Comparative Analysis of Theories of Thermocapillary Inclusion Motion Under Zero-Gravity Condition

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ABSTRACT

The motion of a fine viscous inclusion in a viscous medium under zero-*g* condition is subjected to theoretical examination. This motion can be qualified as an elementary act of the motion of fine phase particles in composition mixtures: suspensions, emulsions, foamed liquid metals, aerosols. It is interesting to notice, when carrying out a theoretical analysis of thermocapillary inclusion dynamics, that attention is usually centered on a description of the behavior of gaseous and liquid inclusions whereas the behavior of solid inclusions is left beyond any considerations. Such a situation can be understood since the theoretical basis commonly used gives no way to describe the behavior of solid inclusion, although the proper experiments are known now. An approach proposed here allows one to describe thermocapillary dynamics of solid, liquid and gaseous inclusions in the reduced gravity (without influence of any other exertions). It may be used in the interpretation of experiments and technological processes with fine particles.

Key words: thermocapillary motion, velocity of solid, liquid or gaseous inclusions, solid particle, drop or bubble in melt, suspension, emulsion, aerosol, foamed metal (material).

1. INTRODUCTION

The theory of hydrodynamic capillary processes under conditions with zero-*g* or reduced gravity is of great importance, at least, for the following cases. Firstly, such a theory will allow one to ascertain various events in the space technique and technology (for example, the behavior of liquid fuel with some inclusions – solid, liquid or gaseous ones – in space vehicles). Secondly, such a theory will focus the attention on the important part of more complex dynamic processes consisting of various other processes with different natures as well (for example, flows which are similar to the gravitation-thermal convection, the electric-magnetic convection both within fluid inclusions and in outer media). The behavior of suspensions in a non-uniform temperature field is very essential for most important scientific fields and technological branches (for example, the motion of refractory materials' particles or slag drops in metal melts). Although in literature there is the theory of motion of bubbles and drops, the absence of theory of the solid particles' thermocapillary motion leads to thought about the necessity of more careful verification of the fundamental equations describing dynamics of foreign inclusions in a viscous medium with a temperature gradient.

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Some novel prospects in materials science, conditioned by the emergence of space-branch technology, motivated us to test carefully the validity of the scientific basis of methods used for studying and processing of materials under zero-g conditions.

It is known that reduced ("scaled") accelerations (i.e. the portions of the acceleration due to gravity on the Earth's surface) at a relatively low height (240 km) are equal to very small quantities: from outer gravitation $4.3*10^{-7}$, from inner gravitation $3.3*10^{-8}$, from control by orientation in flight $4.3*10^{-7}$, from light pressure $3.1*10^{-9}$ /1/. Thus, the proper theoretical considerations of the processes at heights a few hundred kms above Earth's surface and under the exact zero-g condition will be practically the same. The present paper is aimed to give a more precise definition mainly to the above-mentioned problem.

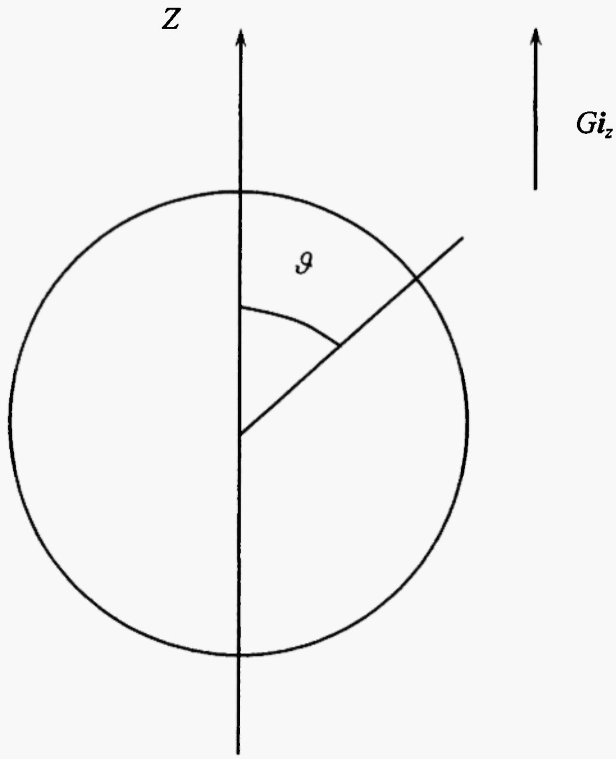

Fig. 1: Model used in the theories analyzed.

