On Thermocapillary Rings from Radioactive Particles Suspended in Liquids

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Thermocapillary motion and its significance with regard the thermal behavior of heat sources and with particular reference to radioactive particles suspended in fluids is discussed. It shown that because the heat source, a thermocapillary flow is induced with the viscous liquid flowing outwards from the particle and then with a drag force propelling the surrounding suspended material away with the formation of a diffusive ring. Utilizing a simplified physical model, it was found a power-law for the concentration of particles surrounding the hot particle. The formation of such a ring may be important as a visual simple way to detect or unveil the presence of radioactive particles suspended in lakes, rivers, ponds or aquifers after,say, an accidental atmospheric release, a new method for detection under controlled conditions or in geophysical science. Computational Fluid Dynamics (CFD) simulations were performed which agree qualitatively and quantitatively well with the analytical model.

Keywords. *Thermocapillary, Radioactive heat decay, Diffusion*

I. INTRODUCTION

While Marangoni convection have been researched through decades in fundamental physics and industrial applications as well [1]-[9], and more recently extensively simulated using the lattice Boltzmann methods,[10]-[13]. Nonetheless, there still remains interesting unexplored phenomena. One of them is the thermal behavior of heat sources suspended in liquids. The object of this work was to analyze the role of Marangoni convection induced by heat sources and particulary by radiogenic particles when they are suspended in fluids. It is shown that because the thermal gradient surrounding the radioactive particle, thermocapillary convection is developed with the viscous liquid flowing outwards from the radiogenic particle and then with a drag force propelling the surrounding suspended material away with the formation of a diffusive ring-like structure. The importance of such hypothetical ring-like structure lies in the possibility to unveil particles suspected to be radioactive suspended in,say, lakes, ponds or similar aquifers environments after, say, an accidental atmospheric release or as a technique of detection in laboratories under controlled conditions. In geological science, the presence of rigs-like structures impressed in rocks could be an indicative of radiogenic activity when the rock was molten.

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II. METHODS

The Marangoni effect or also called the Gibbs-Marangoni effect,[14], is the mass transfer along an interface between two fluids due to surface tension gradient. In the case of temperature dependence, this phenomenon is generally named as thermocapillary convection. In short, because a fluid with a high surface tension pulls more strongly on the surrounding fluid than one with a low surface tension, therefore, the presence of a gradient in surface tension will cause the liquid to flow away from regions of low surface tension to regions of high surface tension. For the case of thermocapillary, the surface tension gradient is induced by the temperature gradient (surface tension is a function of temperature and decrease when temperature increase). For the interested reader above Marangoni convection is referred to the classical work of Landau and Lifshitz,[15].

• Thermocapillary flow induced by temperature gradient from a suspended radiogenic particle

Because the thermal gradient surrounding the radiogenic particle suspended in a fluid -which can be considered of infinite extent in comparison with the dimensions of the particle, a thermocapillary flow is induced where the viscous liquid flows outwards from the radiogenic particle and then with a drag force propelling the surrounding suspended material away from the particle. This flow, being neither,axisymmetric nor irrotational, is difficult to solve analytically, even if the inertial terms are omitted from the Navier-Stokes equation. Hopefully, we may extend the early work by Fedosov,[16] which considered the induced thermocapillary motion of a liquid in a flat container of infinite extent induced by a temperature gradient. This expression altough, of course, is not rigurously valid for our case of study, however, in view of the several uncertainties, this certainly can provide a first assesement of the ring formation surrounding the radiogenic particle.

According with Fedosov,[16] the velocity of the liquid at the interface is given by [16]

$$v = -\frac{h}{4\mu}\frac{d\sigma}{dT}\frac{d\sigma}{dx} \tag{1}$$

where h is the is a depth of the container, μ the dynamic viscosity of the liquid, σ the surface tension, T temperature, and x is the coo-ordinate applied along the container length.
Therefore, we from dimensional analysis, a similar semiempirical relationship may be expected for our case of study to have the form

$$v = -\frac{Fh}{4\mu}\frac{d\sigma}{dT}\frac{d\sigma}{dx} \tag{2}$$

where $F \approx 1$ is a numerical constant which consider the deviation from the real situation.

