900	2.953	4.176	5.114	5.906	6.603

For the same flow regime, regardless of the particle size, increasing the temperature the rate of sedimentation makes greater, probably as a result of the melt viscosity reduction.

It may be admitted that at constant pressure, dynamic viscosity decreases with temperature after an exponential law [11]:

$$\eta = \eta_0 e^{\frac{E_v}{RT}},$$ (4)

where: η_0 is the pre-exponential viscosity (η_0=0.257 mPa·s); E_v - the activation energy for viscous flow (E_v = 13.08 kJ mol^{-1}); R – the gas constant.

At the same temperature, the turbulent flow favours a high settling velocity. Consequently, theoretically, the possibility of an accentuated segregation phenomenon of dispersed material exists during casting.

However the theoretical results are in contradiction with a series of experimental researches. This situation can be explained on the basis that the mixture between the two components is a multiphase fluid, characterized by an apparent viscosity.

If the volumetric concentration of silicon carbide particles is higher than 5%, the next relationship is recommended for the apparent dynamic viscosity:

$$\eta_a = \eta_0 (1 + aC_v + bC_v^2),$$ (5)

where: η_0 is the dynamic viscosity of the melt at the reference temperature; a – the hydrodynamic constant (a = 2.5 for a solid and spherical particle); b = 10.05; C_v - the volumetric concentration.

The value of hydrodynamic constant diminishes if particle deviates from a spherical shape, tending to one ellipsoidal.

For multiphase melts a new equation is used to determine the settling velocity of the particles greater than 100 μm [12]:

$$v_p = v_{St} (1 - C_v)^{4.65},$$ (6)

where: v_{St} is Stokes modified velocity ($v_{St} = \frac{2}{9} gr_p^2 \frac{\rho_p - \rho_l}{\eta_a}$).

However, it should be noted that the proposed equation does not consider the Reynolds number attached to the particles.

Table 3 presents the values of settling velocities of SiC particles at 700°C, using equations 4, 5, 6 and Stokes modified velocity.

<div align="right">*Table 3*</div>

The settling velocity for multiphase melts

C_v, %	$v_p \cdot 10^{-3}$, m/s		
	$r_p = 6 \cdot 10^{-5}$m	$r_p = 8 \cdot 10^{-5}$m	$r_p = \cdot 10^{-4}$m
5	3.548	6.307	9.855
7	3.019	5.367	8.386
10	2.350	4.177	6.527

It can be seen that increasing the concentration of complementary material, settling velocity decreases. Elevated melt viscosity slows the settling of silicon carbide particles.

In practical conditions, the settling velocities are higher due to agglomeration of particles. The necessary condition for the formation of a cluster:

$$\Delta E_\sigma < 0 \qquad (7)$$

is accomplished, the negative value of the surface free energy being provided by the non-wetting conditions from the system Al-SiC ($\theta > 90°$).

To determine the flow capacity of the mixtures with 1.5% Mg, 3 or 5% SiCp, spiral test sample was used. Experimental results are presented in Figure 1.

The sample with 1.5% Mg has a lower flow capacity due to occurrence of Al_2Mg_3 compounds. This intermetallic compound has a negative effect on the viscosity of the melt. Also, silicon carbide particles grow the internal friction forces in the same time with the increase of solid component proportion.
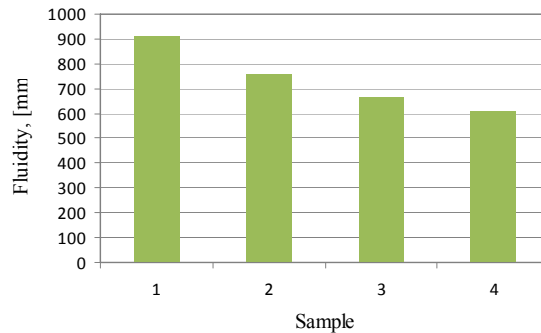

Fig.1. Fluidity variation: 1 – Control sample; 2 – Al–1.5% Mg; 3 – Al-1.5% Mg/3% SiCp; 4 - Al-1.5% Mg/5% SiCp.

