

A NEW TECHNIQUE FOR MEASURING
THE WETTABILITY OF FIBROUS PREFORMS
BY LIQUID METAL

by

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ABSTRACT

In this thesis a method to dynamically measure the capillary pressure drop during infiltration of a fibrous preform by pure metal is derived from first principles. This capillary pressure drop results from the metal and the fibers forming a non-wetting system. Experiments are conducted with Saffil fibers and 99.999% aluminum to measure values for capillary pressure drop. The results of the experiments show a capillary pressure drop on the order of 1 Mpa with approximately 15% measurement error.

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Introduction

In recent years interest in metal matrix composites has been steadily increasing due to the remarkable mechanical properties that can be achieved with these materials and the availability of affordable fibrous reinforcements. Much research interest has been focused on composites reinforced with "Saffil", a silica bonded alumina fiber preform manufactured by ICI in the U.K. This fiber is used commercially by Toyota in producing a reinforced aluminum piston head, the first metal matrix composite application to be mass produced. This part is manufactured by the squeeze casting method, whereby molten metal is forced into the fibrous preform by a mechanical ram.

The potential of casting methods for economical production of metal matrix composite materials has recently motivated research into the fundamentals of metal flow and solidification during infiltration of a fibrous preform. One of the chief difficulties in the solidification processing of fiber reinforced metals is the fact that usually the metal does not wet the fibers. A simple analogy is that of a water repellent fabric which can not under normal circumstances be combined with water. When the metal is forced into the fiber preform, a pressure drop due to surface tension effects is observed at the infiltration front. This capillary pressure drop has been theoretically predicted from the wetting action of liquid metal and fibers, but a value has not been

determined experimentally to date. In this thesis a method to experimentally measure this capillary pressure drop is developed. Experiments are conducted to find actual values for the capillary pressure drop when pure aluminum is infiltrating Saffil preforms under various processing conditions. When producing a final MMC part, such as the Toyota piston head, capillary pressure is an important factor to know because it is the minimum pressure necessary to achieve any infiltration.

Theory

A model describing the flow of pure metal into a fibrous preform was recently developed by L. Masur [1]. The theory pictures flow of metal into the voids of a fiber preform. The metal first enters the larger pores and solidifies as a sheath around the fibers if the latter are initially at a temperature below the melting point of the metal. Behind the solidification tip new metal continues to flow past the solidified metal, encountering new fibers where the process occurs again. This flow and solidification would continue indefinitely if the cooling effects of the mold wall did not eventually choke off flow, ceasing any further infiltration. It is presented that infiltration of a preform can be described by the combination of a flow through a porous medium solution and a solidification during flow solution.

For this thesis, equations describing flow through porous media are used to establish a method for determining capillary pressure.

As is described by Masur, Darcy's law describes flow of the metal through the fibrous preform. When flow is unidirectional and gravity effects are neglected Darcy's law can be written as:

$$V_o = \frac{K dP}{\mu dx} \quad (1)$$

$$V_o = (1 - V_f) \frac{dL}{dt}$$

K = permeability of fibrous preform

μ = viscosity of liquid metal

V_f = volume fraction of preform

Preform permeability, K , is a function mainly of fiber volume fraction and temperature, and is calculated as presented in reference [1]. With K constant everywhere and when infiltration is at constant pressure, integrating equation (1) gives:

$$\frac{L}{\sqrt{t}} = \sqrt{\frac{2 \Delta P \mu K}{\mu (1 - V_f)}} \equiv \Psi \quad (2)$$

Infiltration is then parabolic, and Ψ is a measure of permeability specific for a given set of processing conditions. As the parameters of fiber temperature, metal

temperature, volume fraction and pressure are changed, both the preform permeability and the metal viscosity change, resulting in a ψ specific to that set of conditions.