Now, for our specific case of study working with a radiogenic particle a heat source, some further analysis is possible. First, the thermal gradient surrounding the radiogenic particle may be derived from the Fourier equation

$$-\kappa\frac{dT}{dr} = q(r)A_r \tag{3}$$

where $q(r)$ is the heat flux at which heat is conducted at a radial distance r from the radiogenic particle through a plane of area $A_r = 2\pi r dz$ normal to this direction with z the vertical coordinate, and with a temperature gradient $\frac{dT}{dr}$ and being κ the effective thermal conductivity of the surrounding liquid, where it was considered a cylindrical particle which is a acceptable simplification for prelimnary assesment when particles are of very small size as is our case.

On the other hand, we know by a simple balance of energy that the hat flux at a certain distance r from the radiogenic particle must be related with the heat flux at its surface by $q_o 2\pi r_o = q(r)2\pi r$ being r_o the radius of the particles and q_o the heat flux at its surface, respectively. Thus, Eq.(3) becomes

$$-\kappa\frac{dT}{dr} = \frac{q_o r_o}{r} \tag{4}$$

Substituting Eq.(4) into Eq.(2) one obtains

$$v_r(r) = -\frac{Fq_o r_o h}{4\mu\kappa r}\frac{d\sigma}{dT} \tag{5}$$

Finally, the heat flux at the surface of the particle may be related with its radiogenic activity by the following expression

$$q_o \simeq \frac{\epsilon\rho_p\eta N_A\lambda r_o}{2\bar{m}_p} \tag{6}$$

where ϵ is the energy per disintegration, ρ the density of particle, η its mineral content (fraction of radiogenic material), N_A the Avogadro number, λ the disintegration constant, $\bar{m}_p$ the molecular weight and r_o the radius of the particle. Substituting Eq.(6) into Eq.(5) one obtains

$$v_r(r) = -\frac{\epsilon\rho_p\eta N_A\lambda r_o^2 h_1}{8\bar{m}_p\mu\kappa_e r}\frac{d\sigma}{dT} \tag{7}$$

- **Ring formation**

With the calculated velocity profile we can derive an expression for the formation of the diffusive ring as follows.

First, let us assume that the suspended radiogenic particle is surrounded by a cloud of particles homogeneously dispersed in the liquid . Let us also say that the container has a radius R, for example the radius of the test tube used in the laboratory. Because the thermocapillary flow, particles are pushed away from the radiogenic particle and as result a radial distribution of particles will appear as pictorially depicted in Fig. 1.

The flux of particles J across any plane parallel to the z-axis is given by

$$J = cv_r \tag{8}$$

where c is the total concentration of particles at a given radial distance r, and v_r is the radial velocity which was previously calculated. On the other hand, the opposite flux due to particle concentration gradient is given by

$$J_p = -D_p\frac{dc}{dr} \tag{9}$$

where D_p is the local particle diffusivity. At steady state this flux must be equated to the flux across any plane parallel to the z-axis , and then with $J_p = -J$, from Eq.(8) and Eq.(9) we obtain

$$\frac{dc}{c} = \frac{v_r}{D_p}dr \tag{10}$$

inserting Eq.(7) into Eq.(10) and if we define the concentration of particles at the wall container as c_o, then after integration one obtains the following power-law for the concentration

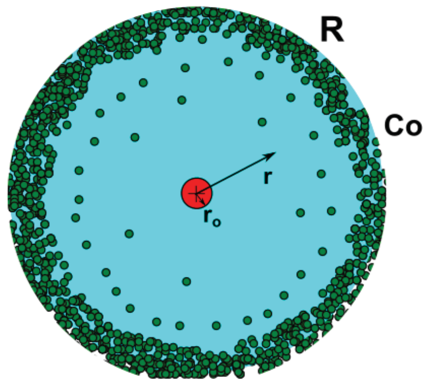

FIG. 1: Ring-like deposit formation surrounding a radiogenic particle induced by thermocapillary motion.

