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DIRECTIONAL SOLIDIFICATION
AT ULTRA-HIGH THERMAL GRADIENT

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16. Abstract

This report summarizes work at M.I.T. leading to the "HGC" (High Gradient Controlled Solidification) furnace, and presents work conducted under NASA grant to develop the HGC furnace. The HGC furnace comprises a "pancake" shaped hot zone which is continuously fed solid or liquid metal and from which solid metal is continuously withdrawn. The thin "pancake" of liquid metal permits obtaining extremely high thermal gradient while maintaining low metal superheat.

In the course of this program, an HGC furnace was designed and successfully operated to continuously produce aluminum alloys. Over the last several years, many design modifications were made and incorporated to improve its reliability and quality of metal produced, and thermal gradients. Gradients up to 1800°C/cm have been achieved - the highest ever achieved in a continuous or semi-continuous directional solidification apparatus. A recent improtant modification is the complete elimination of rubber "O rings" for the water-cooling chamber, while still maintaining water-cooling directly onto the solidified metal.

An HGC unit has also been designed and operated for high temperature ferrous alloys. The hot zone of this furnace is under vacuum to permit its use for superalloys. Design and operation of this furnace was a final phase of the project research. Successful runs were made with cast iron, at thermal gradients up to 500°C/cm.

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## FINAL REPORT

## DIRECTIONAL SOLIDIFICATION AT ULTRA-HIGH THERMAL GRADIENT

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## Summary

This report summarizes work at M.I.T. leading to the "HGC" (High Gradient Controlled Solidification) furnace, and presents work conducted under NASA grant to develop the HGC furnace. The HGC furnace comprises a "pancake" shaped hot zone which is continuously fed solid or liquid metal and from which solid metal is continuously withdrawn. The thin "pancake" of liquid metal permits obtaining extremely high thermal gradient while maintaining low metal superheat.

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the highest ever achieved in a continuous or semi-continuous directional solidification apparatus. A recent important modification is the complete elimination of rubber "O rings" for the water-cooling chamber, while still maintaining water-cooling directly onto the solidified metal.

An HGC unit has also been designed and operated for high temperature ferrous alloys. The hot zone of this furnace is under vacuum to permit its use for superalloys. Design and operation of this furnace was a final phase of the project research. Successful runs were made with cast iron, at thermal gradients up to 500°C/cm.

## Introduction and Background Research

Beginning in the early 1960's, the writer and his co-workers developed a series of innovative furnaces for directionally solidifying metal under steep thermal gradient. The dual purpose of these designs has been to achieve directional solidification with (1) steep thermal gradient, and (2) minimum convection. Only with these two conditions can homogeneous single crystals or homogeneous "in-situ composites" (off-eutectic multi-phase alloys) be grown from liquid melts of finite size.

## TABLE I

## MAXIMUM THERMAL GRADIENTS ACHIEVED IN ALUMINUM ALLOYS

|  | Year | Maximum Gradient °C/cm |
|---|---|---|
| Typical Boat-Type Crystal Growing Furnace | <1965 | <50 |
| Rinaldi, et al (Al-Cu)[8] | 1972 | 375 |
| Dunn and Flemings (Al-Cu) [9] | 1976 | 825 |
| Neff et al (Al-Cu)[11] | 1977 | 1043 |
| D. S. Lee, This Work | 1978 | 1800 |

Table 1 summarizes the success of this work, using studies on aluminum alloys as the example. Prior to these studies, typical boat-type furnaces achieved perhaps as high as 50°C/cm gradient, and this with much thermal convection.

Over the last decade or more, we have made a series of steps forward and in this work have achieved experimental furnaces with thermal gradients as high as 1800°C/cm, and with vastly reduced thermal convection.

In the mid-1960's, the writer and a co-worker, F. R. Mollard[1,2], concluded that it should be possible to

grow off-eutectic alloys with a plane front provided the
solidification could be caused to occur at high thermal
gradient, G, and low growth velocity, R (i.e., at high G/R).
To accomplish this, they built the first "High G" crystal
growing furnace, Figures 1 and 2. This furnace became the
prototype for many other furnaces subsequently built and
now being used throughout the world by crystal growers
producing many different types of alloys and "in situ
composites" (i.e., off-eutectic multiphase crystals).
Examples of such furnaces are those of Perry, Giamei, Young,
Cline, and Chadwick.[3-7]

$\sim$ Figure 3 is a schematic illustration of the basic
principle of construction of the Mollard-Flemings "High G"
furnace. Heating coils are placed as closely as possible
to cooling coils, and a liquid-solid interface maintained
between the two so as to obtain a high heat throughput across
the liquid-solid interface and therefore a steep thermal
gradient at the interface according to the relation

$$q^* = -kG \qquad\qquad (1)$$

where $q^*$ is heat flux across the interface, $k$ is liquid
thermal conductivity and G is gradient in the liquid at the
liquid-solid interface.

Using this furnace design, gradients, G, as high as 480°C/cm were obtained, the highest ever obtained up to that date.

In later work at M.I.T., in the late 1960's, Rinaldi, Sharp, and Flemings[8], and later Dunn et al[9] employed the Mollard-Flemings principle to construct a furnace for growth of aluminum alloys, a sketch of which is shown in Figure 4. Gradients as high as 825°C/cm were obtained in these studies.

In work at M.I.T. in the mid-1970's sponsored by NASA-Lewis, Neff, Rickinson, Young, and Flemings[10] modified the basic Mollard-Flemings concept to make it applicable to superalloys. This was done by using liquid gallium as the cooling fluid (and water-cooling the gallium). Using gallium as the direct cooling fluid eliminated problems encountered earlier from steam vaporization. Figure 5 is a sketch of that furnace. Gradients in excess of 1000°C/cm were obtained with this unit.

## The HGC Furnace

The "HGC" furnace (high gradient, continuous) was first
designed by Rickinson and built by Neff et al to overcome two
limitations of earlier furnaces. First, it eliminates
a problem that plagued earlier furnaces of excessively
superheating liquid metal when high power was added to the
liquid in order to obtain a high heat throughput and hence
high gradient. It eliminates this problem by reducing the
liquid zone to, in the limit, only a thin film. The second
limitation the HGC furnace overcomes is that earlier designs
could not be continuous.

Figure 6 shows the principle schematically. A more
detailed schematic, Figure 7, shows a heat source located
directly above a solidifying in-situ composite. The solid-
liquid interface is maintained at a fixed position while
the composite is withdrawn. The first furnace utilizing
this concept, constructed for aluminum alloys, is shown in
Figure 8. Gradients in excess of 1000°C/cm were obtained
with this design. A modification of this design subsequently
developed by D. Lee reached the highest thermal gradient yet
achieved for aluminum, 1800°C/cm (Table 1). He reached this
gradient while keeping maximum temperature in the liquid

below 900°C. A schematic illustration of that design is
shown in Figure 9.