In the equation above ΔP_μ represents the pressure drop due to viscous drag. It is, in fact, the pressure drop in the metal measured between the entrance of the preform and a point right behind the infiltration front. Therefore, if ΔP_T is the total pressure applied to drive the metal into the preform and ΔP_γ is the capillary pressure drop at the infiltration front, we have

$$\Delta P_T = \Delta P_\mu + \Delta P_\gamma \quad (3)$$

The capillary pressure drop can be envisioned as the minimum pressure necessary to force the metal to enter the voids in the fibrous preform. Normally the metal and fibers are a non-wetting system. As with a drop of water sitting on water resistant fabric, the liquid forms a bead and does not wet the fibers. In both cases a certain amount of hydrostatic pressure is required before the liquid will enter the spaces among the fibers. This pressure can be quantified by:[2]

$$\Delta P_\gamma = \frac{2V_f(\sigma_{FL} - \sigma_{FA})}{(1 - V_f)r_f} \quad (4)$$

ΔP_γ = capillary pressure due to surface tension

σ_{f1} = fiber/liquid metal interfacial energy

σ_{fa} = fiber/atmosphere interfacial energy

r_f = radius of fibers

In the work of L. Masur $\Delta P\gamma$ was measured by an isothermal breakthrough experiment (see reference [1]). The results of this experiment showed that a nominal breakthrough pressure of 0.3 Mpa is needed to infiltrate the majority of voids in the preform. This value was largely treated as an insignificant factor, and thus only the viscous pressure drop was considered in further calculations. When capillary pressure drop is included in equation 2:

$$\Psi \equiv \frac{L}{\sqrt{t}} = \sqrt{\frac{2(\Delta P_T - \Delta P_\gamma) K}{\mu(1 - V_f)}} \quad (5)$$

ΔP_T = total pressure drop

ΔP_γ = capillary pressure drop

Rearranging this equation into a linear form.

$$\frac{\mu(1 - V_f)}{2K} \Psi^2 = \Delta P_T - \Delta P_\gamma \quad (6)$$

ΔP_γ can now be determined experimentally by using equation 6. Values for Ψ can be measured at various ΔP_T by plotting L vs \sqrt{t} , where the slope is defined as Ψ . Measurement of Ψ in this manner has been shown by Masur to be quite accurate. Results of several experiments at different ΔP_T can be plotted in the form of equation 6. By plotting the data in a linear manner

$\Delta P\gamma$ can be read from the graph as the X-axis intercept. One originality in this experimental technique is that wettability is measured dynamically, while the metal infiltrates and exchanges heat with the fibers. Inherent in this method is the assumption that $\Delta P\gamma$ is a constant for different applied pressures.

Experimental Apparatus and Procedure

For these experiments the apparatus and materials were identical to those used by L. Masur. A schematic diagram of the pressure casting device is shown in figure 1. This type of caster is unique in its ability to independently control metal temperature, fiber temperature and pressure. To operate the device a fibrous preform is placed into a quartz tube and secured to the cap through pressure fittings. A resistance heater is put in place to heat the fibers to the preheat temperature T_f . In the bottom of the vessel another furnace melts metal in a graphite crucible. When the correct metal and fiber temperatures are reached the cap is lowered onto the vessel and secured within a steel frame. As nitrogen gas is delivered to the chamber through the inlets, hydrostatic pressure on the metal forces it into the fibers.

Figure 2 shows the arrangement used to measure the liquid metal position. To measure metal position during infiltration a SiC fiber is pushed through a preform. At the

top it is connected to the circuit as shown in the figure. When the metal, coming up the quartz tube, touches the bottom of the SiC fiber the circuit is completed and the chart recorder reads a voltage. As the metal continues into the preform the effective length and the net resistance of the SiC fiber decreases. The chart recorder measures the change as a decreasing voltage. The voltage trace can then be transformed into length and time data.

In these experiments 99.999% pure aluminum was used for the matrix metal. The entire melt was replaced every 8-10 experimental runs to prevent excessive contamination of the metal. As mentioned by Masur a significant difference in infiltration behavior is noticed once the metal purity reaches 99.95%.

The experimental procedure consists of running one set of fiber temperature, metal temperature and volume fraction conditions at different pressures. The actual experimental run is done identically to that described by L. Masur. Data from the SiC fiber is transformed into L and t and plotted as L^2 vs. t . A typical plot is shown in figure 3. Ideally this plot should be linear with a slope of ψ^2 . However the curve is only linear in the middle section. For the first ~1.2 seconds nitrogen is entering the vessel and increasing the pressure to the final constant value. Because the pressure is not constant, ψ is not constant during that period. ψ also changes at the end of the infiltration curve when

solidification from the die walls starts to choke the flow of metal. Thus when measuring the ψ value only the first three data points after 1.2 seconds are used for the least square fit.

