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To cite this article: J Wang et al 2016 IOP Conf. Ser.: Mater. Sci. Eng. 117 012051

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Convection and solidification influenced by thermo-electric effect

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Abstract. We analyse the thermo-electric-magnetic (TEM) forces both on the liquid metal and immersed solid particles which mimics equiaxed grains. Firstly, we provide a summary of analytical works performed in a previous paper. Then, we present experimental works on in situ and real time visualization of the directional solidification of Al-10wt%Cu alloy by means of synchrotron X-ray radiography during directional solidification under a transverse 0.08 T permanent magnetic field. The experiments bring definite evidence of the existence of various phenomena which could be related to the thermoelectric effect acting directly on equiaxed grains. Comparisons between the predicted and observed horizontal deviations of the particles are in fairly good agreement.

1. Introduction

Convection induced by magnetic field is now widely used in metallurgy to master liquid metal flows during solidification and thus the final properties of the materials. Usually, AC magnetic fields are commonly used to generate fluid flows whereas DC fields are supposed to create an anisotropic damping of liquid motions [1-3]. However, the presence of temperature gradient may lead to the generation of thermo-electric currents both in the liquid and in the solid phases near the solidification interface. The interaction of non-parallel thermo-electric current and the applied magnetic field creates electromagnetic forces which act both on the liquid and on the solid [4]. In previous works, experimental and numerical analyses of thermo-electric magnetic convection (TEMC) for different length scales under various magnetic fields up to 10T have been investigated, and it has been found that besides TEMC in the melt thermoelectric magnetic (TEM) force on the solid was important as well [5-8]. In general, all materials during solidification have thermal gradient at the interface and different material properties between the liquid and the solid, including thermal and electric conductivities as well as absolute thermo-electric powers. The existence of temperature gradient having a non-zero component along the interface (e.g. the liquid-solid front) may cause the appearance of internal electric current along the interface in accordance with the well-known Seebeck effect, associated with the Thomson effect. Thermo-electric effect may be encountered in various situations as in solidification processes of electrically conducting materials. When static magnetic fields are superimposed there appears Lorentz forces (TEM forces) which may be responsible for

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thermo-electric hydromagnetic flows, the so-called TEMHD [4]. Various types of effects may be expected according to the magnetic field amplitude and direction as well as the physical properties of both the liquid and solid. The existence of electrical currents depends on the thermo-electric power difference $S_l - S_s$ [4, 8] between the liquid and solid. Furthermore, the TEM forces are able to drive a liquid motion only if $\nabla S_l \times \nabla T \neq 0$ [4].

In the present paper we focus our attention on the TEM forces and motions on equiaxed grains in a solidifying Al-10wt.%Cu alloy. Firstly, we summarize the theoretical results obtained in a previous works [9]. Then, we present experimental results related to in situ and real time observations of the equiaxed grain motion during directional solidification of Al - 10 wt.% Cu under static magnetic field which were carried out by means of synchrotron X-ray radiography. The latter work was published in [10].

2. Theory of the motion of a spherical particle under thermo-electric effect

We consider now an isolated solid spherical particle immersed in a liquid metal submitted both to a constant vertical temperature gradient and a static magnetic field. The geometry is illustrated in figure 1. Details of the theoretical analysis may be found in [9]. We only provide here a summary of the results. The thermo-electric current has a dipole-type distribution [9]. The interaction of the TEM electric current density \vec{j}_i and a superimposed static magnetic field \vec{B}_0 produces a TEM body force \vec{F}_i . In the case of a transverse magnetic field, the orientation of the TEM force acting on the sphere is illustrated in figure 1. The force is perpendicular both to the temperature gradient and the magnetic field. The analytical expression of the body force is $\vec{F}_i = \vec{j}_i \times \vec{B}_0$, i = l or *s* respectively in liquid and in solid. In spherical coordinates (r, θ, ϕ) the expressions of the electric current both in the liquid and the solid are [9]

$$\vec{j}_s = -\frac{2\sigma_s \sigma_l}{(\sigma_s + 2\sigma_l)} (S_s - S_l) G \vec{i}_z,$$
(1)

$$\vec{j}_l = \frac{\sigma_s \sigma_l}{(\sigma_s + 2\sigma_l)} \left(S_s - S_l\right) G R^3 \left(\frac{2}{r^3} \cos\theta \, \vec{i}_r + \frac{1}{r^3} \sin\theta \, \vec{i}_\theta\right)$$
(2)

The symbols R, G, σ_i , S_i respectively denote the sphere radius, the applied constant temperature gradient, the electrical conductivity and the absolute thermo-electric power of each phases.



