# EXPLOITING THE TEMPERATURE/CONCENTRATION DEPENDENCE OF MAGNETIC SUSCEPTIBILITY TO CONTROL CONVECTION IN FUNDAMENTAL STUDIES OF SOLIDIFICATION PHENOMENA

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# INTRODUCTION

The objective of this new research project is to demonstrate by experiment, supplemented by mathematical modeling and physical property measurement, that the effects of buoyancy driven convection can be largely eliminated in ground-based experiments, and further reduced in flight, by applying a new technique. That technique exploits the dependence of magnetic susceptibility on composition or temperature. It is emphasized at the outset that the phenomenon to be exploited is fundamentally and practically different from the magnetic damping of convection in conducting liquids that has been the subject of much prior research.

The concept suggesting this research is that all materials, even non-conductors, when placed in a magnetic field gradient, experience a force. Of particular interest here are paramagnetic and diamagnetic materials, classes which embrace the "model alloys", such as succinonitrile-acetone, that have been used by others investigating the fundamentals of solidification. Such alloys will exhibit a dependence of susceptibility on composition. The consequence is that, with a properly oriented field (gradient) a force will arise that can be made to be equal to, but opposite, the buoyancy force arising from concentration (or temperature) gradients. In this way convection can be stilled.

The role of convection in determining the microstructure, and thereby properties, of materials is well known. Elimination of that convection has both scientific and technological consequences. Our knowledge of diffusive phenomena in solidification, phenomena normally hidden by the dominance of convection, is enhanced if we can study solidification of quiescent liquids. Furthermore, the microstructure, microchemistry and properties of materials (thereby practical value) are affected by the convection occurring during their solidification. Hitherto the method of choice for elimination of convection has been experimentation in microgravity. However, even in low Earth orbit, residual convection has effects. That residual convection arises from acceleration (drag on the spacecraft), displacement from the center of mass or transients in the gravitational field (g-jitter). There is therefore a need for both further reducing buoyancy driven flow in flight and allowing the simulation of microgravity during ground based experiments.

### **PREVIOUS INVESTIGATIONS**

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Two publications of great relevance to this investigation are those of Braithwaite et al. (1) and Beaugnon et al. (2). These investigators at the Centre de Recherches sur les Très Basses Température in Grenoble, have described how a material in a magnetic field gradient is subjected to a force (per unit volume)

$$\mathbf{F} = -\left(\frac{\chi}{2\mu_0}\right)\nabla(\mathbf{B}^2) \tag{1}$$

where  $\chi$  is the magnetic susceptibility,  $\mu_0$  is the permeability of vacuum and B is the magnetic flux density.

If the susceptibility varies with the temperature in a fluid with a temperature gradient, then the effect is to enhance or oppose the normal buoyancy driven convection. The effect can be described by a magnetic Rayleigh number given by these works as

$$\mathbf{Ra}_{\mathrm{m}} = \mathbf{Ra} \left( 1 + \frac{\mathbf{F}}{\mathbf{g}} \left( 1 + \frac{\gamma}{\beta} \right) \right)$$
(2)

where Ra is the usual Rayleigh number, g is the acceleration due to gravity (or microgravity),  $\beta$  is the volumetric expansion coefficient and  $\gamma$  is the fractional rate of change of susceptibility with temperature:

$$\gamma = \frac{\left|\frac{1 \, \mathrm{d}\chi}{\chi \, \mathrm{d}T}\right|}\tag{3}$$

In the case where the magnetic force is aligned opposite to the gravitational force (F and g have opposite signs), the magnetic Rayleigh number is reduced to zero at some value of  $\nabla(B^2)$  for any liquid, halting buoyancy driven flow.

Fig. 1 is taken from the results of these French investigators; these are measurements of heat flux (expressed as a Nusselt number) from a hot to a cold plate separated by a paramagnetic solution (gadolinium nitrate in water). The apparatus was located in a superconducting magnetic and the parameter on the curves is the value of  $\nabla B^2$  in  $T^2/m$ . With the magnetic force applied opposite to gravity, convection was suppressed, apparently being eliminated at 15  $T^2/m$ .