From these L^2 vs t curves, ψ values are measured for several different infiltration pressures and then are plotted in the form of equation 6, where K is calculated as in reference [1].

Results

The variation in processing parameters were limited by several factors. First, volume fractions available were limited to a range of 23% to 25% solid fraction. This is a limitation imposed by the batch of Saffil preforms that were available. Also fiber temperature, pressure and metal temperatures were limited by the apparatus. It was not possible to use processing conditions that resulted in a final composite of greater than 45 mm in length.

Another limitation existed at low pressure (< 1.3 Mpa). In this situation the total infiltration time is on the order of 5 seconds. In this time period a linear portion of the L vs \sqrt{t} plot does not exist because the die solidification effects become pronounced just as the pressurization transient finishes. Thus it was not possible to measure ψ values below pressures of 1.3 Mpa.

The result of a series of experiments at one set of processing conditions is a curve of Ψ vs pressure. Results for three sets of processing parameters are given in figures 4,5, and 6. Note that in all three curves, excepted for the data measured at the maximum pressure, all points lie on a straight line. These points are fit to a least square line where the x-axis intercept is $\Delta P\gamma$. As can be seen in figures 4,5, and 6 values of $\Delta P\gamma$ range from 0.92 Mpa to 1.22 Mpa.

Discussion

The results from these experiments show a $\Delta P\gamma$ value approximately three times as large as that measured by Masur in his breakthrough experiments. This is not surprising because the breakthrough experiments were run at isothermal temperatures where $T_f = T_m$. In these experiments the interfacial surface temperature is somewhere between T_f and T_m . At the surface of a fiber very rapid heat transfer occurs that raises fiber temperatures to some T_i while reducing metal temperatures to the same value. Thus the T_i is lower in this case than the isothermal experiment. As is demonstrated by S. Oh with SiC particles and aluminum, temperature has a marked influence on capillary pressure [3].

When $\Delta P\gamma$ of figure 4 is compared to figure 5 a lower value is found. This is as expected from equation 4 because a

smaller volume fraction is used. Equation 4 can also be used to gather a measure of the error in the experimental procedure. For figures 4 and 5 the only difference in processing parameters is the difference in volume fraction. Thus the ratio of interfacial energies and fiber radius is constant for both series of experiments. When the ratio $V_f/(1 - V_f)$ is used to compare the results of the two figures a discrepancy on the order of 15% is found. This will be taken as an estimation of experimental error in these experiments.

When a comparison is made between figures 5 and 6 theory is not supported. Theory calls for a lower $\Delta P\gamma$ for figure 5 because a higher fiber temperature is used, just the opposite is found. The error can be attributed to the experiments in figure 6. In this series of runs the low fiber temperature used caused a short infiltration length, making it difficult to accurately read ψ . Uncertainty in the measurement of ψ might lead to the error found in $\Delta P\gamma$.

If theoretical values of Ψ are plotted by equation 6 with a $\Delta P\gamma = 0$ a line of slope one, intercepting the origin results. Ideally the experimental results should be a line parallel, but shifted to the right by $\Delta P\gamma$. The figures show slopes of the experimental lines are 20- 30% greater than one, however this is within an acceptable margin of error.

In fitting a least squares line to the data, points at pressures greater than 4.3 Mpa were ignored. This was done because the ψ values obtained were consistently lower than the

theoretically predicted values. It is likely that at higher pressures some sort of mechanical limitation to flow takes place. One possibility is that the metal flows so quickly into the fibers that the displaced gas creates a back pressure, slowing the metal flow and resulting in a lower measured value of ψ .

Another possibility is that the preform could be compressed by the metal. At these pressures the ceramic support rod leaves an indentation in the top of the preform, something not seen in lower pressure experiments. This is caused by the force of the metal when it first reaches the fibers. If the fibers did compress, the infiltration behavior will have varied in a complex manner resulting in a lower measured ψ value.

Conclusions

1.) A method is found for determining capillary pressure due to the non-wetting behavior of the system. This involves experimentally determining values for the parameter ψ and then plotting the results in a linear form. The X-axis intercept of the line is the capillary pressure.