Figure 1. Sketch of the geometry and illustration of the thermo-electric current distribution both in the solid and in the liquid as well as the corresponding TEM force under a transverse magnetic field.

Note from equations (1) and (2) that the TEM force acts both on the liquid and the solid. The TEM force on the solid tends to generate a deviation of the particle along the y-direction in the present geometrical conditions. Neglecting the influence of the surrounding flow, an estimate of **the particle**

TEM velocity U may be obtained from the balance between the TEM force on the solid and the Stokes drag. We then obtained the following expression:

$$U = \frac{2R^2}{9\mu} F_s$$
(3)

 μ denoting the dynamic viscosity of the fluid.

The latter phenomenon will be confirmed by the in-situ experiments shown in the next paragraph. The TEM force also generates fluid flows around the particle. Figure 2 provide an example of computed flow obtained by introducing the analytical force expressions (1) and (2) in ANSYS/FLUENT.



Figure 2. View of the flow pattern around the spherical particle in the case of the transverse magnetic field. The parameter values are G = 3000 K/m, sphere radius R = 0.05 mm, B = 0.08 T. The velocity scales is in m/s. $U_{max} = 0.00143$ m/s.

3. In situ observations of the thermo-electric effect on equiaxed grains

In situ experiments were carried out at the European Synchrotron Radiation Facility (ESRF, Grenoble, France). Directional solidification experiments were performed inside a Bridgman furnace, which has been described in detail previously [11]. The thermal gradient *G* (in the *z*-direction) was imposed by two separated heating elements and the solidification was controlled by applying the same cooling rate *CR* on the two heating elements. The temperature adjustment was monitored using two embedded K-type thermocouples, one at the top and one at the bottom on either side of the sample, 2 cm away from each other. A series of solidification experiments were carried out with a same cooling rate *CR* = 2 K/mn, the same magnetic field value, and various temperature gradients *G* decreasing from 20 K/cm to nearly zero. The thin samples studied were 35 mm in length, 5 mm to 6 mm in width and 150 µm to 200 µm in thickness. To minimize grain sinking phenomena, solidifications were conducted on Al – 10 wt.% Cu alloy for which the grain density is weakly larger than the surrounding liquid [12]. The static magnetic field was generated by a cubic neodymium magnet, which was fixed close to the Bridgman furnace and imposed a permanent magnetic field *B*₀ equal to 0.08 Tesla. The direction of the field was set before the experiment parallel (*x*-direction) to the beam, and its intensity was almost uniform over the sample. Some details of the experiments may be found in [10, 11]. The solid-liquid

interface was visualized by X-ray radiography; the main surface of the sample (y-z plane) was set perpendicular to the incident monochromatic X-ray beam.

3.1. Experimental evidence of the TEM force

Figure 3 shows the time evolution of the solidification microstructure for a temperature gradient G = 10 K/cm. The presence of an axial temperature gradient in the molten sample led to a non-homogeneous nucleation phase, from the bottom to the top of the sample. More specifically, first grains nucleated and showed up earlier in the cooler bottom part of the sample. As discussed in a previous paper [13], it can be reasonably assumed that the grain nucleation takes place from heterogeneous nucleation events at the crucible wall/oxide skin at liquidus temperature, or negligibly below. After the nucleation, few grains remained stuck on the oxide layer of the sample but the great majority of the grains moved roughly from the right side to the left side of the sample as indicated by the white arrows in figure 3a to figure 3d. It is worth noticing that the grains continued to grow during their motion. The global movement of the grains caused the equiaxed grains to form a tightly packed dendritic matrix on the left side of the sample, which, gradually moved towards the right side of the sample. This effect is more easily visible in the videos from which figure 3 are taken because grains were forced to move towards one side of the sample during the equiaxed growth. The number of grains across the transverse direction of the sample was non-uniform, as was their size. The fact that all free grains moved from the right side toward the left side of the sample definitely demonstrates that the grains were pushed by a horizontal TEM force in y-direction. Furthermore, the experiments showed that the deviation of the particles with respect to gravity increased with the temperature gradient in agreement with the theoretical analysis.