2. MODEL. THEORETICAL STUDY

We use here the well-known conventional simple model, that is: an inclusion is immersed in a viscous fluid unlimited (Fig. 1). An inclusion material is free from soluble elements. An inclusion is a smooth unchanged sphere that does not interact chemically (or by diffusion) with a medium. There is no convection in a medium. The heat flow is unidirectional. The thermal conductivities and the viscosities of the inclusion and medium are uniform and temperature independent. The temperature field in a medium (without the inclusion's influence) has a constant gradient. Such conditions were adopted in a lot of studies /2-12/. In the work /13/ the model has presupposed the motion in the infinite medium under joint thermocapillary/gravity influence. In the thermocapillary part of the theory /13/ the method, which takes into account the dependency of the surface (interfacial) energy on the location, was outlined previously. The gravity is absent in the model considered. Also assume that the Capillary, Reynolds and Marangoni (Pèclet) numbers are small (the Pèclet number in thermocapillary processes is often called the Marangoni number).

Let us consider from the beginning the behavior of a viscous inclusion in a viscous medium. The following equations for a steady regime are the governing ones:

$$\nabla p = \eta \Delta \boldsymbol{v} \tag{1}$$

$$\nabla p_i = \eta_i \Delta_i \boldsymbol{v}_i + \boldsymbol{f}_i \tag{2}$$

$$\nabla \boldsymbol{v} = 0 \tag{3}$$

$$\nabla \boldsymbol{v}_i = 0 \tag{4}$$

In eqs. (1)-(4) p and p_i are the pressures, $\boldsymbol{v}$ and $\boldsymbol{v}_i$ are the velocities in fluids, η and η_i are the viscosities (values with index "i" belong to an inclusion, values without "i" belong to a medium), $\boldsymbol{f}_i$ is the "thermocapillary force" introduced conditionally, namely, the "density" of the whole force due to the exertion of non-uniform interfacial (surface) tension ***F=Fi*** on an inclusion/medium interface ("thermocapillary force"). The "thermocapillary force" is not true "body force": it acts on inclusion's surface. It is worth to remember that the "buoyancy (Archimedes) force" in the gravitation conditions is the difference between the superficial forces, which act from the surrounding fluid on inclusion's surface, and the body force, which is the result of the action of gravitation on the inclusion's mass (the latter force is the exact body force). But the final results of the proper effects (both thermocapillarity and buoyancy) on the "closed body" (inclusion) as a whole is equivalent to the effect of some "body force" acting on the inclusion as a whole. Eqs. (1), (2) are the Stokes equations, eqs. (3), (4) are the continuity equations.

The average "density" of the thermocapillary force in the case of a spherical inclusion is

$$f_i = \frac{3F}{4\pi r_i^3} \tag{5}$$

where /14/

$$F = -4\pi r_i^2 \sigma_T \frac{\partial T}{\partial z}\ , \tag{6}$$

$\sigma_T = \partial\sigma/\partial T$ is the interfacial tension temperature coefficient, T is the temperature, $\partial T/\partial z$ is the value of the temperature gradient inside an inclusion, z is the coordinate, r_i is the inclusion's radius; it should be noticed that in equations for thermocapillary force in /14/ (see eq. (4) in /14/) the numerical factor must be substituted: $\pi^2 \rightarrow 4\pi$, i.e. eq. (6) in the present work is true instead of eq. (4) in /14/.

Let us express the interfacial (surface) tension as it was made in /3/

$$\sigma = \sigma_0 + \sigma' r_i \cos\vartheta \tag{7}$$

where σ_0 is the value of the interfacial (surface) tension at the point of the inclusion center (at $z=z_1$ or z_2, Fig. 2),

$$\sigma' = \sigma_T \frac{\partial T}{\partial z} \tag{8}$$

Taking into account the well-known equation for the temperature inside a sphere in the condition of the adopted model /15/, one can write

$$\sigma' = \sigma_T G \frac{3k}{2k + k_i} \tag{9}$$

where $G=[\partial T/\partial z]_\infty$ is the value of the temperature gradient far from an inclusion, k and k_i are the thermal conductivities of the medium and the inclusion, respectively.