## Final Design, HGC Unit for Aluminum Alloys

The design shown in Figure 9 proved to be excellent
for achieving high thermal gradients but had one significant
disadvantage - the "O" ring required to contain the water
had a short life and leaked after only a few centimeters of
material were produced, when very high gradients were employed.

A major step forward was taken in the course of this
work in completely eliminating the "O" ring and the associated
water leakage. This was done by taking the two important
steps shown schematically in Figures 10 and 11. First, the
"chilling chamber" (where water contacts the solidified ingot)
had previously been kept full of water and fast flow velocities
used to minimize vapor formation and associated pressure
buildup. However, especially with the higher melting point
metals such as aluminum and above, some vapor formation
cannot be prevented and it is the pressure "peaks" that
result from such formation that cause water leakage into
the hot zone with disastrous results.

The first part of the new development was to reduce the pressure within the chilling chamber by pumping the water (or water-steam mixture) out of the chamber under slightly reduced pressure. At sufficiently low pressure, the chamber is probably not completely full of water but probably comprises a strong water jet with some steam formation as shown schematically in Figure 10. The important aspect is that the pressure in the chilling chamber must not exceed atmospheric pressure so that water or steam cannot pass upward to the hot zone.

Once the above condition was met experimentally, the "O" ring was found to be unnecessary and in its place was put several spring steel O-rings with locking spring to keep it in place (Figure 11). These modifications have now been made and incorporated in the HGC unit for aluminum alloys. Water leakage has been eliminated, as has the "O" ring life problem. Design of the overall apparatus is shown in Figure 12.

## Experimental Results - Aluminum HGC

Five experiments have now been made using Off-eutectic Al-Cu alloy to test the modified directional

solidification apparatus. Figure 13 shows the result of one such experiment, in which 7.4 cm of material was grown.

All experimental difficulties in producing continuous sound lengths of aluminum under steep thermal gradient now appear to be overcome.

For some time, the remaining problem appeared to be that there was some convection resulting from electromagnetic stirring that is influencing the structure. Finally, this problem has been resolved by use of a thicker susceptor (7/16") and a thinner melt (1/16") in the hot zone than those in the previous experiments. Figures 14 and 15 show a quenched interface and a grown lamellar structure of Al-31.5 wt%Cu, respectively. The process utilizes a 20 KW - 10 $KH_z$ RFC inverter unit as an induction power source, with the power used in these experiments ranging from 80 to 85% of the maximum available 20 KW.

## High Temperature HGC

A high temperature, atmosphere-controlled HGC has been built and tested with Fe-based alloys. This furnace

is shown in Figure 16 and schematically in Figure 17.
A 20 KW, 10 kHz inverter induction source couples directly
to a 1 cm thick, 7 cm diameter liquid metal disk in an
alumina crucible. A solid metal rod is fed continuously
during growth into the liquid pool through a water-cooled
vacuum seal. Inert gas is fed in along the feed rod
periphery to prevent melting before it enters the liquid
region shown in Figure 17.

Solidification occurs as shown in Figure 8 in a
12 mm diameter section of the alumina mold. The interface
is maintained at a constant level within the alumina mold,
while solid rod is continuously withdrawn down through the
chill. The liquified metal is entirely under vacuum to
permit this furnace to be used for superalloys.

Note the chilling arrangement here is different from
that of the aluminum HGC. Cooling is within a thin graphite
mold. The graphite mold is cooled to a location as close as
possible to the liquid-solid interface by liquid gallium.
The liquid gallium is cooled by water.

Initial experiments performed using this apparatus
have been successful. One example is shown in Figure 13 and

19 wherein 9 cm of white cast iron of approximately eutectic composition were grown with a temperature gradient of 500°C/cm. This apparatus operates smoothly and efficiently and it is presumed that very high thermal gradients could be reached by incorporating the water-chill design of Figure 10.

## References

1. F. R. Mollard, M. C. Flemings, "Growth of Composites from the Melt, Part I", Trans. Met. Soc., AIME, v. 239, 1967, pp. 1620-1625.

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7. G. A. Chadwick, I.S.I. Publ., 100, 1968, p. 138.

8. M. D. Rinaldi, R. M. Sharp, M. C. Flemings, "Growth of Ternary Composites from the Melt, Part I and Part II", Met. Trans., v. 3, 1972, pp. 3133-3148.

9. E. M. Dunn, R. A. Wasson, K. P. Young, M. C. Flemings, "Growth of In-Situ Composites of Al-Cu-Ni Alloys", Conference on In-Situ Composites-II, Xerox Corporation, Lexington, Massachusetts, 1976.

10. M. A. Neff, B. A. Rickinson, K. P. Young, M. C. Flemings, "The Growth and Morphology of Directionally Solidified Nickel-Based $\gamma/\gamma'-\delta$ Superalloys", Met. Trans. B, v. 9B, September, 1978, pp. 469-476.

11. NASA Tech Brief

## APPENDIX

## LIST OF PUBLISHED PAPERS, THESES, AND TECHNICAL
## PRESENTATIONS GIVEN ON WORK SUPPORTED BY THIS CONTRACT

### Published Papers

1.  M. A. Neff, B. A. Rickinson, K. P. Young, M. C. Flemings, "The Growth and Morphology of Directionally Solidified Nickel-Based $\gamma/\gamma'-\delta$ Superalloys", Met. Trans. B, v. 9B, September, 1978, pp. 469-476.

2.  M. C. Flemings, D. S. Lee, M. A. Neff, K. P. Young, B. A. Rickinson, "A New Furnace for High Gradient Directional Growth", Conference on In-Situ Composite - III, Ed. J. L. Walter, et al, Ginn Custom Publishing Co., Lexington, Mass., 1979, pp. 69-77.

3.  NASA Tech Brief

### Theses

1.  M. A. Neff, S.B.-S.M., "The Morphology of Directionally Solidified Nickel-Based $\gamma/\gamma'-\delta$ Superalloys.

2.  R. Ewasko, S.B., "The Morphology of Directionally Solidified Nickel-Based $\gamma/\gamma'-\alpha$ Superalloys.

3.  D. S. Lee, Sc.D. (in progress), "Development of a High Gradient Continuous Caster (HGC) for Directional Solidification of Al-Cu Alloys".

4.  M. A. Neff, Sc.D. (in progress), "Development of a High Gradient Continuous Casting Process for Nickel-Based Eutectic Composites".

