Theoretical considerations regarding upward velocity of graphite spherical particles in pure aluminum during melting

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Abstract. The paper presents some theoretical aspects regarding the upward velocity of particles in the cast aluminum - graphite composites. Many types of particles are used to analyze the influence of shape factor on upward velocity in aluminum melts. The calculations were made by using modified Stokes’ equation for different Reynolds number attached to graphite particles. At temperature 700-900°C the upward velocity increases about 8.5%. The viscosity of multiphase fluids depends on many factors such as: the properties of aluminum melt and complementary material; temperature of the metallic bath; concentration, shape, fineness of graphite particles and degree of agglomeration of the dispersed phase.

1. Introduction
Metal matrix composites are an engineered combination between metals and some complementary materials (particles or fibres) in order to obtain certain improved properties in operating conditions [1]. Aluminum alloys associated with graphite have been received with considerable attention due to the potential of using these materials for aerospace or electrical applications [2]. A large variety of anti-friction parts can be obtained, graphite particles acting as a lubricant in the contact area (automotive components, bearings etc). Also, aluminum-graphite composites are inexpensive and lightweight materials with high-temperature properties. In addition, they have good machinability, a high thermal or electrical conductivity, and relatively low values of the thermal expansion coefficient [3, 4, 5, 6]. Although gravitational casting seems to be the cheapest method to obtain these materials, two important problems must be solved during processing.

Firstly, the wetting conditions are difficult to achieve in the aluminum-graphite system. The values of contact angle reported in different conditions, at conventional casting temperatures, reach up to 160° for aluminum and its alloys [7,8]. The presence of an oxide layer at the surface of the melt favours non-wetting conditions in the system. The thin layer of alumina, with an amorphous structure, appears at temperatures above 450°C [9]. At high- temperature aluminum reacts with alumina layer and forms a volatile compound (Al₂O) [10]. Also, graphite can react with aluminum and form aluminum carbide, especially at temperatures above 627 °C. Al₄C₃ is a brittle compound, highly sensitive to moisture contact and reduce mechanical properties and thermal conductivity. The appearance of carbide may be limited by the presence of silicon in the aluminum melt [11, 12, 13].

To improve the wetting conditions is recommended: alloying of the melt with superficial active elements (magnesium, titanium, lithium etc.), overheating of the melt, coating of graphite particles with nickel or copper, and preheating of complementary material.
Secondly, after incorporation in the aluminum melt, graphite particles have an upward movement due to the difference of density between components.

2. Theory

The laminar movement of a spherical graphite particle through resting aluminium melt is described by Stokes’ equation:

\[ v_{St} = \frac{2}{9} \frac{g r_p^2 (\rho_l - \rho_p)}{\eta} \]

(1)

Where: \( g \) is gravitational acceleration, \( r_p \) – radius of a particle; \( \rho_l, \rho_p \) – melt and particle densities, respectively; \( \eta \) - dynamic density of aluminum melt.

At constant pressure, dynamic viscosity decreases with temperature. This dependence can be analysed by using Arrhenius’ equation [14]:

\[ \eta = \eta_0 e^{\frac{E_v}{RT}} \]

(2)

Where: \( \eta_0 \) is the viscosity of the liquid aluminum at melting point (\( \eta_0 = 0.257 \) mPa·s), \( E_v \) - the activation energy for viscous flow (\( E_v = 13.08 \) kJ mol\(^{-1}\)); \( T \) – the overheating temperature; \( R \) – gas constant.

At different temperatures the density of the aluminum melt can be determined by Luca’s relationship [7]:

\[ \rho_{Al} = 2.369 - 3.11 \times 10^{-4} (T - T_{melt}) \]

(3)

Where \( T_{melt} \) is the melting point of aluminum.

3. Calculations and Results

The calculated values of Stokes velocity for a spherical graphite particle at different overheating temperatures are presented in Table 1.

| \( T \), °C | \( \eta \times 10^3 \), N·s/m² | \( \rho_l \), kg/m³ | \( r_p = 25 \) µm | \( r_p = 50 \) µm | \( r_p = 75 \) µm | \( r_p = 100 \) µm |
|---|---|---|---|---|---|---|
| 700 | 1.294 | 2357 | 3.75 | 15.01 | 33.78 | 60.05 |
| 800 | 1.113 | 2325 | 3.98 | 15.93 | 35.85 | 63.73 |
| 900 | 0.983 | 2294 | 4.08 | 16.33 | 36.74 | 65.31 |
| 1000 | 0.884 | 2263 | 4.06 | 16.23 | 36.51 | 64.9 |

