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COMPUTER CALCULATIONS OF DIFFUSION FROM A SOLID INCLUSION IN THE SURROUNDING LIQUID CURRENT-CARRYING METAL UNDER ELECTRIC CURRENT ACTION

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ABSTRACT

A novel approach was developed for the problem of mass transfer from a solid metal inclusion surrounded by the liquid current-carrying metal. The primary mechanism of this process is diffusion under electric current action. The named phenomenon provides more effective diffusion for the alloying process under some conditions than the usual atomic diffusion. Computer calculations and experimental investigations are presented that show this. The systems investigated were Cu(sol)-Al(liq) and Ni(sol)-Al(liq) compositions. The relation between the conductivity of a solid inclusion and the surrounding liquid metal are opposite in the two systems.
Knowledge of the phenomena analysed might be useful for the improvement of alloying technologies.

NOMENCLATURE

B	magnetic induction
C	const
c	concentration
D	diffusion coefficient
$E\equiv E(\alpha,\beta)$	elliptic function of 1st kind
$F\equiv F(\alpha,\beta)$	elliptic function of 2nd kind
i	unit vector
f	Lorentz-force
H	magnetic intensity
J	electric current density
K	parameter
k	coefficient of cell
p	pressure
Q	activation energy
R	const
r	radial coordinate
Sh	Sherwood number (criterion)
T	temperature
U	velocity of fluid far of inclusion
v	velocity
V	reduced velocity
y	part of radial distance
α	parameter
$\Gamma(x)$	gamma function
ζ	parameter
η	dynamic viscosity
ϑ	angular coordinate
Λ	parameter
λ	conductivity
μ	magnetic permeability
φ	parameter
ψ	Stokes' stream function

Subscripts

a	approximate
el	electrical case
e	surrounding medium
i	inclusion
Me	metal
r	radial value
s	outer surface of layer e
ϑ	tangential value
φ	azimuth
∞	infinity
0	zero value
1…4	ordinal number

INTRODUCTION

A series of solutions to the problem of interaction between a foreign solid inclusion and a surrounding current-carrying liquid substance under the passage of a direct current is known: in case of a non-conductive inclusion (Chow, 1966) and in case of a conductive inclusion (Raychenko O.I., Raychenko O.O., and Chernikova, 1993; Raychenko O.I., Raychenko O.O., Chernikova, and Miroshnichenko, 1993). Inclusions in suspension have, as a rule, conductivity and some other properties different from the external medium, and also often have a higher melting point. In manufacturing or operations with alloys of some compositions, there is the need to understand the character of interactions between such foreign inclusions and the surrounding current-carrying medium. Processes such as mass-transfer can arise due to passage of the electric current in this situation.

MODEL DESCRIPTION

We use the following conventional scheme (Fig. 1). There is a solid sphere i (with radius r_i and electric conductivity λ_i), a spherical layer e (outer radius $r_e=kr_i$, where k is the coefficient of the cell) filled by a liquid conductor (with the electrical conductivity λ_e and viscosity η). The origin of the spherical coordinate system (r, ϑ) is located in the center of the sphere. The direct current is passing through the whole model in direction ϑ=0 (direction OA, see Fig. 1.) with current density far away the inclusion that is equal to J_0. The present model has axial symmetry. In general, the mentioned scheme may be applied to heterogeneous compositions with different components where the whole volume is exposed at a temperature between melting points of the two component substances: the low-melting ingredient therefore exists in the liquid state. We divide in mind a whole suspension up into a number of cells with one inclusion in center of each cell. Then let us change a cell-polyhedron to a cell-sphere with the same volume.

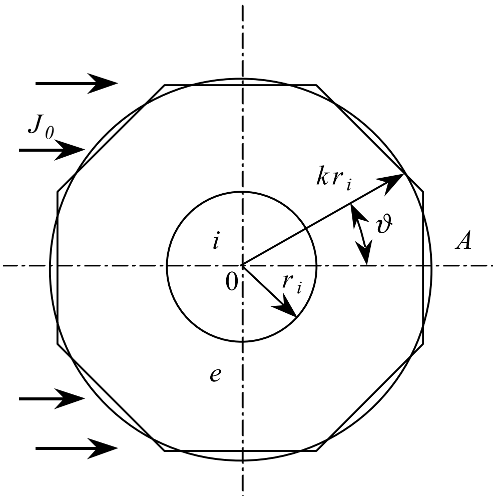


Figure 1 : Scheme of the model.