### Oral Presentations

1.  TMS-AIME Fall Meeting, 1977, "The Morphology of Directionally Solidified $\gamma/\gamma'-\delta$ Superalloys", by M. A. Neff in the High Temperature Materials Session.

2.  NASA, January, 1980, "Furnace Requirements for High Gradient Processing", by M. C. Flemings in Lecture Series, Materials Processing in Space, Houston, Texas.

**TO DRIVE MECHANISM**

SAMPLE

TRANSITE

CONSTANT
VOLTAGE
SOURCE

STAINLESS
STEEL TUBE

LAVA SLEEVE

CHROMEL "A"
WINDING

COPPER CHILL

WATER
PUMP &
HEAT
EXCHANGER

O-RINGS

TO TEMPERATURE
RECORDER

SQUARE
SHAFT

BALL BEARING
ASSEMBLY
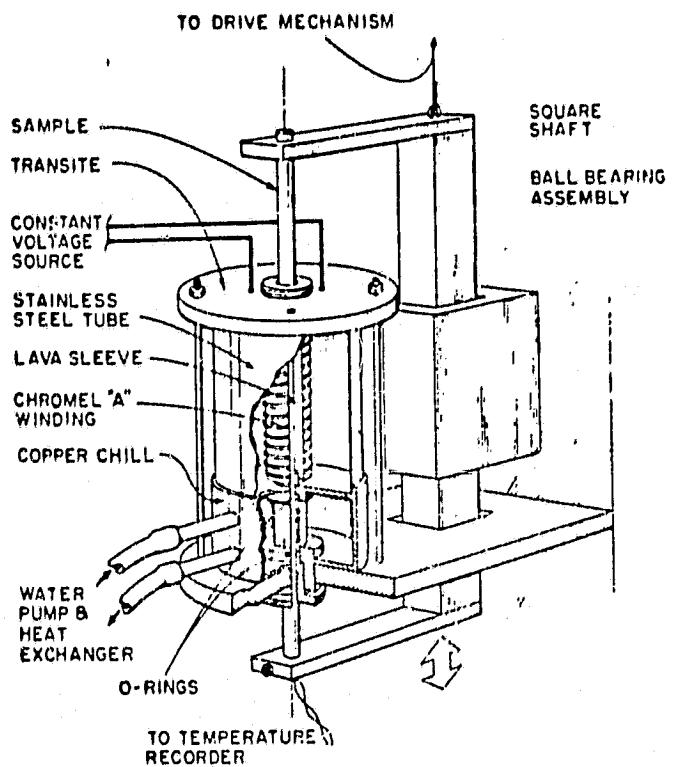
Figure 1.   Overall view of high thermal gradient furnace
first built by Mollard.(ref. 2)
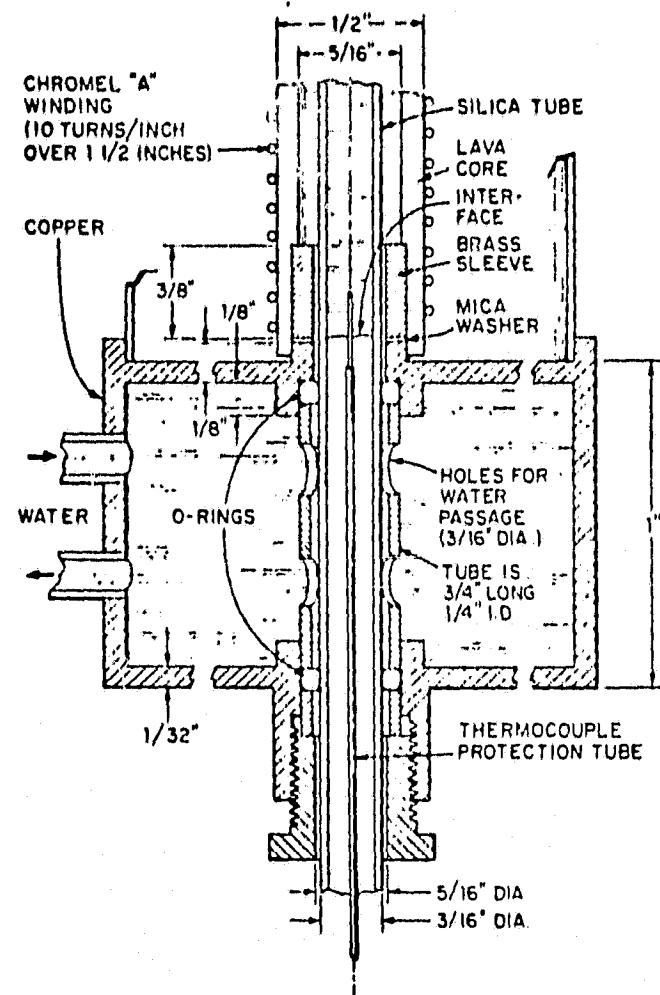
Figure 2.   Central portion of high gradient furnace
built by Mollard.(ref. 2)
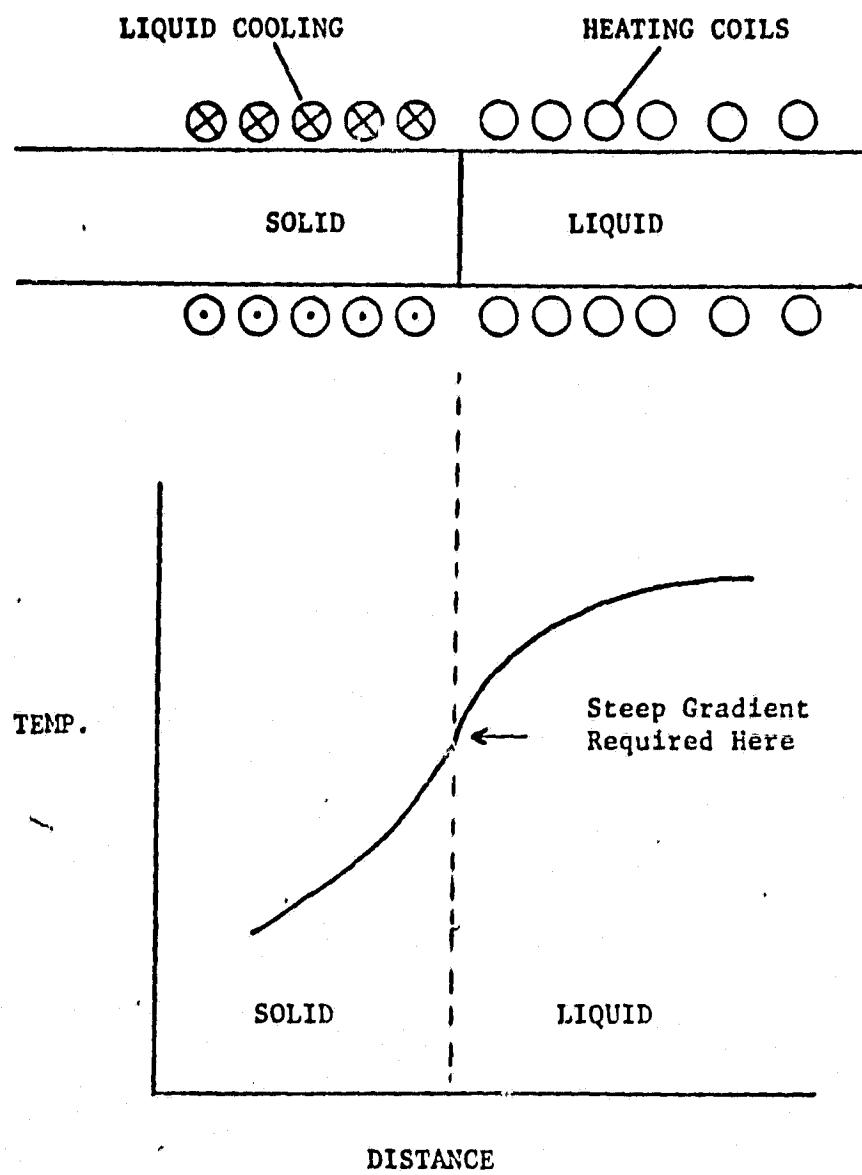
Figure 3. Schematic illustration of principles of "High G" Furnaces.
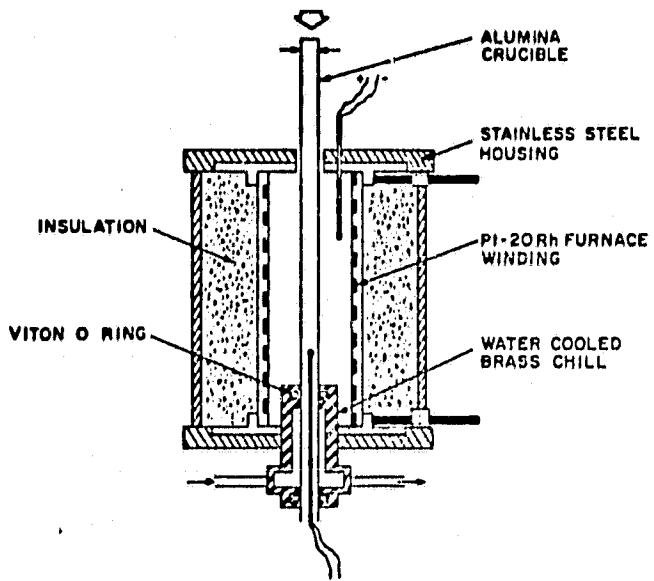
**Figure 4.** Schematic view of Rinaldi furnace for high gradient directional growth. (Ref. 8)
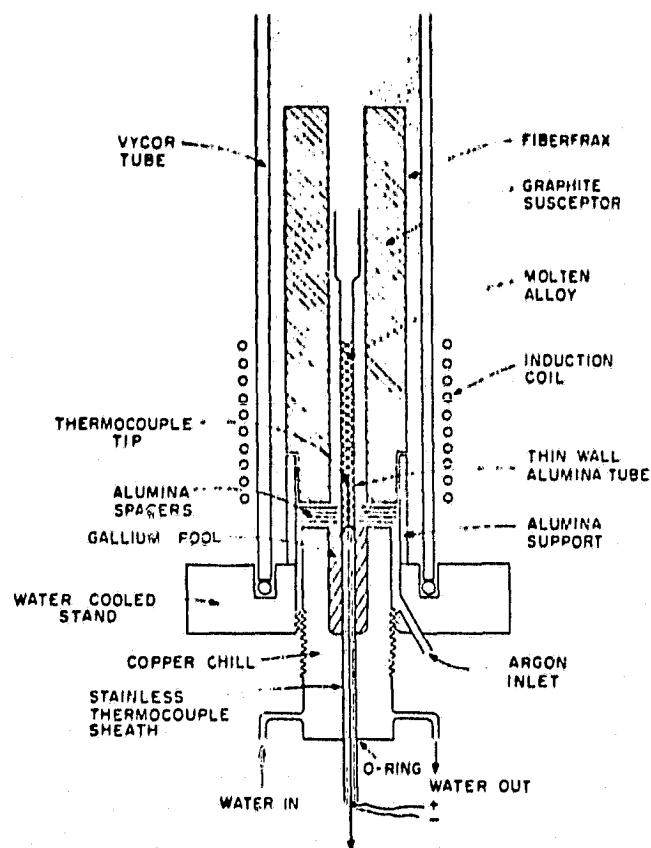
Figure 5. High gradient furnace built by Neff et al
for directional solidification of high temperature
materials. This was the first furnace to use gallium
cooling. (ref. 10)