$$\frac{c}{c_o} = \left[\frac{r}{R}\right]^{-n} \tag{11}$$

where an exponential index n was defined as

$$n = \frac{\epsilon \rho_p \eta N_A \lambda r_o^2 F h}{8 D_p \bar{m}_p \mu \kappa_e} \frac{d\sigma}{dT} \tag{12}$$

because the very low velocity regime, it is permissible to use the Einstein-Stokes diffusivity equation for the diffusivity term which is valid for low Reynolds numbers given by

$$D_p = \frac{\kappa_B T}{6\pi\mu a} \tag{13}$$

where κ_B is the Boltzmann constant, T the temperature, and a the radius of the particle suspended surrounding the radiogenic particle. Inserting Eq.(13) into Eq.(12) yields

$$n = \frac{3\pi a \epsilon \rho_p \eta N_A \lambda r_o^2 F h}{4 \kappa_B T \bar{m}_p \kappa_e} \frac{d\sigma}{dT} \tag{14}$$

• **Discussion**

To obtain an idea of the shape of the curves predicted by Eq.(11) using the derived exponential index from Eq.(14), we assume some typical values of the parameters for a somehow arbitrary radiogenic particle with: $\epsilon = 20$ keV; $\rho_p = 2000$ kg/m^3; $\eta = 0.1$; $N_A = 6.02 \times 10^{23}$ mol^{-1}; $\lambda = 0.13$ years^{-1}; $\bar{m}_p = 60$ g/mol; and for the properties of water: $\kappa_e = 0.6$ W/(m)(K); and $\frac{d\sigma}{dT} = -1.66 \times 10^{-4}$N/(m)(K); and $\kappa_B = 1.38 \times 10^{-23}$m^{2}kg/(s^2)(K), and with a liquid temperature $T = 300$ K and assuming $F = 1$. The resulting curves are shown in Fig. 2. It is seen that the distribution curve is very strongly dependent on the radius of the particle.

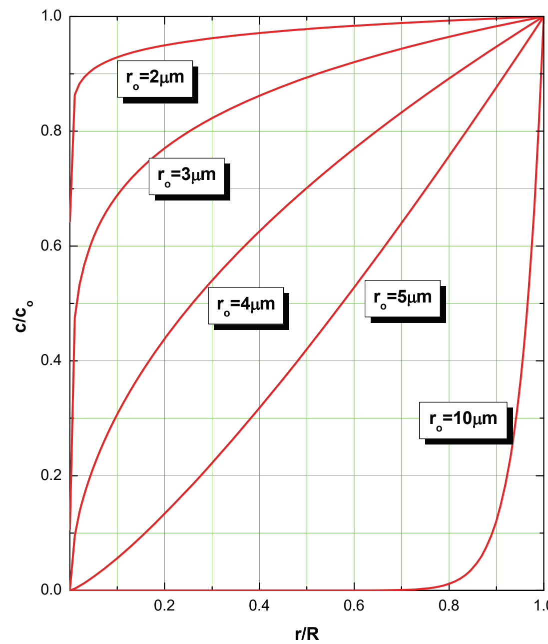

FIG. 2: Radial concentration distribution calculated from Eq.(11) and using the exponential index given by Eq.(**??**) for a radiogenic particle with a decay energy of 20 keV for various radius of the particle and considering the same radius for the surrounding particles.

The effect of ionizing radiation on the Marangoni convection

Because the radioactive decay, ionization of the liquid occurs and then a local electric field developed. If one considers that the surface tension is the result of a distribution of charges in the fluid, it is permissible to think that a local electrical field can affect the surface tension generating a gradient and then inducing Marangoni effect. At this point and although there is some dispute over the effects of electric and magnetic fields on surface tension. However, electric field have been reported to lower the surface tension of natural water by up to 8% [17], if so, the ionization of the liquid will also contribute to the formation of the diffusive ring because the surface tension gradient generated by the electrical field will be in the same direction than the gradient generated by the temperature. Nevertheless it must be keep in mind that the reach of an electrical field in water is very reduced and is given by the Debye length, which for an electrolyte or a colloidal suspension, is just a few nanometers.