Obviously, at the same temperature, the upward velocity grows when the radius of the particle increases. It can be seen an increase in particle velocity by approximately 8.5%, regardless of its size, in the temperature range of (700-900) °C. This increase can be explained especially by the effect of the temperature on the melt viscosity. The upward velocity reaches a maximum value at 930-940°C. After this range of temperature, the analysed parameters begin to decrease slowly. Consequently, at high temperature, the effect of reducing density passes in the foreground. It should be noted that up to 1000°C the velocity reduction does not exceed 0.5%.
Figure 1. Maximum value of upward velocity for a spherical graphite particle ($r_p = 25 \mu$m)

Because Stokes equation neglects the inertial effect (the scenario that can be applied only for very small velocities), other shapes of particles, or the presence of several particles in the melt, there is a difference between the calculated values and the experimental data. Also, must consider that graphite particles tend to agglomerate due to the non-wetting conditions.

To approach the calculated values by real situations it can be used some modified equations [15].

So, if it is taken into account the inertial effect of the motion, Oseen equation can be used [16].

\[
    v_p = \frac{v_{St}}{1 + \frac{3}{8} \text{Re}_p}
\]

Where; \( \text{Re}_p \) is Reynolds number of particles.

The modified values of Stokes’ velocity for a spherical particle of graphite ($r_p = 25 \mu$m) are presented in Figure 2. From the obtained data it results that at Reynolds number of the particles close to unity, the velocities are less than 75% of the initial one, regardless the melt temperature (Table 2).

Figure 2. Modified values of Stokes velocity taking into account the inertial effect.
Table 2. Loss of velocity due to the inertial effect

| $Re_p$ | $T = 700^\circ C$ | $T = 800^\circ C$ | $T = 900^\circ C$ | $T = 1000^\circ C$ |
|--------|-----------------|-----------------|-----------------|-----------------|
| 0.2    | 6.89            | 6.90            | 6.93            | 7.06            |
| 0.4    | 12.96           | 12.97           | 13.00           | 13.12           |
| 0.6    | 18.29           | 18.30           | 18.32           | 18.44           |
| 0.8    | 23.01           | 23.02           | 23.04           | 23.14           |

The deviations that arise due to the presence in a metallic melt of some particles with an irregular shape are highlighted by the equation:

$$v_p = \frac{2}{9} g \frac{Re \rho_f - \rho_p}{\eta} \frac{r_{eq} - \rho_p}{\lambda}$$

(5)

Where: $r_{eq}$ is the equivalent radius of the the particle (a spherical particle radius with the same volume), $\lambda$ - a shape factor of a particle. For $Re_p < 0.2$, Chowdhury-Fritz equation can be used [17]:

$$\lambda = \frac{1}{1 + 0.862 \log \psi}$$

(6)

$\psi$ - Where sphericity of the particle:

$$\psi = \frac{\text{surface area of sphere of same volume}}{\text{surface area of particle}}$$

(7)

The calculated values of equivalent radius, sphericity and shape factor for some types of particles are presented in Table 3 ($r_p = 50 \mu m$)

Table 3. The values of equivalent radius, sphericity and shape factor

| Particle type | $r_{eq} \cdot 10^2$, m | $\psi$       | $\lambda$     |
|---------------|------------------------|--------------|---------------|
| Cylinder, $r = r_p$, $h = 2r_p$   | 0.57                   | 0.873580     | 1.053293      |
| Cone, $r = r_p$, $h = 2r_p$      | 0.40                   | 0.778674     | 1.121887      |
| Truncated cone, $R = r_p$, $r = r_p/2$, $h = 2r_p$ | 0.48                   | 0.842706     | 1.068453      |
| Ellipsoid, $a = r_p$, $b = 2r_p$ | 0.40                   | 0.911580     | 1.035901      |

In Figure 3 are comparatively presented the values of upward velocity in aluminum melt ($T_{melt} = 700^\circ C$) for the analyzed particles. It can be observed that the maximum value of upward velocity, due to the high value of equivalent radius, is characteristic for particles with a cylindrical shape. Similarly, conical or ellipsoidal particles have the lowest upward velocities. Consequently, in operating conditions, through the graphite concentrating in the contact area, cylindrical particles can provide improved properties of wear resistance. Conical or ellipsoidal particles are recommended for parts with a relatively uniform distribution of graphite.