HEAT
INPUT

THIN LIQUID
ZONE

SOL'D OR
LIQUID FEED

S-L INTERFACE

COOLING

SPECIMEN
WITHDRAWAL
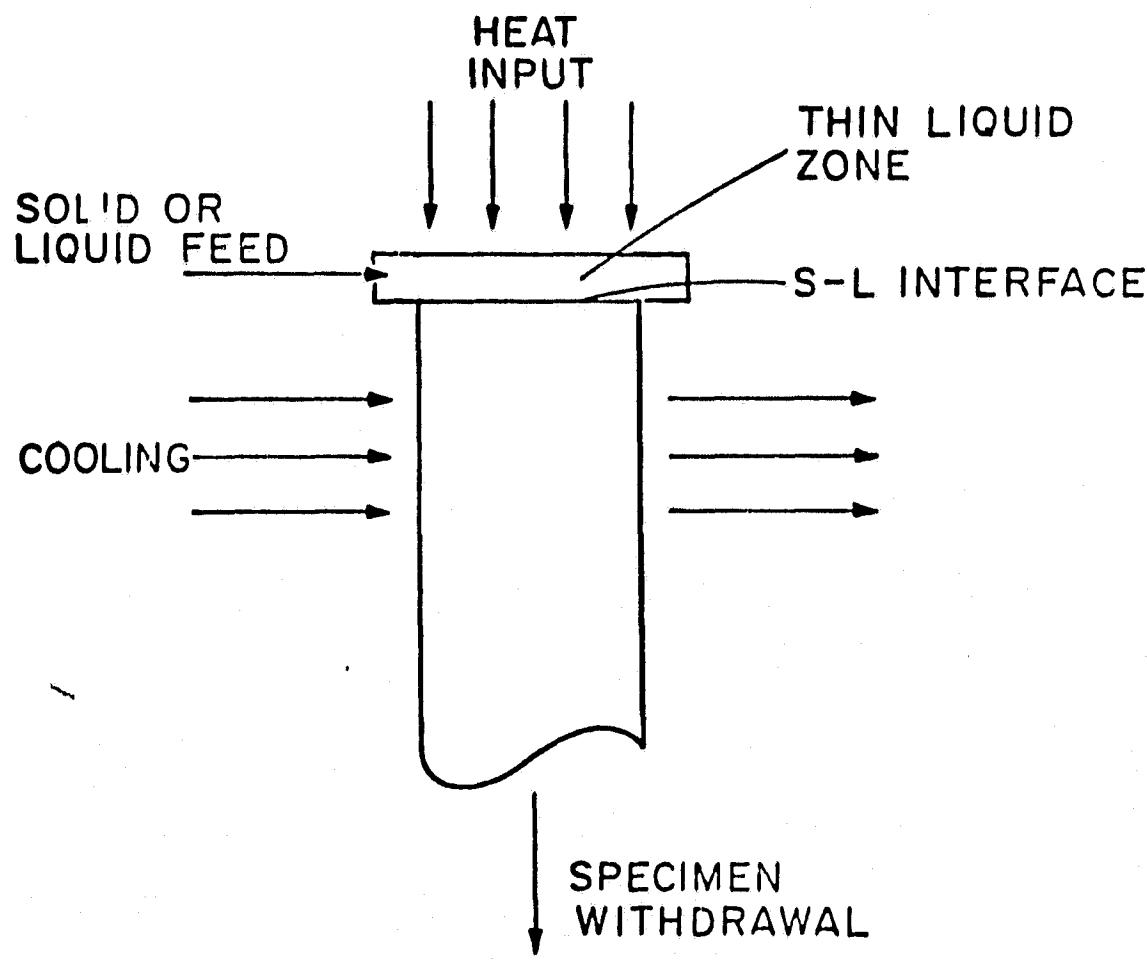
Figure 6.   Schematic illustration of principles
of "HGC" Furnaces.

INDUCTION HEAT SOURCE

RESISTANCE
HEATING

HEAT SINK

A – ALLOY POOL
B – GRAPHITE SUSCEPTOR
C – CERAMIC MOLD
D – LIQUID-SOLID INTERFACE
E – FLOWING WATER
F – STEEL SHELL
G – INSULATION