2.) Experiments with Saffil fibers and 99.999% aluminum show the value for $\Delta P\gamma$ range from 0.92 Mpa to 1.22 Mpa. with an estimated experimental error of 15%.

3.)At high pressures the experiments show consistent discrepancies with the theoretical predictions . This is attributed to a mechanical limitation of the apparatus.

Special thanks goes to Larry Masur for his hours of help and guidance.

Nomenclature

K = preform permeability

L = distance from bottom of preform to metal front

μ = metal viscosity

ΔP = applied pressure

$\Delta P\gamma$ = capillary pressure drop

$\Delta P\mu$ = viscous pressure drop

ΔP_T = total pressure drop

r_f = fiber radius

σ_{f1} = fiber/liquid metal interfacial energy

σ_{fa} = fiber/atmosphere interfacial energy

t = time from start of infiltration

T_f = fiber preheat temperature

T_i = interfacial surface temperature

T_m = liquid metal temperature

V_f = fiber volume fraction

V_o = velocity of metal front

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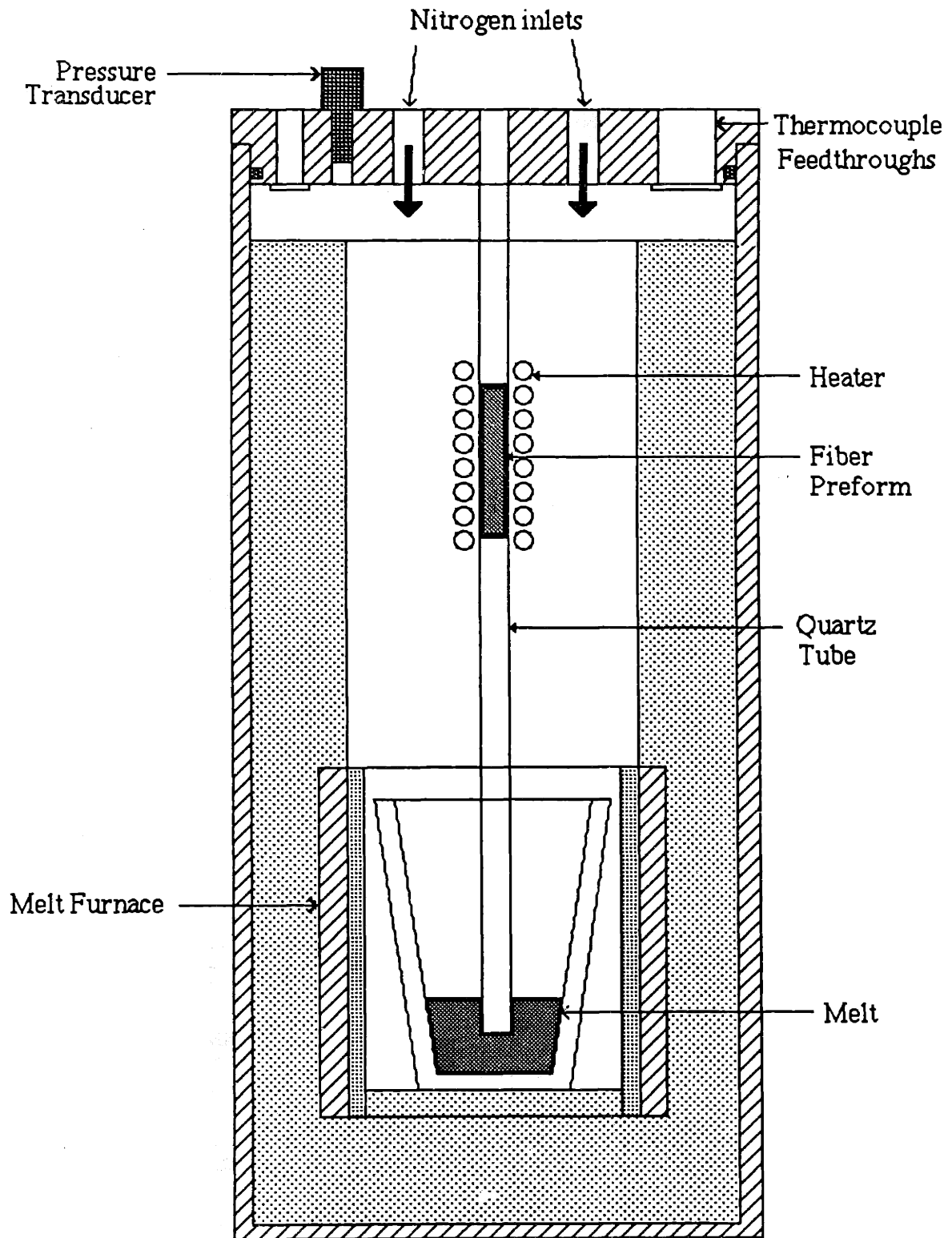