In the present case two pairs with the following component compositions: $Cu_{sol.}$-$Al_{liq.}$ and $Ni_{sol.}$-$Al_{liq.}$ were taken for theoretical comparative analysis and practical test. A number of elements such as that depicted in Fig. 1 composes the suspension. All physical characteristics of the mentioned components are the same ones for conditions that are valid for experiments, they are taken from (Sosedov, 1975; Kikoin, 1976; Samsonov, 1976; Drits et al., 1985; Larikov et al., 1987) and placed in Table 1.

Parameter	Components		
	Cu	Al	Ni
λ, Ohm^{-1} m^{-1}	23.4×10^6	10.0×10^6	2.3×10^6
η, MPa s	-	1.076	-
	System		
	Cu-Al		Ni-Al
c_s	0.31		0.0325
D_0, m^2 s^{-1}	0.15×10^{-4}		1.1×10^{-6}
Q,, kJ mol^{-1}	126.4		83.32

Table 1: Properties of the systems investigated.

THEORETICAL ANALYSIS

Convection movement at electric current passage

Suppose that the basic Navier-Stokes equation is one of electro-magneto-hydrodynamics in inertialess and non-induction approximation (Chow, 1966)

$$grad\, p + \eta\, curl\, curl\ \mathbf{v} = \mathbf{f}\,, \tag{1}$$

where **f**=**J**×**B** is the spatial Lorentz-force, p is the pressure, **J** is the current density, **B** is the magnetic induction, **v** is the velocity of liquid. The velocity components of the Lorentz electroconvection (the spatial Lorentz-force **f** is placed in the right side of eq. (1)) are

$$\left.\begin{aligned} v_r &= \frac{1}{r^2 \sin\vartheta}\frac{\partial\psi}{\partial\vartheta}\,, \\ v_\vartheta &= -\frac{1}{r\sin\vartheta}\frac{\partial\psi}{\partial r}\,. \end{aligned}\right\} \tag{2}$$

where ψ is Stokes' stream function. To determine the velocity components we will apply the following boundary conditions:

$$\left.\begin{aligned} & v_r\big|_{r=r_i} = 0, \quad v_r\big|_{r=kr_i} = 0, \\ & v_\vartheta\big|_{r=r_i} = 0, \quad v_\vartheta\big|_{r=kr_i} = v_{\vartheta,\infty}\big|_{r=kr_i}, \end{aligned}\right\} \tag{3}$$

where $v_{\vartheta,\infty}$ is the velocity ϑ-component in the model solid particle – infinite liquid medium (Raychenko O.I., Raychenko O.O., and Chernikova, 1993; Raychenko, Popov, Burenkov., Istomina, and Derev'yanko, 1997), i.e. the present scheme is the "soft cell" unlike the previous case studied (Raychenko O.I., Raychenko O.O., and Chernikova, 1993; Chow, 1966). First, by analogy with (Tikhonov and Samarsky, 1953) we can write expression for the current density in medium e:

$$\begin{aligned} \mathbf{J} = J_0 \Bigg\{ & \left[1 - \left(\frac{r_i'}{r}\right)^3\right]\cos\vartheta\,\mathbf{i}_r - \\ & - \left[1 + \frac{1}{2}\left(\frac{r_i'}{r}\right)^3\right]\sin\vartheta\,\mathbf{i}_\vartheta \Bigg\}, \end{aligned} \tag{4}$$

where $\mathbf{i}_r$, $\mathbf{i}_\vartheta$ are the unit vectors, $r_i' = r_i \cdot \Lambda$,

$$\Lambda = \left(2\frac{\lambda_e - \lambda_i}{2\lambda_e + \lambda_i}\right)^{\frac{1}{3}}.$$