SPECIMEN
REMOVAL
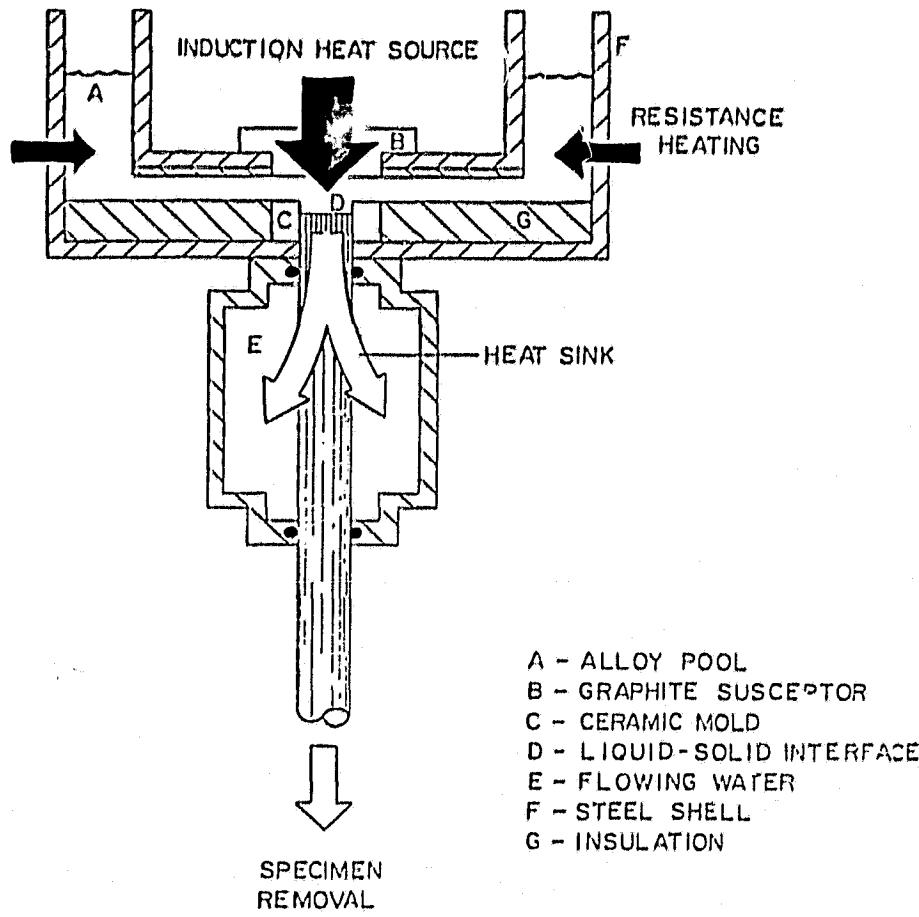
Figure 7.   Schematic illustration of the
HGC design.
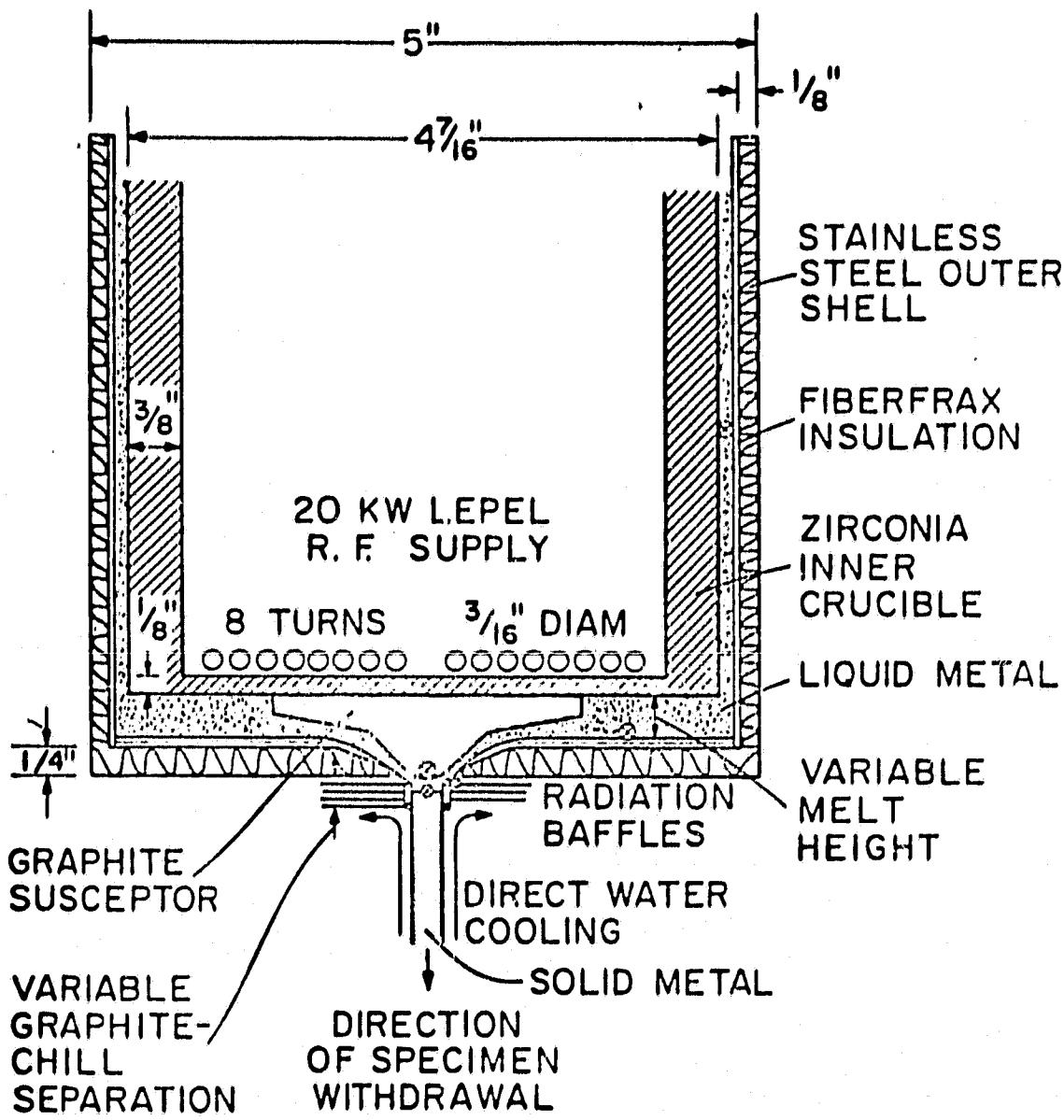
Figure 8.   Liquid metal feed HGC tested for aluminum alloys.
