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TEMPERATURE PROFILES IN HIGH GRADIENT FURNACES

by

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INTRODUCTION

Accurate temperature measurement of the furnace environment is very important in both the science and technology of crystal growth as well as many other materials processing operations. A high degree of both accuracy and precision is acutely needed in the directional solidification of compound semiconductors in which the temperature profiles control the freezing isotherm which, in turn, affects the composition of the growth with a concomitant feedback perturbation on the temperature profile.

Directional solidification, by its very nature, requires a furnace configuration that will transport heat through the sample being grown. A common growth procedure is the Bridgman Stockbarger technique which basically consists of a hot zone and a cold zone separated by an insulator. In a normal growth procedure the material, contained in an ampoule, is melted in the hot zone and is then moved relative to the furnace toward the cold zone and solidification occurs in the insulated region. Since the primary path of heat between the hot and cold zones is through the sample, both axial and radial temperature gradients exist in the region of the growth interface.

The interaction of temperature profile and interface shape was first shown in the seminal paper by Chang and Wilcox[1] and then expanded by others[2-4]. The effect of the interface shape on composition and convection in the liquid has also been shown in the literature[5-8]. The temperature and thermophysical properties of the sample also interact with those of the furnace wall and result in a growth rate that does not equal the relative furnace-ampoule translation rate[9-11]. From this discussion, there is clearly a need to know the temperature profile of the growth furnace with the crystal that is to be grown as the thermal load. However it is usually not feasible to insert thermocouples inside an ampoule and thermocouples attached to the outside wall

of the ampoule have both a thermal and a mechanical contact problem as well as a view angle problem.

The objective of this paper is to present a technique of calibrating a furnace with a thermal load that closely matches the sample to be grown and to describe procedures that circumvent both the thermal and mechanical contact problems.

PROCEDURE

The procedure is actually in three parts. First to be discussed is the selection of a suitable material that will properly emulate the crystal to be grown. Next the thermocouples have to be fixed to this test sample to overcome the mechanical, thermal and view factor problems. The third step is to perform an in-situ measurement of the thermocouple positions at the growth temperature. Each of these steps will be discussed in turn.

Since the primary heat flow between the hot and cold furnace zones is through the sample (and ampoule walls) it is necessary to match the thermophysical properties of the sample to those of the crystal to be grown. A complete match would include matching the thermal conductivity, thermal diffusivity and emissivity of the two. Since these factors are functions of temperature, with a discontinuity at the interface, a full match is virtually impossible especially if the latent heat of fusion is considered. The most important single factor and easiest to match is thermal conductivity. The thermal diffusivity becomes important in transient portions of the testing and can also be important when using high growth rates or in samples of high values of diffusivity. The match of emissivity can be accomplished by choosing the same ampoule for the test sample as will be used for the crystal. This ampoule choice is necessary if its thermal conductivity is on the same order or higher than that of the sample. However if the ampoule is transparent over a significant range of the thermal emission of the furnace (note Wein's law) then the sample/ crystal emissivity match still has to be considered.

Once the sample material has been chosen, it has to be instrumented for temperature measurement. Although various temperature sensitive resistors might be used for measurement the most common procedure is to use bimetallic thermocouples and this will be the only technique discussed in this paper.

The thermocouples have to be attached to the ampoule to obtain a sound mechanical and thermal contact. If pressed only to the surface of the sample the thermocouple is only in partial contact and will measure an average temperature between the sample and the ambient. The lack of a sound contact will also result in a relative motion between the sample and the thermocouple due to the mismatch of thermal coefficients of expansion. If the thermocouples are inside an ampoule then the emissivity factor can be ignored to the extent that the ampoule is opaque to the incident radiation.

Concomitant to the problem of a sound thermal contact is the need to minimize the heat flow in the thermocouple wires.[11] This is especially important for low thermal conductivity samples in which a significant portion of the total heat flux could be in the metal thermocouple leads.

The final test for mechanical integrity of the contact is to x-ray the instrumented sample at both room temperature and at the maximum operating temperature to observe the movement of the thermocouple wires. Since most operational furnaces are not amenable to x-ray penetration, a special furnace can was built for this test. The precise temperature distribution was not important for this test.