By obvious extension of the work of these investigators, solutal convection should be suppressed in the case of a solution with a concentration dependent susceptibility when

$$\mathbf{Ra'_m} = \mathbf{Ra} \left( 1 + \frac{\mathbf{F}}{\mathbf{g}} \left( 1 + \frac{\gamma'}{\beta'} \right) \right)$$
(4)

equals zero, where Ra'<sub>m</sub> is the magnetic Rayleigh number for solutal convection

 $\beta' = \begin{vmatrix} 1 & \frac{d\rho}{\rho} \\ \frac{dQ}{dQ} \end{vmatrix}$  is the fractional change of density with concentration  $\gamma' = \begin{vmatrix} 1 & \frac{d\chi}{dQ} \end{vmatrix}$  is the fractional change of susceptibility with concentration.

Again it is emphasized that this halting of buoyancy driven flows is different from the usual magnetic damping. Conventional magnetic damping relies on induced currents arising from the cross product of the velocity vector and the magnetic flux density; if the velocity is zero (or if the material is non-conducting) the damping force is zero, i.e., the force is only operative when motion exists and should therefore completely halt the flow only in the limit of an infinite magnetic field. Furthermore, because model alloys are usually non-conducting (being transparent for ease of observing the solidification) they cannot be arrested by this conventional magnetic damping. The opportunity for controlling flow by exploiting the variation in dependence of susceptibility of the fluid has been noted by Edwards and co-workers (3).

## **RESEARCH PROJECT DESCRIPTION**

The investigation, extending over four years, entails construction of an apparatus in which buoyancy driven flows can be measured during solidification of transparent liquids, selection of suitable liquids (model alloys) by physical property measurement, running the apparatus in the High Magnetic Field Solidification Facility at MSFC to demonstrate that convection can be controlled or eliminated, and mathematical modeling of the convection within a magnetic field. The apparatus will then be used for studying convection free solidification as well as other phenomena (e.g., Soret effect) that require minimization of convection.

### THEORY

The total (gravitational plus magnetic) body force components acting on a material placed within a magnet with axial symmetry are

$$F_{z \text{ Tot}} = \rho g - \frac{\chi}{\mu_0} \left( B_z \frac{\partial B_z}{\partial z} + B_r \frac{\partial B_r}{\partial z} \right)$$

$$F_{r \text{ Tot}} = -\frac{\chi}{\mu_0} \left( B_z \frac{\partial B_z}{\partial r} + B_r \frac{\partial B_r}{\partial r} \right)$$
(5)

For the moment it will be assumed that a region can be found inside the magnet where the axial magnetic body force

$$F_{z \text{ Mag}} = -\frac{\chi}{\mu_0} \left( B_z \frac{\partial B_z}{\partial z} + B_r \frac{\partial B_r}{\partial z} \right)$$
(6)

is uniform (showing no variation in the axial or radial direction), i.e.,  $B_z \frac{\partial B_z}{\partial z} + B_r \frac{\partial B_r}{\partial z}$ 

is constant. Furthermore it is assumed that the radial component of the force is negligible. If buoyancy driven by concentration gradients is to be eliminated,

$$\frac{\mathrm{dF}_{\mathrm{Tot}}}{\mathrm{dC}} = 0 \tag{7}$$

Introducing the fractional variation of density ( $\beta'$ ) with concentration (C), there will be no buoyancy driven convection when

$$B_{z}\frac{\partial B_{z}}{\partial z} = g\rho\beta'\mu_{0}/\frac{d\chi}{dC}$$
(8)

The magnetic field and magnetic forces for a material with the susceptibility of water located within the superconducting magnet of the High Magnetic Field Solidification Facility at MSFC have been calculated and are depicted in Figs. 2 and 3 respectively. The current through this magnet is controllable and the units in Fig. 2 are T/Amp while in Fig. 3 they are  $N/m^3A^2$ . The origin of the co-ordinate system is at the center of the magnet and the region depicted is from that center to the inner radius of the magnet (radially) and to just above the mouth of the magnet (vertically). The magnetic field appearing in Fig. 2 is the anticipated one; the field is strongly axial inside the magnet, diverging as the mouth is approached. It appears from Fig. 3 that the assumption of a uniform axial force and zero radial force is approximately satisfied over a volume of 10cms height and 10cms radius roughly halfway between the center and mouth of the magnet. It is in this volume that the experiment is to be conducted.