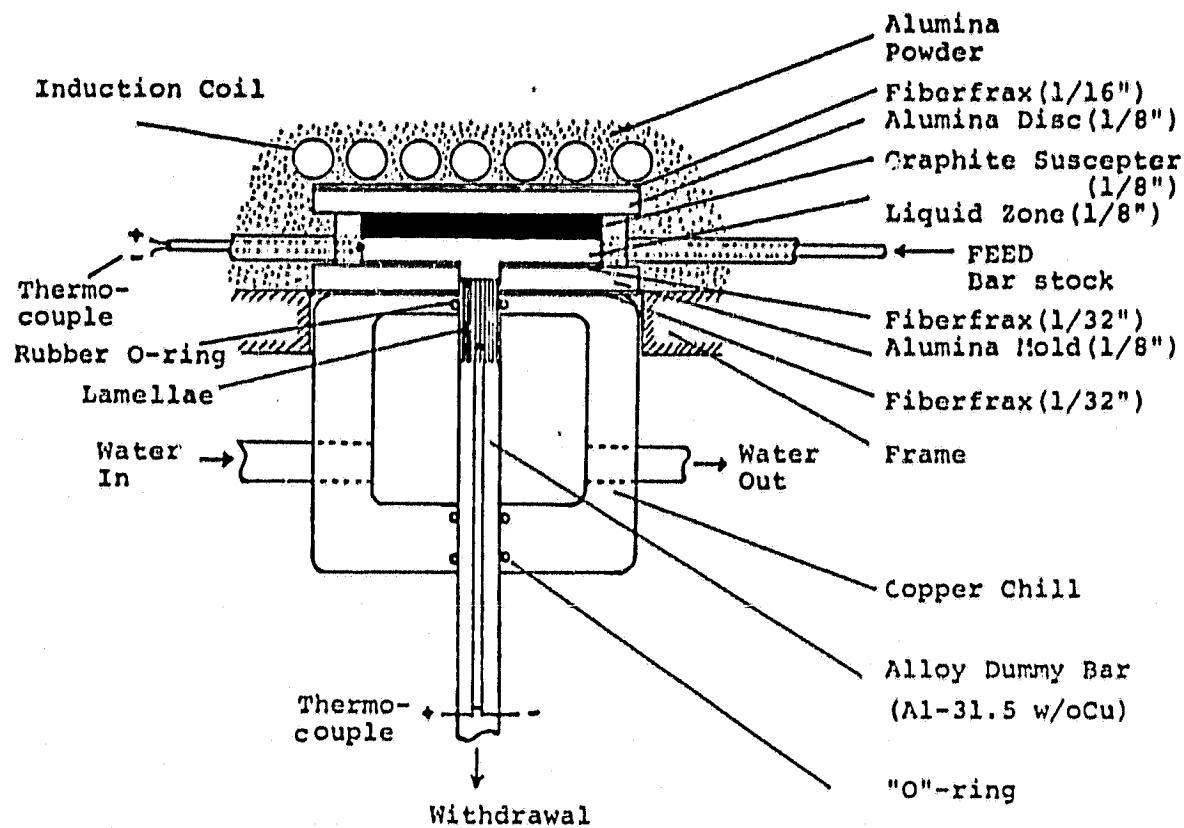
Figure 9.  HGC Furnace for aluminum employing solid feed.
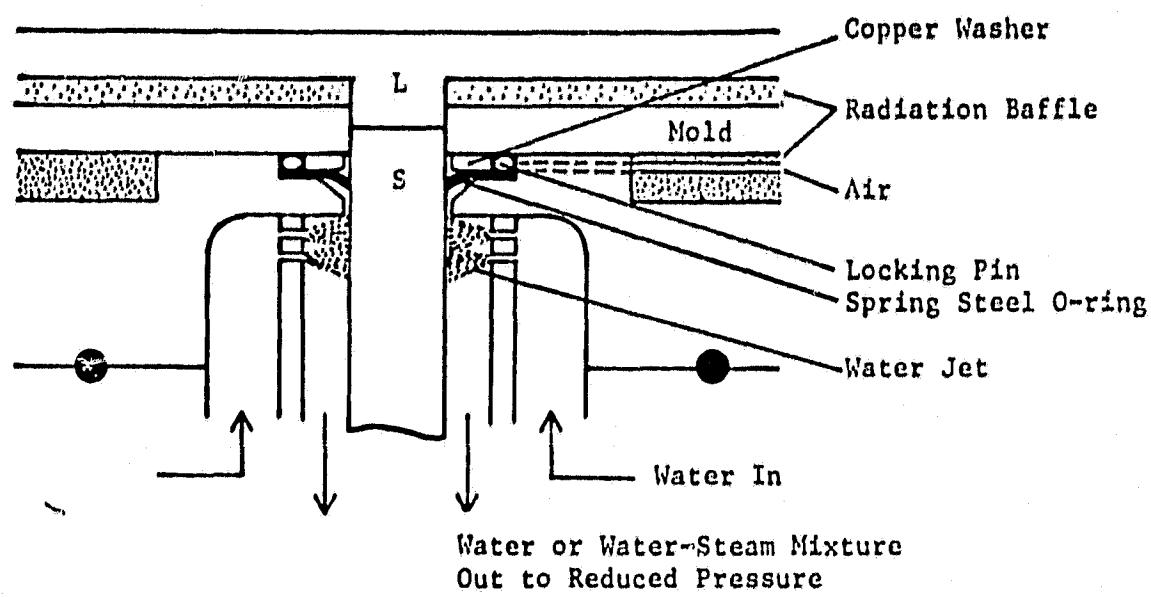
Figure 10.  New Chilling Chamber Design