Figure 1: Schematic drawing of pressure caster

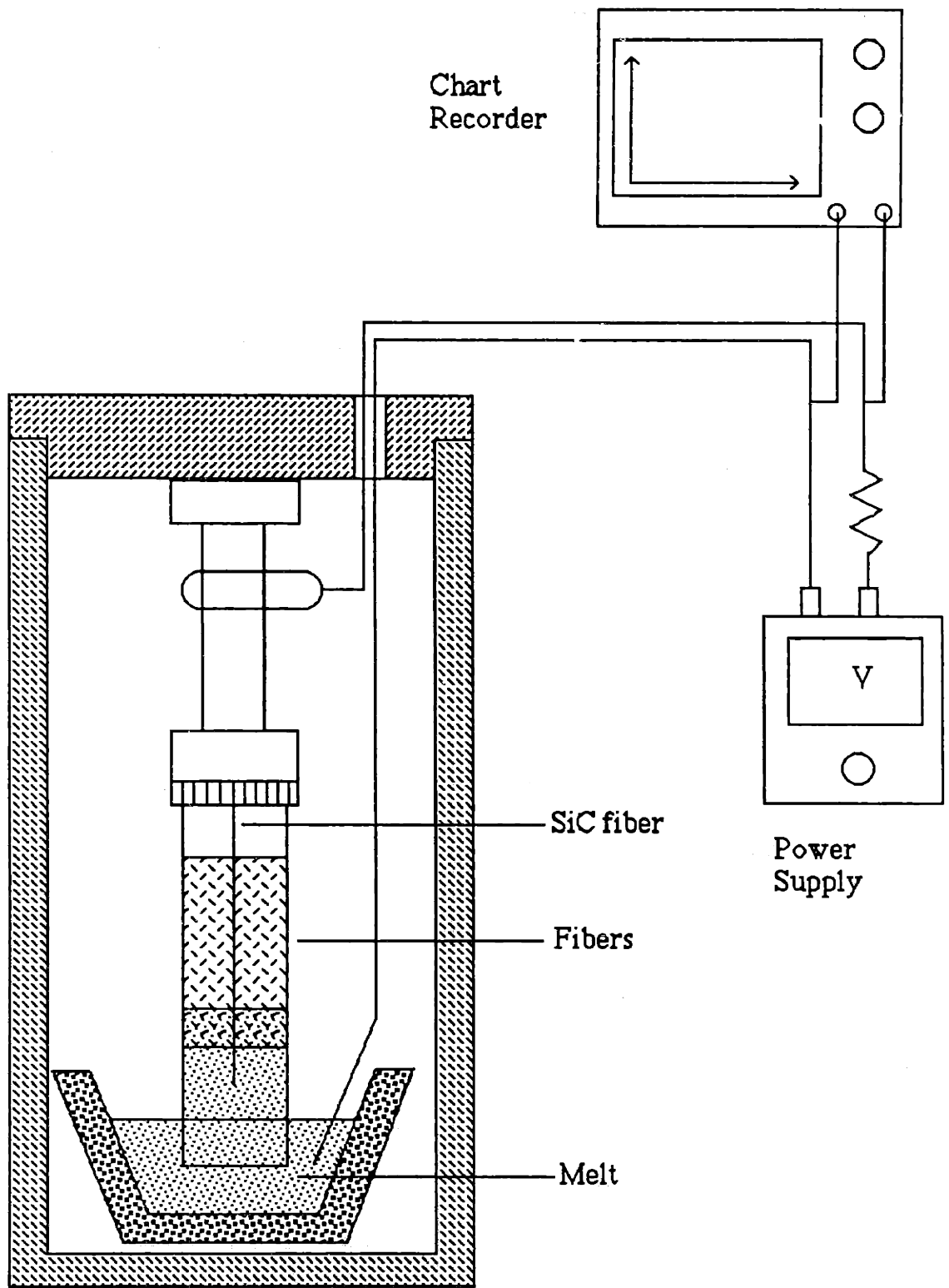
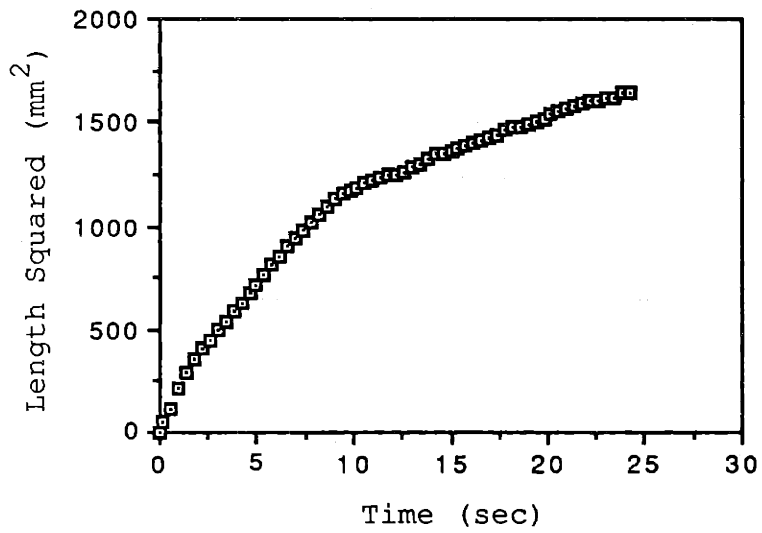