Furthermore, it is possible to obtain the magnetic intensity, arising from the electric current (Raychenko O.I., Raychenko O.O., Chernikova, and Miroshnichenko, 1993):

$$\mathbf{H} = \frac{\mu_0 J_0 r}{2}\left[1-\left(\frac{r_i^{'}}{r}\right)^3\right]\sin\vartheta\,\mathbf{i}_\varphi, \tag{5}$$

where μ_0 is the magnetic constant, $\mathbf{i}_\varphi$ is the azimuth unit vector. Introducing eqs. (4) and (5) into eq. (1) after analogous calculation as in (Chow, 1966), we obtain Stokes' stream function

$$\psi_{el} = -\frac{\mu J_0^2 r_i^{'5}}{16\eta}\left[\frac{r_i^{'}}{r}+\left(\frac{r}{r_i^{'}}\right)^2+C_1\left(\frac{r_i^{'}}{r}\right)^2+\right.$$
$$\left.+C_2+C_3\left(\frac{r}{r_i^{'}}\right)^3+C_4\left(\frac{r}{r_i^{'}}\right)^5\right]\sin^2\vartheta\cos\vartheta, \tag{6}$$

where $\mu=\mu_0\mu_{Me}$, μ_0 is the magnetic constant, μ_{Me} is the relative magnetic permeability of the liquid metal (usually $\mu_{Me}\approx 1$), C_1, C_2, C_3, and C_4 are the constants, η is the dynamic viscosity. Substitution of eq. (6) into eq. (2) gives the velocity components:

$$v_r = -\frac{\mu J_0^2 r_i^{'3}}{16\eta}\left[\left(\frac{r_i^{'}}{r}\right)^3+1+C_1\left(\frac{r_i^{'}}{r}\right)^4+\right.$$
$$\left.+C_2\left(\frac{r_i^{'}}{r}\right)^2+C_3\frac{r}{r_i^{'}}+C_4\left(\frac{r}{r_i^{'}}\right)^3\right]\times \tag{7}$$
$$\times\left(3\cos^2\vartheta-1\right),$$

$$v_\vartheta = \frac{\mu J_0^2 r_i^{'3}}{16\eta}\left[-\left(\frac{r_i^{'}}{r}\right)^3+2-2C_1\left(\frac{r_i^{'}}{r}\right)^4+\right.$$
$$\left.+3C_3\frac{r}{r_i^{'}}+5C_4\left(\frac{r}{r_i^{'}}\right)^3\right]\sin\vartheta\cos\vartheta. \tag{8}$$

The constants C_1, C_2, C_3, and C_4 can be determined from eq. (3) which are the boundary conditions (Raychenko et al., 1997). The velocity magnitude is determined from the usual expression: $v=\sqrt{v_r^2+v_\vartheta^2}$. The computation based on formulae (7), (8) gives a reconstructed picture of the velocity field around the inclusion *i* which is caused by Lorentz electroconvection (Fig. 2). Figure 2 shows schematically the field of the dimensionless normalized (reduced) velocities of such a moving liquid in the first quadrant of the cell $V = v\frac{16\eta}{\mu J_0^2 r_i^{'3}}$ (for example, in the system $Cu_{sol.}$-$Al_{liq.}$) at $r_i \sim 35\times10^{-5}$ m, $J_0=4.0\times10^6$ A m^{-2}, temperature 1000 K, and k=2. The velocity component v_r near the line ϑ=0 has a negative value. Figure 2 is a conventional vector arrow plot.

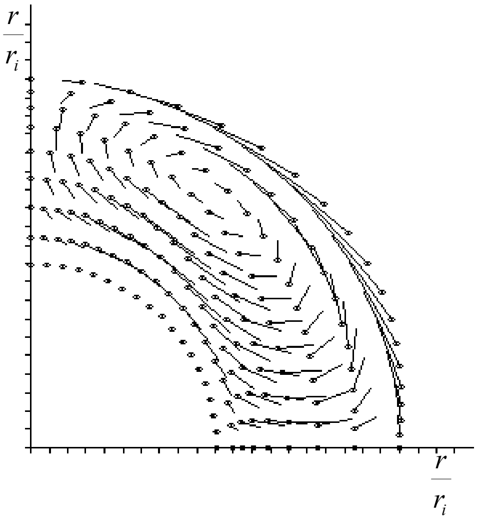


Figure 2: The velocity field computed using eqs. (7), (8).