A:

B:

Slit

Small Hole
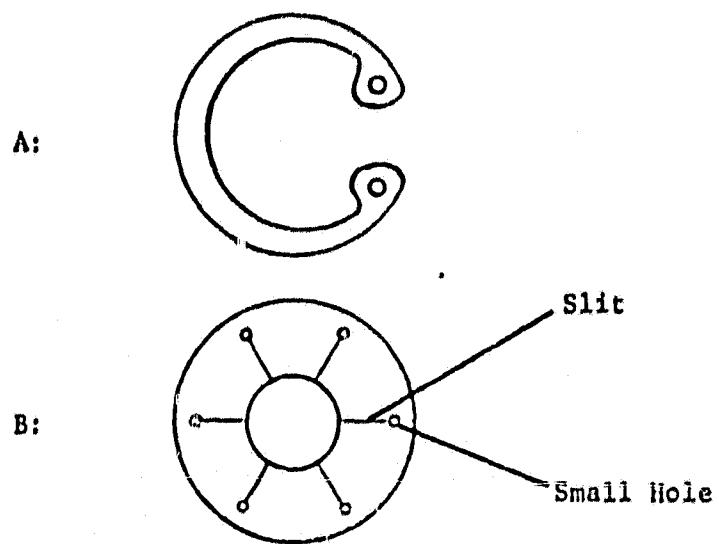
Figure 11.   A:   Locking pin of spring steel O-ring (thickness = 0.035").