Figure 2: Schematic of liquid metal position sensor



99.999% Al
Alumina fiber
3.8 μ dia.
Tf = 285 C
Vf = 0.25
 $\Delta P = 4.38$ Mpa

Figure 3: Example plot of L^2 vs t

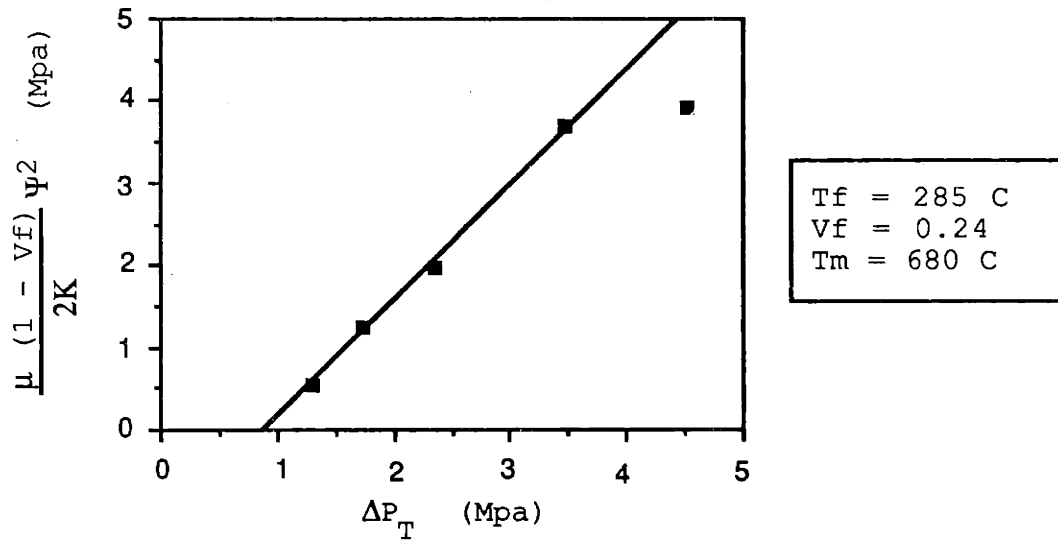


Figure 4: Plot of Ψ in form of equation (6)