An additional variable that has to be considered in furnace calibration is the pull rate or simulated growth rate of the sample. Heat is transferred from the hot zone to the cold zone by both thermal conductivity and, when the sample is physically moving, by heat capacity and the temperature difference between zones. This effect, often characterized by the Peclet number, is not easily quantifiable without numerical analysis or physical measurement.

RESULTS

The material chosen for the test sample was fused silica (SiO_2). This material was chosen because of its inert nature, the ease of thermocouple insertion and, primarily, because of its thermal conductivity match to the crystalline material of interest, PbSnTe.

The relative movement of the thermocouples with respect to the sample is shown in figure 1. The thermocouples are attached to a fused silica rod by various techniques and inserted into a furnace. The cross hairs are wires placed on the exterior of the furnace and are not affected by the heating of the furnace; hence they represent a fixed reference point. Figure 1a shows the setup at room temperature and figure 1b shows the same setup at 1000°C. The relative movement of thermocouples with respect to the lab frame is as much as 2.5mm.

This extent of movement is clearly unacceptable when trying to characterize a high temperature, high gradient furnace in which it is required to measure a 1°C difference between the sample center and surface while the axial gradient may be in excess of 150°C/cm. Also note that the attachment schemes shown in figure 1 do not compensate for axial heat flow nor do they form a sound contact to the sample.

Figure 2 is a drawing that shows an improved technique in which a similar silica rod is used but the thermocouples are attached in a superior fashion. In this technique of thermocouple attachment small holes are drilled into the fused silica with an ultrasonic impact grinder. The holes are sized to accommodate the sheathed thermocouple.

The effect of translation speed on the thermal profile is shown in figure 3. In this experiment a thermocouple is attached to a fused quartz rod and translated through the furnace first at one centimeter per hour then at one centimeter per minute. The slow pull rate is a quasi static measurement and it can be seen that the higher pull rate can displace the liquid solid interface by as much as 2.5cm. This amount of displacement can move the interface from the hot zone to the cold zone which means that the interface could potentially change from convex to concave by just changing the pull rate. This type of profile shift with respect to ampoule pull rate has also been shown by Clyne[11] in liquid metals.

SUMMARY

The thermal profile within the crystal growth ampoule must be accurately known to check thermal modeling and to predict interface shape and extent of fluid convection. An instrumented, inert, sample can be constructed to measure axial gradients in the typical Bridgman Stockbarger directional solidification furnace. Specific care has to be taken to avoid undesired thermocouple movement.

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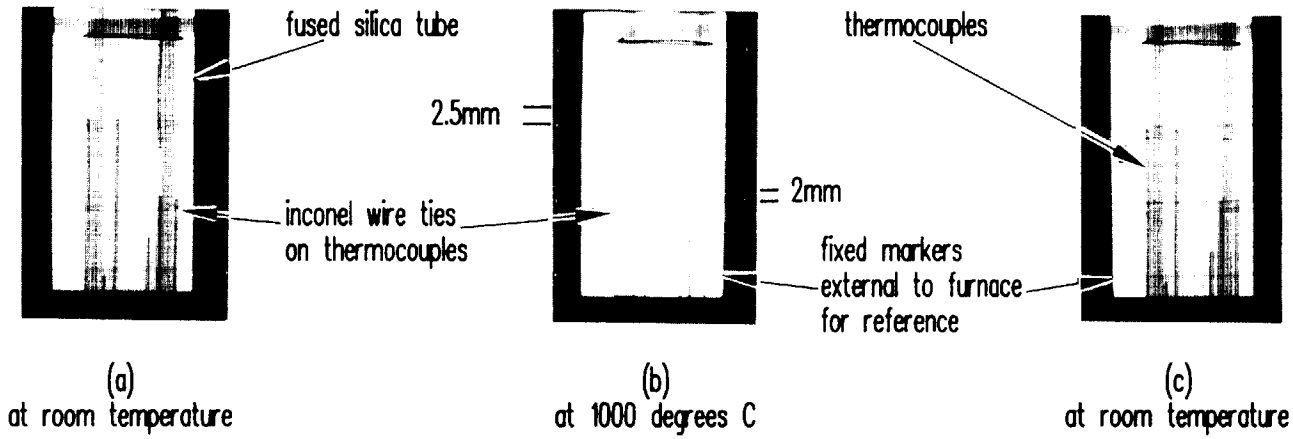

FIGURE 1. X-ray image of thermocouple movement due to temperature change. Six thermocouples were placed circumferentially in grooves on a fused silica tube. All thermocouples were tied to the tube with inconel wire. Three were additionally bonded with ceramic cement. Figure 1a is an X-ray image taken before heating. Figure 1b is an image taken at 1000 degrees C. Figure 1c is an image taken after the furnace had returned to room temperature.