Figure 3. Sequence of radiographs showing the equiaxed solidification experiment of Al-10wt%Cu (CR = 2K/mn ; G = 10K/cm)under a 0.08T static magnetic field. Equiaxed grains nucleated from the bottom to the top, according to the temperature gradient direction, and then moved from right to left in the sample as indicated by few arrows in figure 3a to figure 3d.

3.2. Analysis of grain velocity as a function of grain size

Measurements of the grain surface and the grain velocity were carried out at each step of the solidification experiments between two successive radiographs. The shape of a dendritic grain is obviously very complex and far from a spherical solid in three-dimensions. For a non-spherical particle, an "equivalent surface diameter" d_s can be defined as the diameter of the disc that would have the same area as the projected image of the irregular dendritic grain. This definition does not take into account phenomena which can take place in the third dimension (sample thickness direction). Figure 4 shows the evolution of the grain velocity U for seven grains as a function of the equivalent diameter d_s . The grains were selected in the same area of the sample to prevent as most as possible any influence of sample thickness, temperature gradient or solute segregation. Despite these precautions, a large dispersion was observed for each value of diameter. In our experiments, the maximum Reynolds number Re = $Ud_{st}/v(v)$ being the kinematic viscosity) is much lower than unity, typically about 10⁻². Therefore, in a first approximation we may assume to be in Stokes regime and the velocity U can be deduced from equation (3). The effect of the thermo-electric flow around the particle is neglected following Baltaretu et al. [17].

A comparison of the Stokes velocity (dotted line in figure 4) with experimental data showed a large discrepancy, although the magnitudes are of the same order. Several effects might be responsible for the aforementioned discrepancy, such as:

(i) A confinement effect due to the lateral wall for large size particles,

(ii) The actual morphology equiaxed grains which are different from that of spheres,

(iii) The non-zero permeability of the equiaxed grains.

Those phenomena have an impact not only on the flow field but also on the electric current distribution, hence the TEM forces. To illustrate the previous discussion, some corrections of the Stokes law were introduced to account for the wall interference, that is to say the morphology and permeability of the particles [14]-[16]. Notes that all the corrections tend to decrease the particle velocity. The results illustrated in figure 4 show that owing to the corrections it is possible to obtain a better agreement.



 $d_{S}(\mu m)$

Figure 4. Plot of the velocity as a function of the equiaxed grain diameter. The symbols represent experimental measurements of seven grains, nucleating almost at the same position in the sample. The Stokes model is indicated by the dotted line, the model adding the wall correction to Stokes law is plotted by a dashed line and the model taking into account both wall confinement and grain morphology is plotted in full line.

4. Conclusions

We have analysed the thermo-electric-magnetic (TEM) effect on solid electrically conducting particles in liquid metal under a static magnetic field. That situation is applied to equiaxed solidification in liquid metals. Firstly, a heuristic theoretical model was developed. It was aimed at determining the TEM body forces acting both in the particle and on the surrounding liquid. It was shown that the particle is submitted to a transverse force, which tends to deflect the grain from the vertical direction. We also performed in situ and real time observations of equiaxed grain motion during directional solidification of Al-10 wt% Cu alloy under a weak static magnetic field (0.08 T). The experimental results show that equiaxed grains were moved approximately along a direction perpendicular to both the temperature gradient and the magnetic field direction, which confirms that the grains are forced to move by gravity force and TEM force. The deviation angle in the early stages of growth for the different temperature gradients were determined and compared to the prediction achieved by using an analytical model for a spherical particle. A quite good agreement was obtained. The variation of the grain velocity as a function of the grains diameter was then measured from the radiographs and compared with predictions of analytical models which take into the account the wall confinement and the grain morphology. Despite the corrections, it appears that the drag force in our experiments is much larger, probably due to grain behaviour (tilting and rotation) during the motion, long-distance interaction with other equiaxed grains and local variation of liquid composition related to solute rejection during solidification and modifications of the electric current distribution.

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