Diffusion from solid inclusion into liquid current-carrying medium under electric current action

If we suppose $y=r-r_i \ll r_i$ then it is possible to write Stokes' stream function approximately as

$$\psi_a \cong -\frac{\mu J_0^2 R^3 \Lambda^3}{16\eta}\varphi(\Lambda,k)y^2\sin^2\vartheta\cos\vartheta, \tag{9}$$

where

$$\varphi(\Lambda,k) = 1+\Lambda^3+\frac{1}{k^2K}[\Lambda^3(-6k^{12}+$$
$$+75k^9-75k^8-78k^7+105k^6-15k^5+ \tag{10}$$
$$+9k^2-30k+15)+12k^{12}\text{-}75k^{11}\text{+}180k^9\text{-}$$
$$-159k^7\text{+}30k^5-30k^4+72k^2\text{-}30],$$

$$K = 4k^{10}-25k^7+42k^5-25k^3+4 .$$

Let us apply Levich's method (Levich, 1959) (see for comparison Raychenko O.I., Raychenko O.O., Chernikova, and Miroshnichenko, 1993 also).
We use the equation of the convection diffusion as a starting equation the following one (Levich 1959)

$$\frac{\partial c}{\partial\vartheta} \cong Dr_i^2\sin^2\vartheta\sqrt{3U}\frac{\partial}{\partial\psi}\left(\sqrt{\psi}\frac{\partial c}{\partial\psi}\right), \tag{11}$$

where c is the concentration of diffusant (substance diffusing from the surface inclusion *i*), D is the diffusion coefficient that is $D = D_0\exp\left(-Q/RT\right)$, Q is the activation energy, R is the gas constant, T is the temperature, U is the velocity of a fluid far off an inclusion, $\psi=-\psi_a$. In eq. (11) it is necessary to replace the factor (3/4)U by the expression

$$\frac{\mu J_0^2 \Lambda^3 r_i^3}{16\eta}\varphi(\Lambda,k)\cos\vartheta .$$

Thus, we obtain the equation for the stationary Lorentz diffusion from the surface solid inclusion into the current-carrying liquid medium

$$\frac{\partial \tilde{n}}{\partial \vartheta} = \Phi(J_0,\eta,\Lambda,k,r_i)\times \times \sin^2\vartheta\sqrt{\cos\vartheta}\;\frac{\partial}{\partial\psi}\left(\sqrt{\psi}\;\frac{\partial c}{\partial \psi}\right), \tag{12}$$

where

$$\Phi(J_0,\eta,\Lambda,k,r_i) = \frac{D r_i^3 J_0}{2}\sqrt{\frac{\mu r_i}{\eta}\Lambda^3\left|\varphi(\Lambda,k)\right|} .$$

Let us determine the behaviour of the diffusant in the layer $y<0.1r_i$ near the surface of inclusion using data from (Kikoin, 1976). Derivative $\partial c/\partial y$ gives the r-component of the concentration gradient near the surface of inclusion:

$$\left.\frac{\partial c}{\partial y}\right|_{y=0} = \frac{3(c_s-c_0)}{\tilde{A}(1/3)}e^{-\frac{4}{9}\zeta^3}\left.\frac{\partial \zeta}{\partial y}\right|_{y=0}, \tag{13}$$

where $\tilde{n}_S = \tilde{n}|_{r=r_i}$, $c_0 \equiv 0$ is the concentration in the depth of medium e, $\Gamma(x)$ is the gamma function,