The influence of SiC particles on the solidification parameters was examined by performing some experiments which revealed their ability to stimulate germination and growth of the solid phase.

The solidification process is influenced by the heat accumulation capacity of the mould and by its configuration.

To determine the rate of solidification it was examined the advancement in time of the solidification front represented by the surface of a solid layer formed on a flat surface and at the junction of two walls. The inner surface of the mould was that of a square prism clad with laminate steel coolers (12 mm thickness).

Aluminium - 1.5% Mg and mixtures of this alloy with 5% and 10% SiCp were cast. The melt cast in the cavity of the mould was maintained for 5, 10 and 15 s. After this period the non-solidified mixtures were removed by tipping. The layer thickness δ was measured on the middle of each side and on the corner (where the cooling was more intense due to conductive heat transfer intensified by the presence of the two walls joined at 90°). Finally the arithmetic mean of the four values was calculated.

Variation of the solidified thickness on the two analysed directions is presented in Table 4.

The research carried out by measuring the layer of solidified material indicates its quasi-parabolically evolution in time. Silicon carbide particles will supply a part of the critical size of crystallization grain.

Data given in table 3 allow the determination of the solidification coefficient by using the law of the square root:

$$k = \frac{\delta}{\sqrt{t}}, \tag{8}$$

Table 4

The average thickness of the solidified layer measured in different directions

Material		Side			Diagonal		
	t, s	5	10	15	5	10	15
Al – 1.5 Mg		6.8	12.2	16.2	14.1	20.4	22.4
Al – 1.5 Mg/5% SiC$_p$	$\delta \cdot 10^{-3}$ m	8.8	12.9	17.4	16.4	22.6	25.3
Al – 1.5 Mg/10% SiC$_p$		9.7	14.5	18.6	19.4	25.3	29.1

The solidification coefficient depends on both the thermal parameters of the process (temperature, latent heat) as well as the configuration of the cooling surface.

On the basis of the determined values of *k* coefficient it was calculated (Table 5) and plotted the solidification rate and how this parameter varies on the direction of solidification (Fig. 2), for *t* = 15 s. Solidification rate can be obtained by the derivation of δ in relation to *t*:

$$v_{sol} = \frac{d\delta}{dt} = \frac{k}{2\sqrt{t}} \, .$$

Table 5

The average values of the solidification coefficient

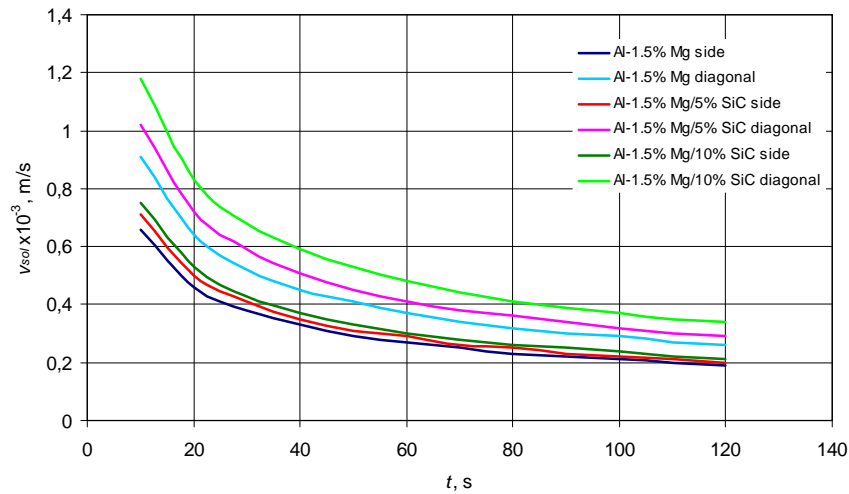| Material	δ·10^{-3}, m (side)	δ·10^{-3}, m (diagonal)	k_s·10^{-3}, m/s$^{1/2}$	k_d·10^{-3}, m/s$^{1/2}$
Al – 1.5 Mg	16.2	22.4	4.2	5.8
Al – 1.5 Mg/5% SiC$_p$	17.4	25.3	4.5	6.5
Al – 1.5 Mg/10% SiC$_p$	18.6	29.1	4.8	7.5

Fig. 2. The variation in time of the solidification rate.