If it takes into account the presence of other graphite particles in the aluminum melts, the following equation for upward velocity could be used:
\[ v_p = \frac{v_{St}}{1 + k\phi^3} \]  
(8)

Where; \( k \) is the distribution coefficient of particles, \( k = 1.3 \ldots 1.9; \) \( \phi \) – the volume fraction of particles.

\[ \eta_a = \eta(1 + 2.5\phi) \]  
(9)

Figure 3. Upward velocity in aluminum melts at (\( T_{melt} = 700^\circ C \)) for a particle with a shape of:
1 – Sphere; 2 – cylinder; 3 – cone; 4 – truncated cone; 5 - ellipsoid

The presence of more particles in metallic bath leads to reduced values of upward velocities at any temperatures (Figure 4). This can be a problem when seeking to obtain composites with a controlled distribution of the dispersed material.

Figure 4. The effect of graphite particles concentration in aluminum melts on upward velocity.

In Stokes equation, the variants have not be taking the mixtures between aluminum melts and graphite particles in the consideration. This will be clear in reality a multiphase fluid characterized by an apparent dynamic viscosity.

Many factors are affecting on the viscosity of multiphase fluids such as: the properties of aluminum melt and complementary material, temperature of the metallic bath, concentration, shape, fineness of particles, and degree of agglomeration of the dispersed phase [18]  

Starting from Einstein equation for a dilute suspension of spherical particles (less than 2% vol.):
Taking considering the collisions between particles and their agglomeration as well as hydrodynamic interactions, a theoretical extension based especially on experimental data is recommended for high concentration of complementary material [19, 20]

\[
\eta_a = \eta\left(1 + k_1C + k_2C^2 + k_3C^3 + \ldots\right)
\]

(10)

Where; \(k_1, k_2\) and \(k_3\) are coefficients of interactions between particles with different evaluations in literature.

Thus, for a volume fraction of complementary material less than 0.1, the next equation can be used:

\[
\eta_a = \eta\left(1 + 2.5\phi + 10.25\phi^2\right)
\]

(11)

Where \(\phi\) is the volume fraction of solid particles.

For volume fractions of particles up to 0.6, Thomas proposed for the apparent dynamic viscosity the next semi-empirical relationship [21]:

\[
\eta_a = \eta\left[1 + 2.5\phi + 10.25\phi^2 + 0.00273\exp(16.6\phi)\right]
\]

(12)

By using equation (8), the calculated values of upward velocity for different values of dynamic viscosity are presented in Figure 5.

**Figure 5.** Upward velocity for different volume fractions of graphite particles in aluminum melt.

It is obvious that the specific values of apparent dynamic viscosity for aluminum – graphite mixtures lead to slowing of upward velocity and has unfavourable effects on the lubrication conditions on the surfaces subjected to wear. The reduction in upward velocity value can reach approximately 25%.

Another equation for the dynamic viscosity of multiphase fluids based on the cumulative influence of size, shape and volume fraction was developed by Wang [22]:

\[
\eta_a = \eta\left[\frac{\xi(1 + d_p^{0.95})}{0.01 + 37.35d_p^{0.95}\phi}\right]
\]

(13)

Where; \(\xi = 1\) for a spherical particle (high/diameter ratio); \(d_p\) diameter of particle in centimetre.
The distribution of graphite particles in different upward velocities can result in collide during displacement through the aluminium melt. To get a cluster (Fig. 6), the formula to be used:

$$\Delta E_\sigma < 0$$  \hspace{1cm} (14)

Where; $\Delta E_\sigma$ is the surface energy variation, is fulfilled due the non-wetting condition from system.

Figure 6. Sketch shown the possible cluster from graphite particles

4. Conclusions:
1- The upward velocity increases when radius of particle increases at the same temperature.
2- When regardless the size of particle, the upward velocity grows with increasing the temperature and reaches a maximum value at 930-940°C.
3- The particles with cylindrical shape have the maximum value of upward velocity comparing with other shapes due to the high value of equivalent radius.