Therefore, let us substitute eq. (6) in eq. (5), then let us substitute eq. (5) in eq. (2), and, finally, let us introduce the force term f_i under the nabla operation symbol with multiplication by coordinate z (just as it was made with other hydrodynamic equations in /16/).

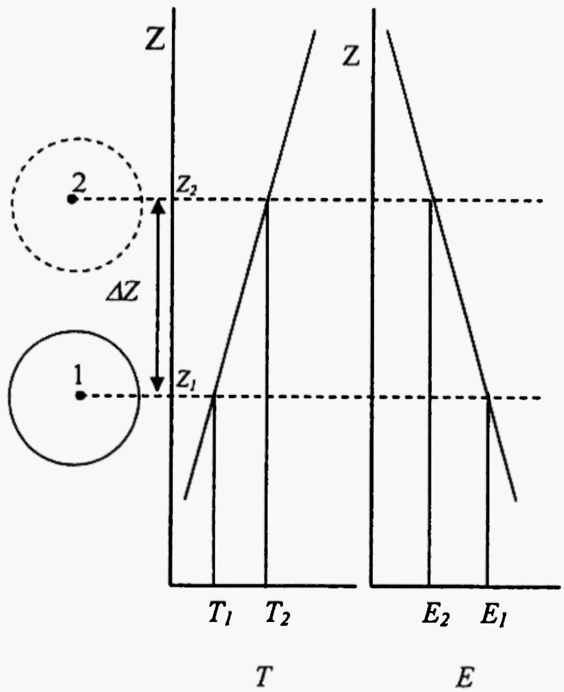

Fig. 2: Scheme of the "interfacial energy depending on location": two close consequent locations of an inclusion in a medium with a temperature gradient (temperature T is a linear increasing function of height).
(1) an arbitrary location (an inclusion center at $z=z_1$);
(2) a consequent location of this inclusion (an inclusion center at $z=z_2=z_1+\Delta z$).
E is the interfacial (surface) energy. E_1 and E_2 are the energies corresponding to locations of an inclusion center at $z=z_1$ and z_2, respectively.

As a result we obtain

$$\nabla\left(p_i+\frac{3\sigma'}{r_i}z\right)=\eta_i\Delta\mathbf{v}_i \tag{10}$$

The only distinguishing feature of the set containing eqs. (1) and (10) in comparison with the proper conventional set, which one usually obtains, is the presence of the term $(3\sigma'/r_i)z$ under the nabla operation symbol in the left side of eq. (10). This term reflects the appearance of the interfacial (surface) energy depending on location in a non-uniform temperature field and, consequently, arising the motive force acting on the inclusion as a whole by changing the integral surface energy E in coordinate z which determinates location of the inclusion's center (see Fig.2).

The boundary conditions are expressed by the equations set (in spherical coordinates)

$$v_r\left(_{r=r_i}=v_{ir}\left(_{r=r_i}=0\right.\right., \tag{11}$$

$$v_\vartheta\left(_{r=r_i}=v_{i\vartheta}\left(_{r=r_i}\right.\right., \tag{12}$$

$$\left[p_i-p+2\eta\frac{\partial v_r}{\partial r}-2\eta_i\frac{\partial v_{ir}}{\partial r}-\frac{2\sigma}{r}\right]_{r=r_i}=0, \tag{13}$$

$$\left[\eta_i\left(\frac{1}{r}\frac{\partial v_{ir}}{\partial\vartheta}+\frac{\partial v_{i\vartheta}}{\partial r}-\frac{v_{i\vartheta}}{r}\right)-\eta\left(\frac{1}{r}\frac{\partial v_r}{\partial\vartheta}+\frac{\partial v_\vartheta}{\partial r}-\frac{v_\vartheta}{r}\right)-\frac{1}{r}\frac{\partial\sigma}{\partial\vartheta}\right]_{r=r_i}=0 \tag{14}$$

Eq. (11) denotes the absence of the radial components of the velocities. Eq. (12) denotes the continuity of the tangent components of the velocities, i.e. the absence of slip. Eq. (13) denotes the continuity of the radial components of the viscous stresses tensor (taking into account the curvature of the inclusion surface by the term $2\sigma/r$). Eq. (14) denotes the continuity of the tangent components of the viscous stresses tensor (taking into account the variation of σ by the term $(1/r)\partial\sigma/\partial\vartheta$).