$$\Delta P_\gamma = 0.92 \text{ Mpa}$$

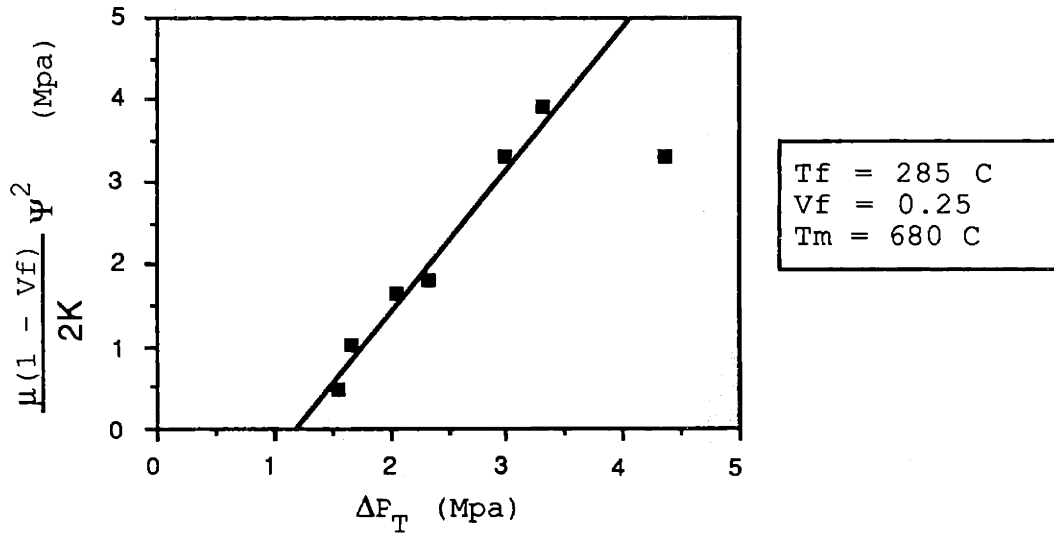


Figure 5: Plot of Ψ in form of equation (6)

$$\Delta P_\gamma = 1.22 \text{ Mpa}$$

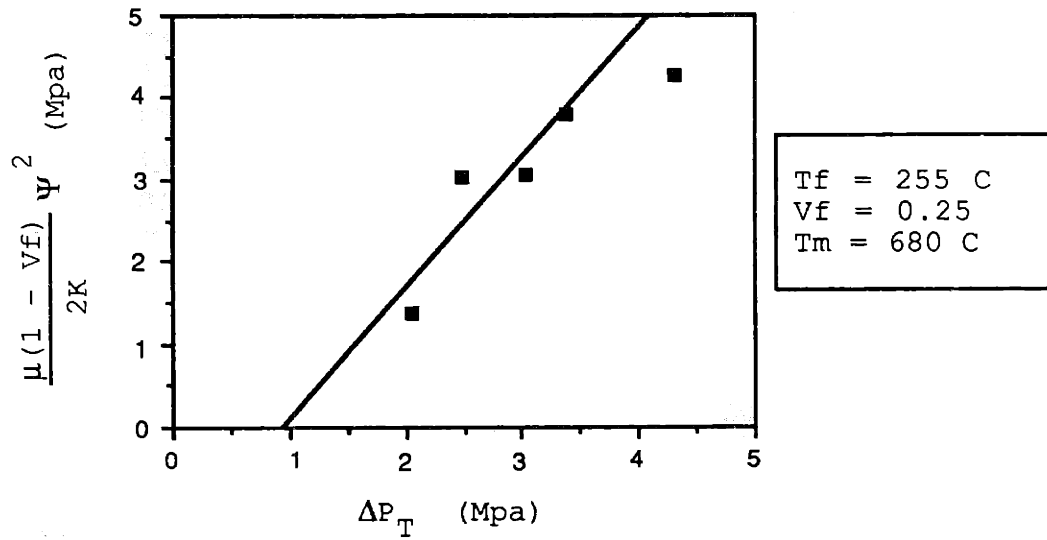


Figure 6: Plot of Ψ in form of equation (6)

$$\Delta P_\gamma = 0.95 \text{ Mpa}$$