In time, solidification velocity varies hyperbolically being great at first, when the temperature of the mould is low (Biot criterion that characterizes the intensity of heat exchange has very high values), but decreases rapidly in time as the mould thermal loading. Solidification velocity has increasing values as it grows the proportion of SiC particles in matrix.

3. Conclusions

■ The mixtures of the system Al – SiC$_p$ represent an interesting category of materials which have specific properties and good resistance to wear under the abrasive friction.
■ By using a liquid matrix such mixtures are obtained with difficulty due to the non-wetting conditions existent in the system.
■ The agglomeration of large sized particles of SiC favoured increasing of the sedimentation velocity.

■ The presence of silicon carbide particles in aluminium melts increases the dynamic apparent viscosity with negative effect on flow capacity. Fluidity of the mixtures decreases with increasing volume fraction of reinforcement particles and their tendency to agglomerate.

■ The presence of the additional material influence the appearance and the growth of solid phase by changing heterogeneous germination conditions and thermal properties of the material.

■ Because the silicon particles have a segregation tendency by sedimentation, the increase of the solidification rate has a favourable effect on the uniform distribution of the complementary material in the matrix. In order to control the degree of segregation and therefore the non-uniformity of the particle distribution in the matrix, the sedimentation rate would be related to the speed of solidification of the metallic material.

R E F E R E N C E S

[1] *M. Singla, D. Deepak Dwivedi, L. Singh, and V. Chawla*, "Development of aluminium based silicon carbide particulate metal matrix composites", in Journal of Minerals and Materials Characterization and Engineering, **vol.8**, no.6, June 2009, pp. 455–467

[2] www.NSM Archives-silicon carbide,

[3]. *S. Balasivanandha Prabu, L. Karunamoorthy, S. Kathiresan, B. Mohan*, "Influence of stirring speed and stirring time on distribution of particles in cast metal matrix composite", in Journal of Materials Processing Technology, **vol. 171**, no. 2, January 2006, pp. 268-273

[4] *R. Davies Joseph*, Aluminum and Aluminum Alloys, ASM International, 1993.

[5]. *V. Laurent, D. Chatain, and N. Eustathopoulos*, "Wettability of SiC by aluminium and Al-Si alloys", in Journal of Materials Science, **vol**. **22**, no. 1, January 1987, pp. 244-250

[6]. *P.K. Rohatgi*, "Cast Metal Matrix/Composites", in Metals Handbook, Ninth Edition, vol. 15, Casting, ASM International, 1988 pp. 840-854

[7]. J. Hashim, L. Looney, M.S.J. Hashim, "The wettability of SiC particles by molten aluminium alloy", in Journal of Materials Processing Technology, **vol**. **119**, no.1, December 2001, pp 324-328

[8]. G. Neagu, Fl. Ştefănescu, A. Mihai, I. Stan, I. Odagiu, "Theoretical and Practical Aspects Regarding Al - SiC Particles Composites Obtaining by Casting", in State Phenomena **vol**. 188 2012, pp. 314-317

[9]. *M. Kobashi, T. Choh*, "The wettability and the reaction for SiC particles/Al alloy system", in Journal of Materials Science, **vol. 28**, no. 3, February 1993, pp. 684-690

[10] *N. Eustathopoulos, J.C. Joud and P. Desre*, "The wetting of carbon by aluminium and aluminium alloy", Journal of Materials Science, **vol. 9**, no. 8, August 1974, pp. 1233–1242.

[11] *A. T. Dinsdale, P. N. Quested*, "The viscosity of aluminium and its alloys-a review of data and models", in Journal of Materials Science, vol. 39, no.24, 2004, pp. 7221-7228

[12]. *A. Ourdjini, K.C. Chew, and B.T. Khoo*, "Settling of silicon carbide particles in cast metal matrix composites, in Journal of Materials Processing Technology, **vol.116**, no.1 October 2001, pp. 72-76