The solution of the problem expressed by eqs. (1), (3), (4), (10)-(14) yields all values, which characterize the velocities and pressures. Let us focus on the equation obtained for the velocity of motion of a viscous inclusion ("drop") as a whole:

$$u=-\frac{2\sigma_T G r_i(4+3\alpha)}{(2+\beta)\eta(2+3\alpha)} \tag{15}$$

where $\alpha=\eta_i/\eta$, $\beta=k_i/k$.

Let us make the limiting transition $\alpha\to\infty$ in eq. (15) and substitute σ' (9) into it. Then eq. (15) turns into the equation for the velocity of a solid inclusion

$$u=-\frac{2\sigma' r_i}{3\eta}=\frac{2r_i\sigma_T}{3\eta}\frac{\partial T}{\partial z}=-\frac{2r_i\sigma_T Gk}{\eta(2k+k_i)} \tag{16}$$

The double limiting transition $\alpha\to 0$, $\beta\to 0$ in eq. (15) yields the equation for the case of a bubble

$$u=-\frac{2\sigma_T G r_i}{\eta} \tag{17}$$

Now let us apply the formula for the thermocapillary velocity of a spherical fluid inclusion from the well-known theory by Young, Goldstein and Block /2/. The thermocapillary part of the formula for velocity of a viscous inclusion after the correction of some errors (see, for example, /5,7,10-12/) is the following

$$u_{YGB}=-\frac{2\sigma_T G r_i k}{(2\eta+3\eta_i)(2k+k_i)} \tag{18}$$

3. DISCUSSION

Let us apply the limiting transition $\eta_i\to\infty$ to eq. (18)

$$\lim u_{YGB}\big|_{\eta_i\to\infty}=u_{YGB,s}=0, \tag{19}$$

i.e. according to the theory by Young *et al.* /2/ the thermocapillary velocity of a solid inclusion $u_{YGB,s}$ is equal to zero. The assertion, expressed by eq. (19), that the thermocapillary motion of a solid inclusion is absent, contradicts both experiments and eq. (16) derived above. The thermocapillary motion (drift) of the solid particles (agglomerates) was observed under zero-g conditions (on the Space Shuttle Columbia during the

US Microgravity Payload 4) /17/. The thermocapillary motion of solid particles was observed under gravitation conditions as well /18-20/ long before the space experiment named. The principal question to eq. (18) is the following one: how the behavior of eq. (18) at limiting transition $\eta_i \to \infty$ (i.e., in the case of a solid inclusion) can be interpreted? The answer will be given below after the comparison between two theoretical approaches which can be represented by works /2/ and /13/.

The adequacy of these approaches may be judged by comparing some experimental data (not only on dynamics of solid inclusions) with calculations based on the proper theoretical equations. The case of the velocities of the solid inclusion derived in frames of the approaches /2/ and /13/ was considered above. Let us now consider the experimental data on the motion of air bubbles in silicon oil /21/. These data were obtained during an International Microgravity Laboratory mission (IML-2). In Fig. 3 in work /21/ the scaled bubble's velocities at various values of the Marangoni numbers Ma are presented (here the scaled velocity is the fraction where the numerator is the experimental value of the bubbles' velocity u_e and the denominator is $u_{YGB,b} = \sigma_T G r_i /(2\eta)$, which is obtained through the limiting transitions $\eta_i \to 0$, $k_i \to 0$ in eq.(18)). If we make the extrapolation of the dependency of the scaled bubbles' velocity $u_e / u_{YGB,b}$ on the Marangoni number depicted in above-mentioned Fig. 3 in /21/ then the result of such an extrapolation will be the following: for the value, let us say, Ma≈10^{-2} one obtains

$$u_e / u_{YGB,b} \approx 3.5.$$

As in the case of an air bubble according to eqs. (15) and (18) (at $\alpha \sim 0$)

$$u_b \approx 4 u_{YGB,b}, \tag{20}$$

another scaled velocity , in which the denominator is u_b (20), will be equal to 3.5/4~0.9. Thus, the calculation based on the equations derived in /13/ (see eq.(15) in the present paper) for the case Re<<1, Ma<<1 yields a result which is much closer to the value obtained by extrapolation of the independent experimental bubble's velocities than the calculation by theory /2/, i.e. with an equation which is obtained from eq.(18) at $\eta_i=0$, $k_i=0$.