III. COMPUTATIONAL SIMULATION

In order to gain certain confidence in the predictions from the derived expressions, a simplified 2-dimensional pressure-based CFD model was developed using the commercially available CFD software Fluent which allows to specify Marangoni stress boundary conditions by specifying the gradient of the surface tension with respect to temperature. An steady condition was used in the calculations and a pressure-velocity was coupled using the SIMPLEC algorithm with a first order upwind discretization scheme for momentum and energy. In order to ensure the independency of the calculations with the grid, the convergency criteria was taken to be residual RMS error values of 10^{-4}. The mesh resolution independence was checked running an initial mesh and ensuring the convergency criteria of RMS of 10^{-4} with an imbalance in the domain less than 1%, then, a second simulation was performed using a second mesh with finer cells throughout the domain, then the simulation was run until the convergency criteria and imbalance in the domain were satisfied. The criteria for selection of the mesh was that the velocity values for two consecutive stimulations were less than a 1%, then the mesh at the previous step was considered accurate enough to capture the result. The schematics of problem and the boundary conditions used are shown in Fig. 3.

The simulation consisted in a box with 10 μm length and 5 μm width with a semi-circular corner with radius 1 μm as schematically depicted in Fig. 3. The boundary conditions were adiabatic walls at the bottom and the top, two pressure outlets at the left and right sides, and the semi-circular corner as a wall with an associated heat flux which represent the heat source. In the top wall, a specific Marangoni stress was associated and then representing the free-surface. The fluid used was water with a temperature of 300 K. The heat flux associated with the semi-circular wall was calculated from Eq.(6) and with the values of the parameters as used in our previous example, which result in a heat flux of $q_o = 1.32 \times 10^{-2}\text{W/m}^2$. The resulting curves are shown in Fig. 4 and Fig. 5. In Fig. 4 it is shown the velocity vectors profile where it is seen that the approximation of zero vertical velocity is a good valid approximation, also the maximum velocity for this hypothetical case at the surface was found around $\approx 8.69 \times 10^{-10}$ m/s, if calculated with Eq.(7) give us $\approx 6.5 \times 10^{-10}$ m/s which is a good enough result. Fig. 5, is the velocity profile calculated at 5 μm from the particle, i.e., in the middle of the box.

IV. SUMMARY OF RESULTS AND CONCLUSIONS

Thermocapillary motion and its significance with regard the thermal behavior of heat sources and with particular reference to radioactive particles suspended in fluids was discussed. Utilizing a simplified physical model it was investigated the formation of diffusive rings surrounding the radiogenic particle and it was found a power-law for the distribution of particles surrounding the radiogenic particle with an strong dependence in the size of the particle. The formation of such a ring may be important as a visual simple way to detect or unveil the presence of radioactive particles suspended in lakes, rivers, ponds or aquifers after an accidental atmospheric release., or as a new method for detection under controlled conditions. In geological science, the presence of rigs-like structures in rocks could be an indicative of radiogenic activity when the rock was molten.

NOMENCLATURE

A_r = Area normal to the radial direction
D_p = diffusivity
$F(z)$ = component radial velocity dependent of depth
g = gravity
h = depth of the container
$\bar{m}_p$ = molecular weight of radiogenic particle
n = dimensionless exponent, Eq.(**??**)
N_A = Avogadro number
q_o = heat flux at the surface of particle
q = heat flux at a distance r
r = radial coordinated directed from the particle outwards
r_o = radius of radiogenic particle
R = radius of the container
T = temperature
v = velocity
z =the z-axis coordinated directed from the surface to the solid boundary