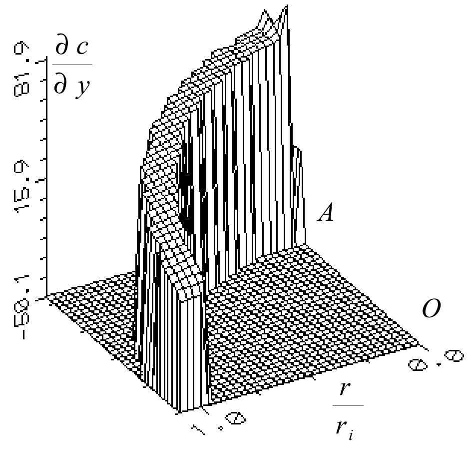
$$\frac{\partial \zeta}{\partial y} = \left\{\frac{5\mu J_0^2 r_i^4 \Lambda^3\left|\varphi(\Lambda,k)\right|}{2^6\eta D}\times \times\frac{\sin^3\vartheta\cos^{\frac{3}{2}}\vartheta}{[\sqrt{2}(2E-F)-\sin\vartheta\cos^{\frac{3}{2}}\vartheta]}\right\}^{\frac{1}{3}} \tag{14}$$

$E \equiv E(\alpha,\beta)$, $F \equiv F(\alpha,\beta)$ are the elliptic functions of 1st and 2nd kinds respectively, $\alpha = \arcsin(\sqrt{2}\sin(\vartheta/2))$.

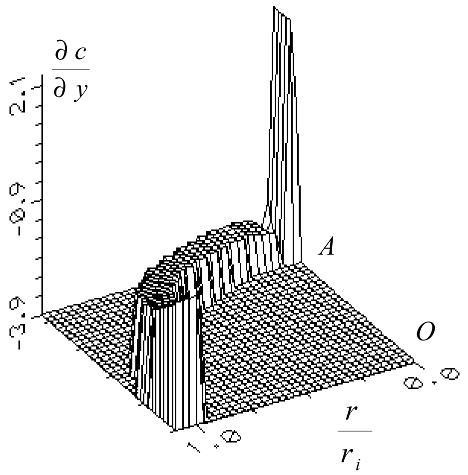
The concentration gradient is computed on the basis of eq. (13) for the layer $y<0.1r_i$ and is shown in figure 3. Parameters Λ and $|\lambda_e\text{-}\lambda_i|$ (see Table 2) influence the calculated concentration gradient (Fig. 3).

Value		System	
		Cu-Al	Ni-Al
Λ		-0.85	0.88
$\lvert\lambda_e\text{-}\lambda_i\rvert$, Ohm^{-1} m^{-1}		13.4×10^6	7.7×10^6
$\partial c/\partial y$	max.	83.83	-50.08
	min.	2.36	-3.94

Table 2: Values used for the calculations.



a)



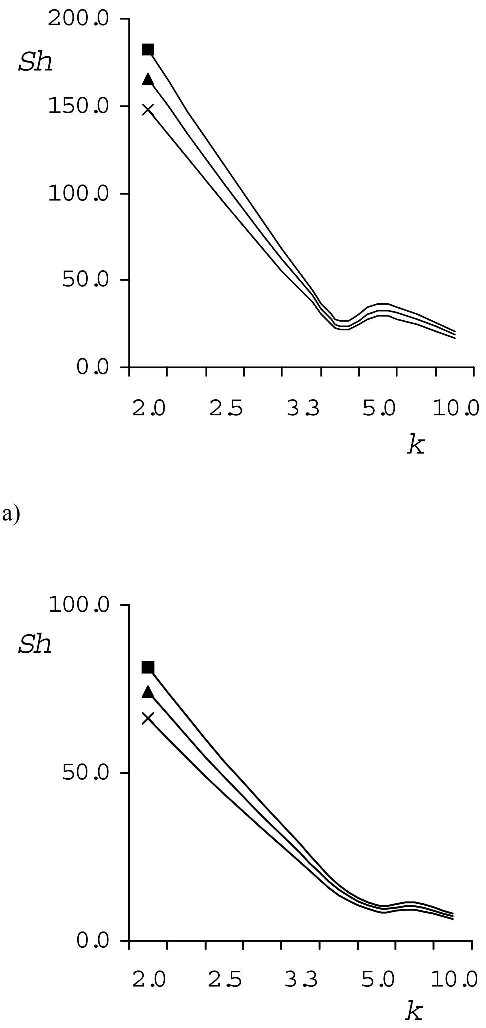
b)

Figure 3: Reconstruction of the concentration gradient in 3D plot for layer y on the surface of inclusion; a) the case of Cu-Al, b) the case of Ni-Al.