By introduction of the "natural velocity scale" /22/

$$u_0 = \frac{|\sigma_T||G| r_i}{\eta}, \tag{21}$$

eq. (15) may be expressed by an equation with the variables reduced

$$U = \frac{u}{u_0} = -sign(\sigma_T G)\frac{2(4+3\alpha)}{(2+\beta)(2+3\alpha)} = \pm\frac{2(4+3\alpha)}{(2+\beta)(2+3\alpha)} \tag{22}$$

where the function *sign(x)* is determined by the argument's sign: *sign(x)*=1 at $x>0$ or -1 at $x<0$.

The physical meaning of the quantity u_0 (21) is the following: u_0 is the velocity of the imaginary solid inclusion with a radius r_i in the medium with the viscosity η and the absolute value of the temperature gradient $|G|$ in the infinity under conditions that the absolute value of the interfacial tension temperature coefficient is equal to $|\sigma_T|$ and $\beta=0$, i.e. the thermal conductivity of this inclusion is equal to zero (an imaginary "thermal insulator").

If $\sigma_T<0$ (this is a typical case), then the sign "+" in Eq. (22) corresponds to the situation with $G>0$, and the sign "-" corresponds to the situation with $G<0$. When considering the inclusion's behavior, many works had been based on theory /2/. Eq. (18) for the viscous inclusion motion velocity in the variables reduced has the form

$$U_{YGB} = \pm\frac{2}{(2+\beta)(2+3\alpha)} \tag{23}$$

(the application of signs here is the same as in eq. (22)).

The equations equivalent to eq. (23) or to equations derived from eq. (23) through transitions $\alpha \to 0$, $\beta \to 0$ were reproduced in various publications devoted to the "thermocapillarity" problems of a fluid inclusion motion without gravitation and any other body forces. One can be sure that the "misadventures" at derivation of eq. (23) arose due to the following reasons: the energetic factor reflecting the energy depending on location of an inclusion (namely, dependency of inclusion's interfacial energy on the coordinate) in a non-uniform temperature field was not taken into account in /2/.

Eq. (22) differs from the well-known eq. (23) by the factor (4+3α). Thus, the ratio of the above-mentioned velocities expressed by eqs. (22) and (23) is equal to

$$n = \frac{U}{U_{YGB}} = \frac{u}{u_{YGB}} = 4 + 3\alpha \qquad (24)$$

It is easy to see how this ratio behaves in the cases of inclusions existing in various states. In the case of a solid inclusion in any fluid medium at $\alpha \to \infty$ eq. (24) yields $n \to \infty$. For a bubble ($\alpha=0$) eq. (24) yields $n=4$. The case of a drop in an another liquid is an intermediate one: $4<n<\infty$.

Thus, for the case of a bubble ($\alpha=0$) the value n (24), i.e. the ratio of two velocities considered, is high, for the case of drops ($0<\alpha<\infty$) this value increases in accordance with linear rule, but for the case of a solid inclusion ($\alpha \to \infty$) the gap between the present approach and the approaches previously used (which is reflected by value n (24)), becomes catastrophic: $n \to \infty$! This leads us to consider the validity of determination of some values, among them σ_T , from experiments with drops and bubbles if equations such as (18) or (23) are used as the basis for computations.

The following circumstances should be emphasized. The fact that the value σ_T in the cases of drops and bubbles determined by equations like (18) or (23) is close to the value σ_T known from independent sources should not be regarded as encouraging. On the other hand, a gap between theoretical and experimental data should not be seen as discouraging. As practice shows, the values under consideration, being determined from similar or close sets of experiments, differ sometimes by an order of magnitude (see, for example, Table II in /8/). Starting from eq. (15) one can write for the interfacial tension temperature coefficient in the case of a solid inclusion in a viscous medium

$$\sigma_{T,s} = -\frac{u\eta(2+\beta)}{2Gr_i}, \quad (\alpha \to \infty,\ 0<\beta<\infty). \qquad (25)$$

Simultaneous solution of the set (1), (10), (3), (4) with the boundary conditions (11)-(14) with the replacement of the term $3\sigma' z/r_i$ in eq. (10) by the value $g(\rho-\rho_i)z$ (the "Archimedes force", where g is the acceleration due to gravity, ρ and ρ_i are the densities of a medium and an inclusion, respectively) yields the well-known classical law by Rybzcynski – Hadamard /23,24/ (this derivation is not adduced in the present paper; the analogous derivation is, for example, in /22/).