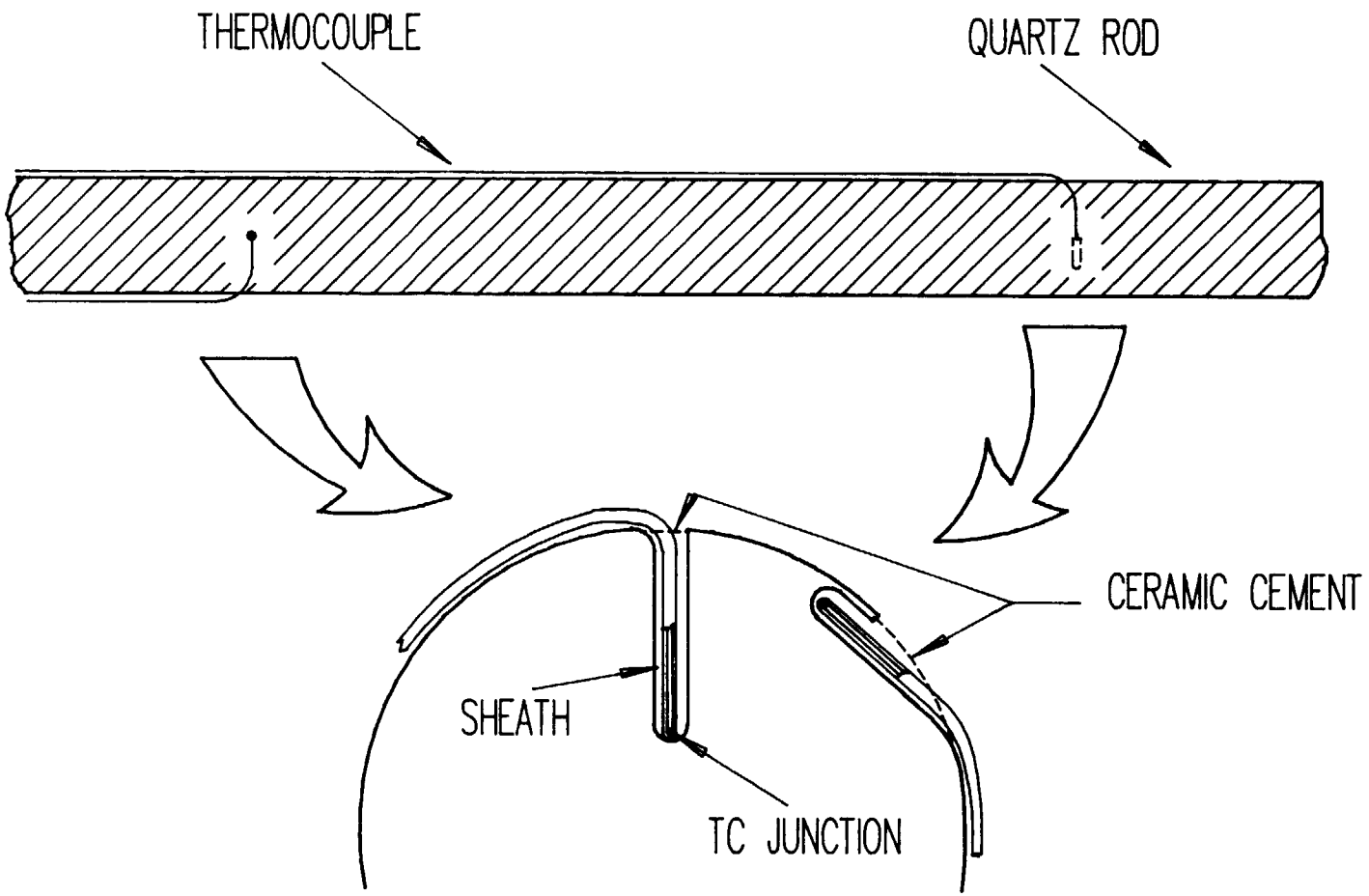

FIGURE 2. Techniques for mounting thermocouples in quartz rod.