Greek symbols

η = radiogenic mineral content of particle
ϵ = energy per disintegration
λ = disintegration constant
μ = dynamic viscosity
ρ = density of liquid
ρ_p = density of radiogenic particle
σ = surface tension
κ = thermal conductivity of liquid

subscripts symbols

p = particle
r = radial
z = vertical coordinate

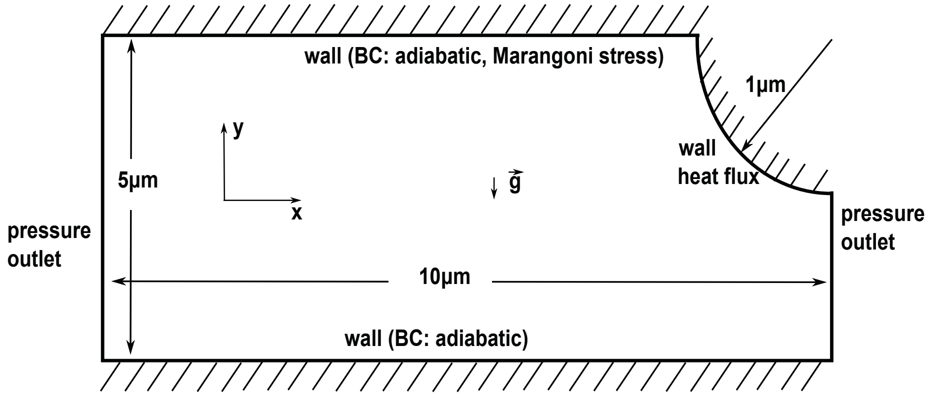

FIG. 3: Problem schematics for the CFD simulation.

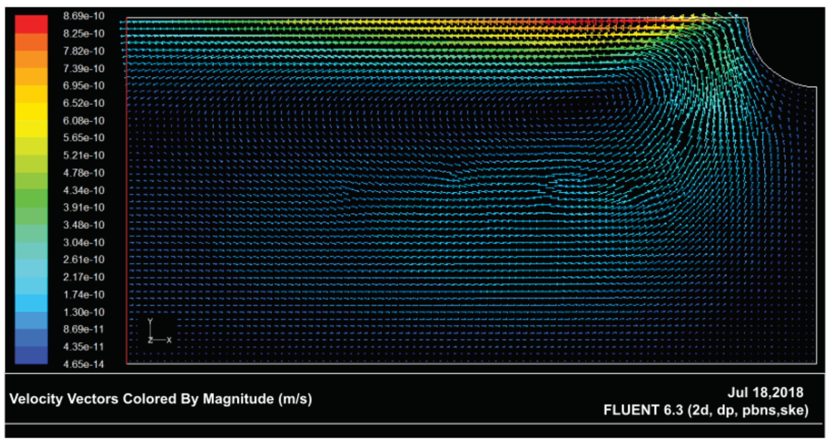

FIG. 4: Velocity vectors profile from the CFD simulation.

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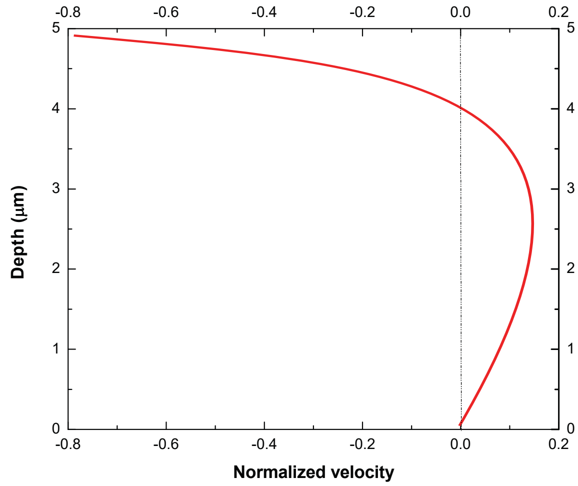

FIG. 5: velocity profile at $x = 5\mu$m from the particle

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