This examination, i.e. obtaining the Rybzcynski – Hadamard law through solution of the set (1), (10), (3), (4) after replacement of the motive force (as it is mentioned above:

$$3\sigma' z/r_i \to g(\rho-\rho_i)z)$$

may be an argument for the validity of the approach contained in /13/.

It is known that determination of the tension on the solid/liquid boundary has always been a problem. However, this parameter reflects a very important aspect of interface phenomena and is of great interest.

Note that experiments with solid inclusions in liquid media under non-uniform temperature, μ-*g* and zero-*g* conditions, might clarify some ambiguities and questionable items mentioned above.

Using eq. (15) it is easy to obtain the equations for the interfacial (surface) temperature coefficients, analogous to the coefficient $\sigma_{T,s}$ (25), for the cases of a drop and a bubble as well:

$$\sigma_{T,d} = -\frac{u\eta(2+3\alpha)(2+\beta)}{2Gr_i(4+3\alpha)},$$
$$(0<\alpha<\infty,\ 0<\beta<\infty;\ \text{drop}) \qquad (26)$$

$$\sigma_{T,b} = -\frac{u\eta}{2Gr_i}, \quad (\alpha=0,\ \beta=0;\ \text{bubble}) \qquad (27)$$

It is clear that eqs. (25)-(27) may be considered as approximate ones since the values η, η_i, k, k_i are considered constants.

For example, let us look at the expected velocities of the motion of a bubble in some molten metals, using eq. (17) with $k_i=0$ and setting verisimilar parameters. Let the main general parameters in the next examples be : $r_i \approx 10^{-4}$ m, $G \approx 10^4$ K/m.

For liquid Cu σ_T = -2.4*10^{-5} N / (m*K) /25/, η=3.33*10^{-3}Pa*s /26/, then

$$u = 1.44 \times 10^{-2} \frac{m}{s}.$$

For bubbles in other melts the analogous proper computations yield:

in molten Al $u = 2.5 \times 10^{-2} \frac{m}{s}$,

in molten Fe $u = 1.73 \times 10^{-2} \frac{m}{s}$,

in molten Ag $u = 1.2 \times 10^{-2} \frac{m}{s}$.

3. CONCLUSION'

What should be done in the field of thermocapillary particle motion as a next step?

The problem of improving the accuracy of experiments is very important. The problem is to separate, with good reliability, the influences of temperature and concentrations of impurities /8,27/. Non-uniformity of the viscosity due to the non-uniform temperature may play its part as well /14/; thus it is necessary to take this important factor into consideration in the future.

The theory, which might be the scientific basis of the research method, ought to be developed more rigorously than it has been until now. Many essential elucidations for understanding interaction between foreign inclusions (mainly, solid particles) and surrounding fluid phases could be obtained from the proper space-experiments designed and organized most efficiently.

When analyzing the dynamics of a bubble during its formation at orifice submerged in a liquid with temperature gradient, it is necessary to account for the bubble's changed shape during three of four stages of this process (at "under critical growth", "critical growth" and "necking" /28/). During the stages named, the thermocapillary force must be described by taking into account the shape of a bubble, which is very different from the sphere, i.e. not by eq. (6). However, when a bubble comes off and becomes spherical, eq. (6) may be used for description of the thermocapillary force acting on a bubble.

The chemical content of gas in a bubble can be of great importance /12/, in particular, for the state of the surface of the inclusion and, hence, for a type of contact inclusion/medium. For example, if a bubble which is submerged in Al melt contains oxygen, then a thin layer of alumina might appear on the inclusion's surface; thus a contact between an inclusion and a medium will transform from the gas/liquid type to the solid/liquid type (namely, Al_2O_3 (sol.)/Al(liq.)). This must determine the choice between eqs. (16) and (17) or their modifications for the analysis of inclusion's behavior.

With regard to the technological applications the right understanding and adequate mathematical expressions, for example, on the behavior of carbon particles in metallurgical melts under various non-uniformities /29/ is a very important factor in high-quality metallurgy. The theoretical approach /13/ might be useful in the analysis of complex processes where the thermocapillary motion might play an essential part.

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