References
[1] B. Vijaya Ramanath, C. Elanchezhian, RM. Annamalai, S. Aravind, T. Sri Ananda Atreya, V. Vignesh and C. Subramanian, 2014 “Aluminium Metal Matrix Composites” - A Review, Rev. Adv. Mater. Sci. 38, pp. 55-60.
[2] S. Rawal. 2001 “Metal matrix composites for space applications”, JOM, 53 (4), p. 14-17.
[3] D.A Saheb, 2011 “Aluminium silicon carbide and aluminium graphite particulate composite”, ARPN Journal of Engineering and Applied Sciences, 6(10), p. 41-46.
[4] T.P. Rajan, R.M. Pillai and B.C. Pai, 1998 “Reinforcement coatings and interfaces in aluminium metal matrix composites”, Journal of Materials Science 33, p. 3491-3503.
[5] A. Mewar and K.K.S. Mer, “Wear characterization of aluminium graphite composites synthesized in open hearth furnace with manually controlled stirring method”, Proceedings of International Conference an Innovation & Research in Technology for Sustainable Development, 1-3 November 2012, India, p. 51-53.
[6] P. Sharmaa, D. Khandujaa and S. Sharmab, 2016 “Dry sliding wear investigation of Al6082/Gr metal matrix composites by response surface methodology”, Journal of Materials Research and Technology 5 (1), p. 29-36.
[7] N. Eustathopoulos, J.C. Joud and P. Desre, 1974 “The wetting of carbon by aluminium and aluminium alloys”, Journal of Materials Science 9, p. 1233-1242.
[8] E.A. Pastukhov, V.P. Chentsov, A.V. Kiselev, L.E. Bodrova, A.V. Dolmatov, E.A. Popova, S.A. Petrova and R.G. Zacharov, 2006 “Wetting of graphite surface by the aluminium alloys melts”, The Fourth International Conference on Mathematical Modeling and Computer Simulation of Materials Technologies, Ariel, Israel, p. 178-181.
[9] K. Keehyun, 2014 “Formation of fine clusters in high – temperature oxidation of molten aluminium”, Metallurgical and Materials Transactions A 45A, p. 3650-3660.
[10] S. Bao, K. Tang, A. Kvithyld, T. Engh, and M. Tangstad, 2012 “Wetting of pure Aluminium on graphite, SiC and Al₂O₃ in aluminium filtration”, *Transactions of Nonferrous Metals Society of China* **22**, p. 495-504.

[11] T. Etter, P. Schulz, M. Weber, M. Wimmler, J.F. Loffler and P.J. Uggowitzer, 2007 “Aluminium carbide formation in interpenetrating graphite/aluminium composites”, *Materials Science and Engineering*, A**448**, p. 1-6.

[12] J.K. Chen, 2013 “Thermal properties of aluminium-graphite composite by powder metallurgy”, *Composites: Part B* **44**, p. 698-703.

[13] G.G. Sozhamannan and S. Balasivananda Prabu, 2009 “Influence of interface compounds on bonding characteristics of aluminium and silicon carbide”, *Materials Characterization*, **60**, p. 986-990.

[14] T. Dinsdale and P.N. Quested, 2004 “The viscosity of aluminium and its alloys-A review of data and models”, *Journal of Materials Science* **39**, p. 7221-7228.

[15] R.C. Iyengar, “Grow and elimination of inclusions in steel”, Ph.D. Thesis, Carnegie-Mellon University, 1970.

[16] A. Venkatalaxmi, B. Sri Padmavati and T.Amaranath, 2007 “A general solution of Oseen equations”, *Fluid Dyn. Res.*, **39**(7), 595-606.

[17] K. C. R. Chowdhury, W. Fritz, 1959 “Sinkversuche mit isometrichen Teilchen in Flüssigkeiten”, *Chemical Engineering Science*, **11**(2), P. 92-98.

[18] K.R. Ravi, R.M. Pillai, B.C. Pai and M. Chakraborty, 2007 “Influence of interfacial reaction on the fluidity of A356 Al-SiCp composites – A theoretical approach”, *Metallurgical and Materials Transactions A*, **38A** (10), p. 2531-2539.

[19] N.S. Cheng and A.W.K. Law, 2003 “Exponential formula for computing effective viscosity”, *Powder Technology*, **129**(1-3), p. 156-160.

[20] K.R. Ravi, R.M. Pillai, K.R. Amaranathan, B.C. Pai and M. Chakraborty, 2008 “Fluidity of aluminium alloys and composites: A review”, *Journal of Alloys and Compounds*, **456**, p. 202-210.

[21] D.G. Thomas, 1965 “Transport characteristics of suspension: VIII. a note on the viscosity of Newtonian suspensions of uniform spherical particles”, *Journal of Colloid Science*, **20** (3), p. 267-277.

[22] J. Wang, O. Guo, M. Nishio, H. Ogava, D. Shu, K. Li, S. He and B. Sun, 2003 “The apparent viscosity of fine particle reinforced composite melt”, *Journal of Materials Processing Technology*, **136**(1-3), p. 60-63.