Dividing the full diffusion flux under electric current action by the full diffusion flux from the surface of the spherical inclusion in case of the stationary atomic diffusion by analogy with (Raychenko O.I., Raychenko O.O., Chernikova, and Miroshnichenko, 1994; Raychenko, Popov, Burenkov, Istomina, and Derev'yanko, 1997) we obtain the expression for the Sherwood number:

$$Sh = 0.677\left[\frac{r_i^4 J_0^2\,\mu\left|\Lambda^3\,\varphi(\Lambda,k)\right|}{\eta D}\right]^{\frac{1}{3}}. \tag{15}$$

Figure 4 shows the dependence of the Sherwood number upon the radius of inclusion. Plots showing the dependences of values Sh upon parameter k (Fig. 4) present the possibility to determine best conditions for alloying. There is a possibility to increase the rate of alloying without increase of the current density through suspension, but by variation of parameter k.



a)

b)

Figure 4: The Sherwood number as function of *k*; ■- 4.36×10^6 A m^{-2}, ▲- 3.78×10^6 A m^{-2}, ✕- 3.20×10^6 A m^{-2}; a) the case of Cu-Al, b) the case of Ni-Al.

For comparison with theoretical analysis, experiments with some systems were performed. The average concentration Cu in $Al_{(liq)}$ was found to be ~37 wt. % (the theoretical computation gives ~12 wt. %); while the average concentration Ni in $Al_{(liq)}$ was found to be ~54 wt. % (the theoretical computation gives ~24 wt. %). These results are for the following conditions: r_i~35×10^{-5} m, J_0=4.0×10^6 A m^{-2}, temperature 1000 K, and k=2. The higher experimental values could be caused by the usual diffusion process that will occur simultaneously with the diffusion caused by electric current action. The latter is the only source of diffusion that is taken into account only in this work.

CONCLUSION

An algorithm for the calculation of the reduced diffusion flux (its measure is the Sherwood number) during the passage of the electric current in the liquid metal surrounding a solid sphere has been developed (Fig. 5). This approach has been applied to models of two suspensions. The dependence of the Sherwood number upon the coefficient of the cell (this value is a measure of inclusion separation) is a decreasing function with a secondary maximum. The proposed simulation can be a basis for calculations of the mass transfer in electro-technological processes.

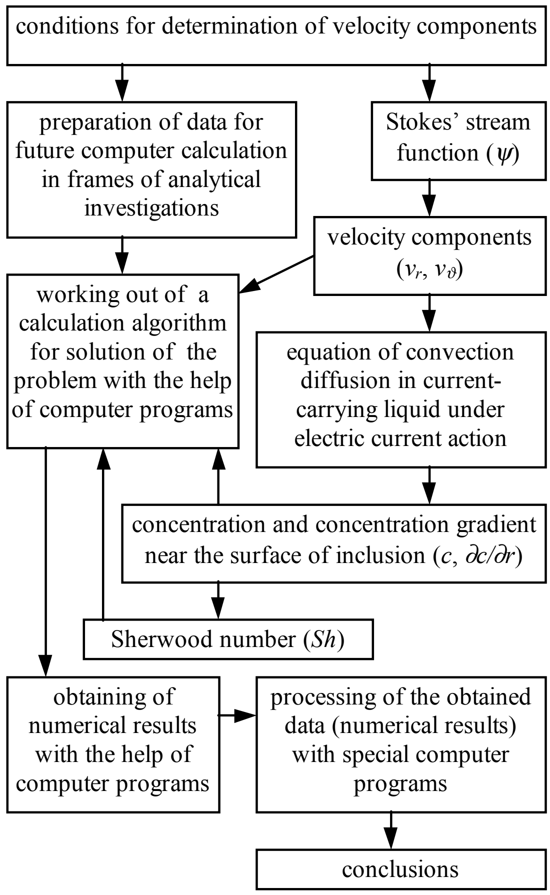


Figure 5: The algorithm of computation (scheme).

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