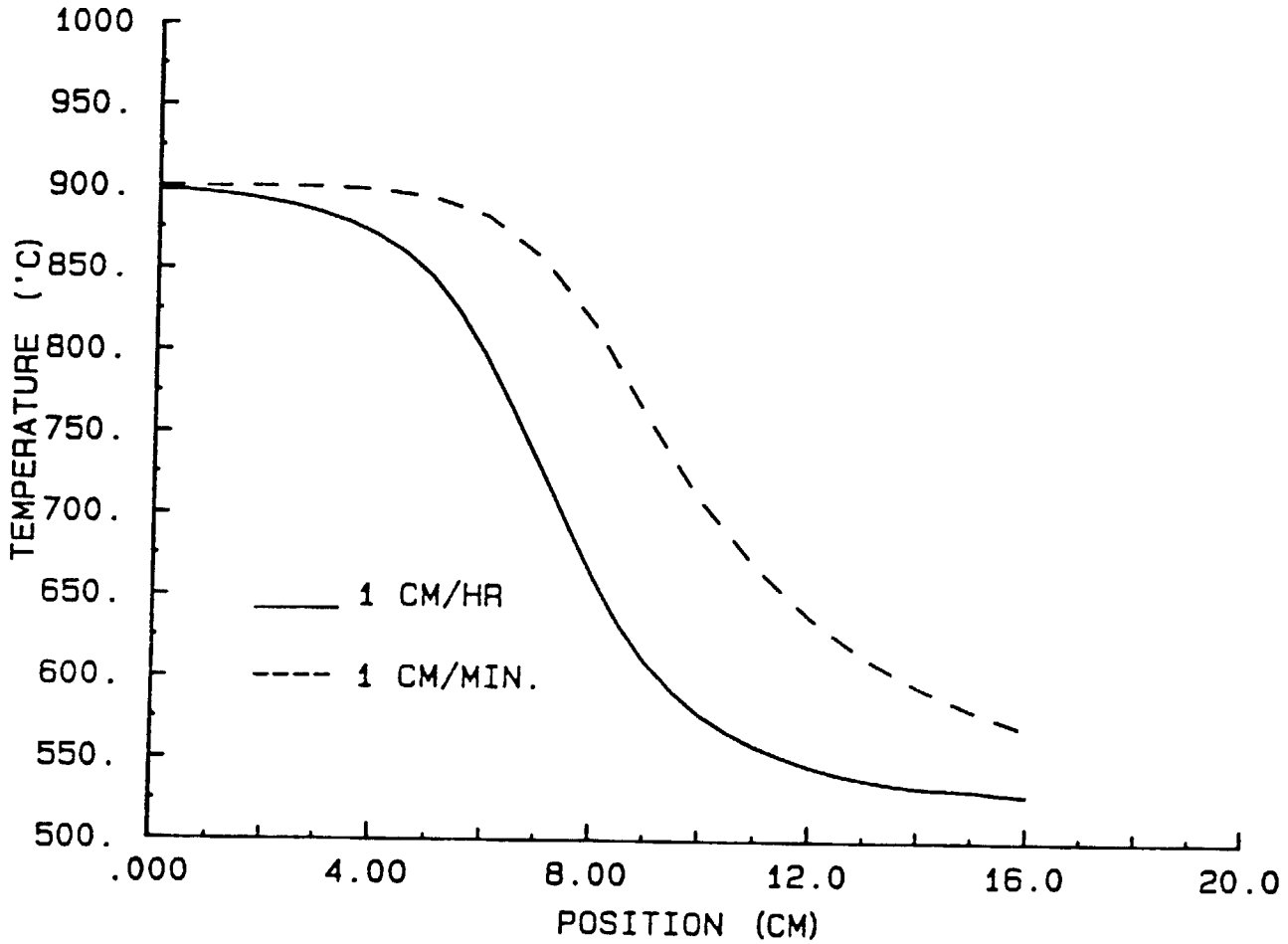

FIGURE 3. The measured temperature profile of the furnace using thermocouples attached to a quartz rod. The hot and cold zones stayed the same; only the pull rate was varied.