#### EXPERIMENTAL

A sketch of the experiment to be incorporated in the superconducting magnet appears in Fig. 4. The model alloy under investigation is contained in a chamber, with temperature controlled walls, located in the bore of the magnet. The motion of the liquid is to be followed as a function of the current of the magnet. The intended technique for measuring the liquid motion is particle image velocimetry. This is a technique whereby motion of particles suspended in the fluid is tracked by a digital camera and velocity maps extracted from the frame-to-frame movement by commercial software. It can only be successful if the terminal velocity of the particles is negligible; in the present case this must hold for terminal velocities resulting from magnetic, as well as gravitational, forces. The particles in one plane are illuminated by a sheet of laser light from the optical scheme shown in Fig. 4. Prior to such experimental work it is necessary to decide on appropriate model alloys. In addition to the usual requirements of mimicking the solidification of practical materials (such as metallic alloys) and transparency, the alloy chosen must exhibit significant variation of susceptibility with composition. Unfortunately, although the susceptibilities of pure liquids are available in the literature, there is little published on susceptibility of solutions. It has therefore been necessary to embark on susceptibility measurements. Fig. 5 shows preliminary results of measurements on ammonium chloride-water which has been used by others (4) as a model alloy. The measurements were made using a Quantum Design magnetic property measurement system (SQUID) with a field strength of 2 Tesla.

## REFERENCES

(1) Braithwaite, D., Beaugnon, E. and Tournier, R., Nature 354, 1991, 134.

(2) Beaugnon, E. et al., J. Phys. I France 3, 1993, 399.

(3) Edwards, B.F., Gray, D. D. and Huang, J., Third Microgravity Fluid Physics Conference, 1996, 711.

(4) Beckermann, C. and Viskanta, R., 1988, PhysicoChem. Hydrodyn. 10, 195.

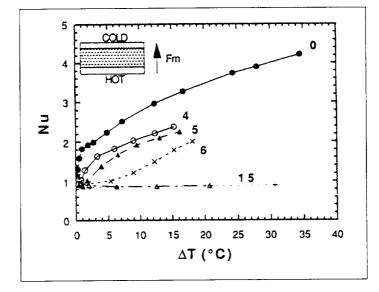


Fig. 1 Plot of Nusselt number versus thermal gradient (taken from ref. 3).

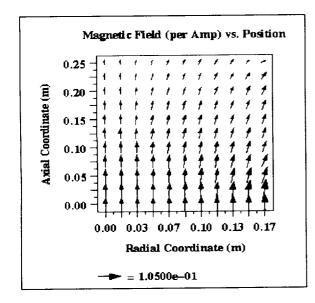


Fig. 2 Plot of magnetic field versus position for the superconducting magnet of the HMF Facility at MSFC.

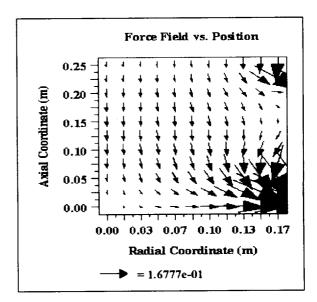


Fig. 3 Plot of magnetic forces for the superconducting magnet of the HMF Facility at MSFC.

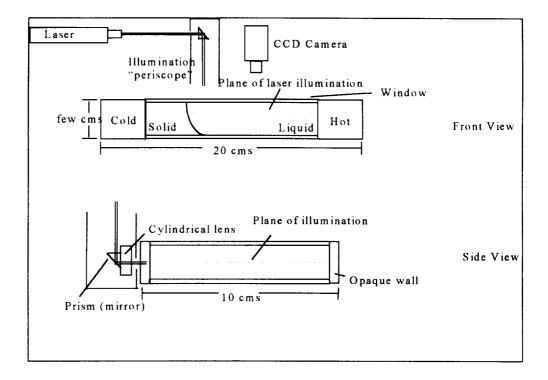


Fig. 4 Sketch of experiment for use in HMF Facility at MSFC.

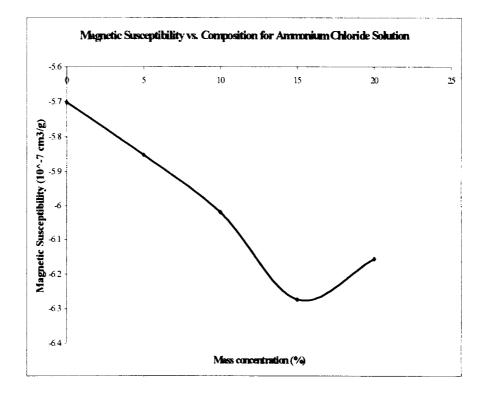


Fig. 5 Plot of magnetic susceptibility versus concentration for ammonium chloride solution.