B:   Spring steel O-ring; ID = 0.24", OD = 0.625",
thickness = 0.007".
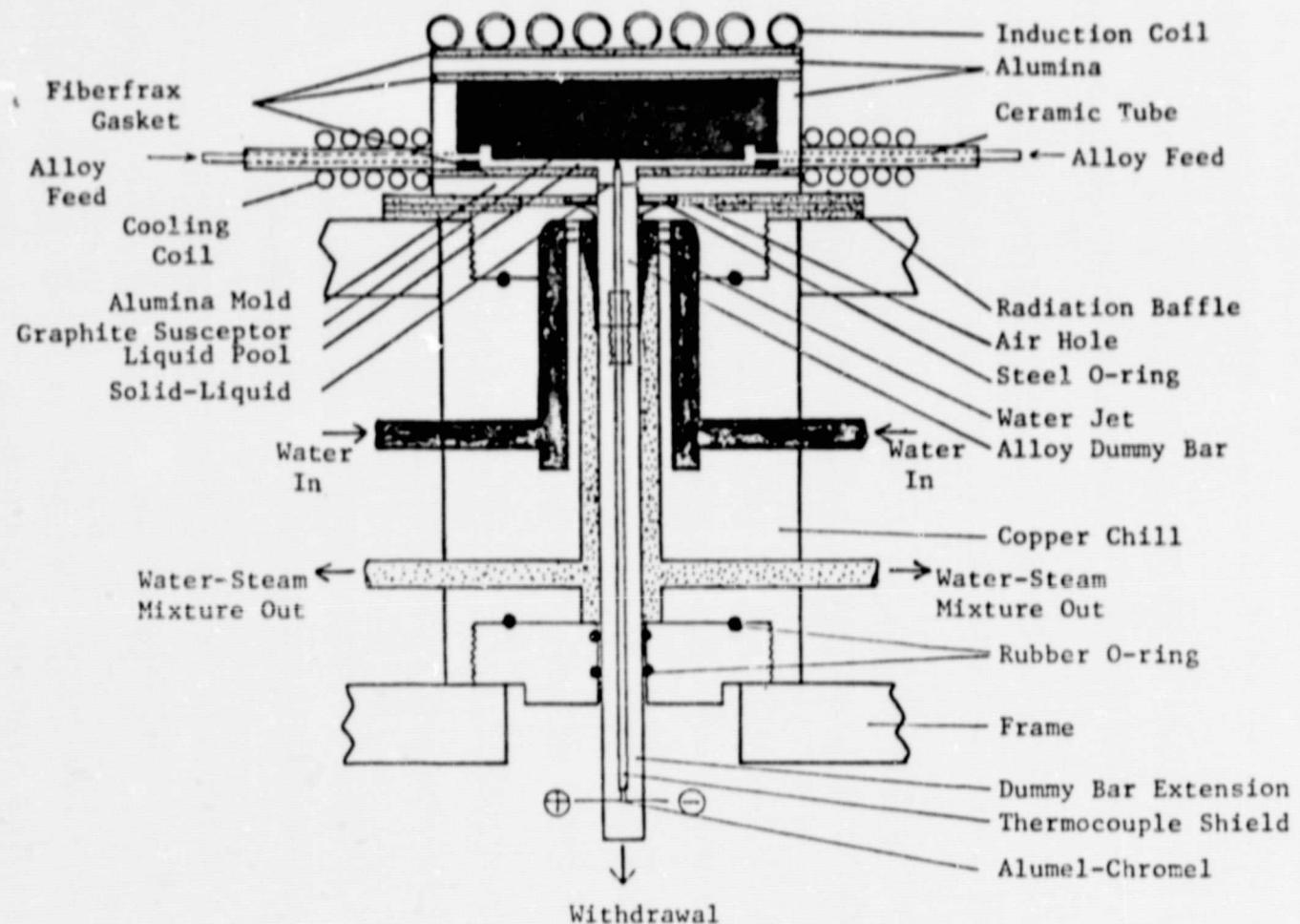
Figure 12. Schematic illustration of the final HGC Furnace.
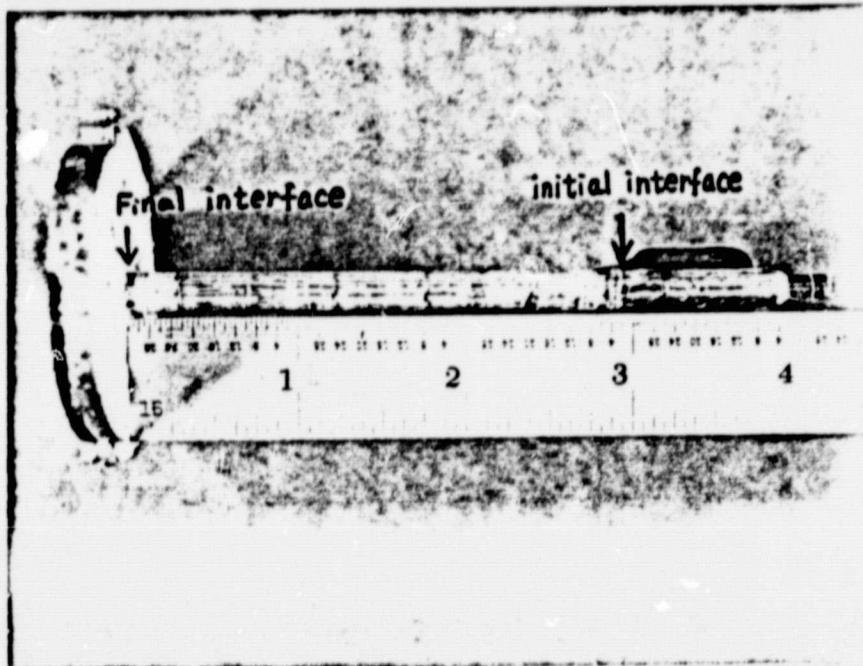
Figure 13. Al-24 wt % Cu specimen directionally grown at various growth rates ranging from 1"/hr to 9.1"/hr. (Run No. 43; G = 913°C/cm.)
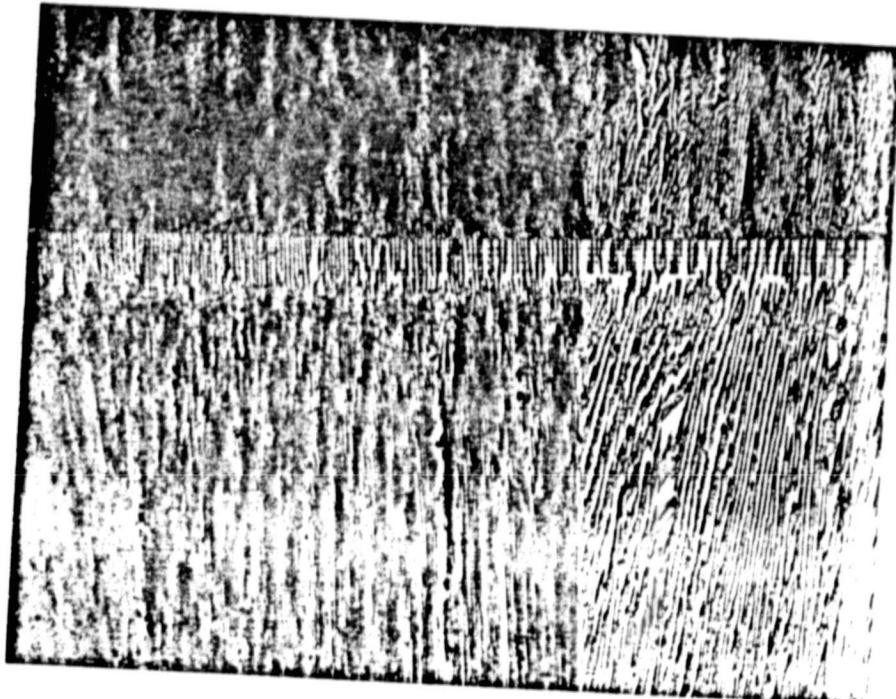
Figure 14.  Planar interface of the lamellar structure observed
at the end of directional growth of Al-31.5 wt% Cu:
calculated $G \simeq 800°C/cm$, $R = 4.7$ cm/hr, X80.
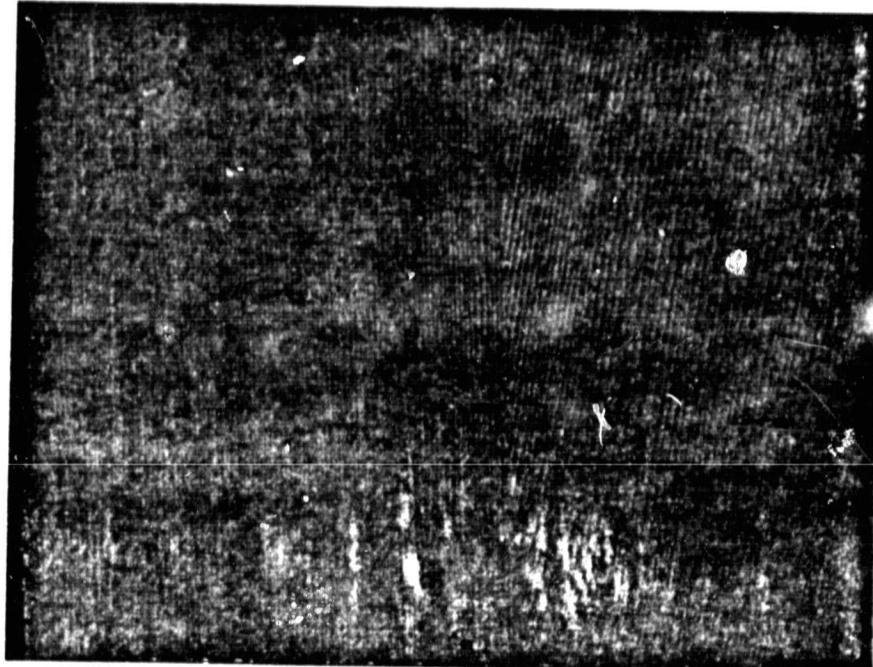
Figure 15.  Microphotograph of a lamellar structure of
A1-31.5 wt% Cu:  G ≃ 800°C/cm, R = 4.7 cm/hr,
$\lambda$ = 2.8μ.  X256.
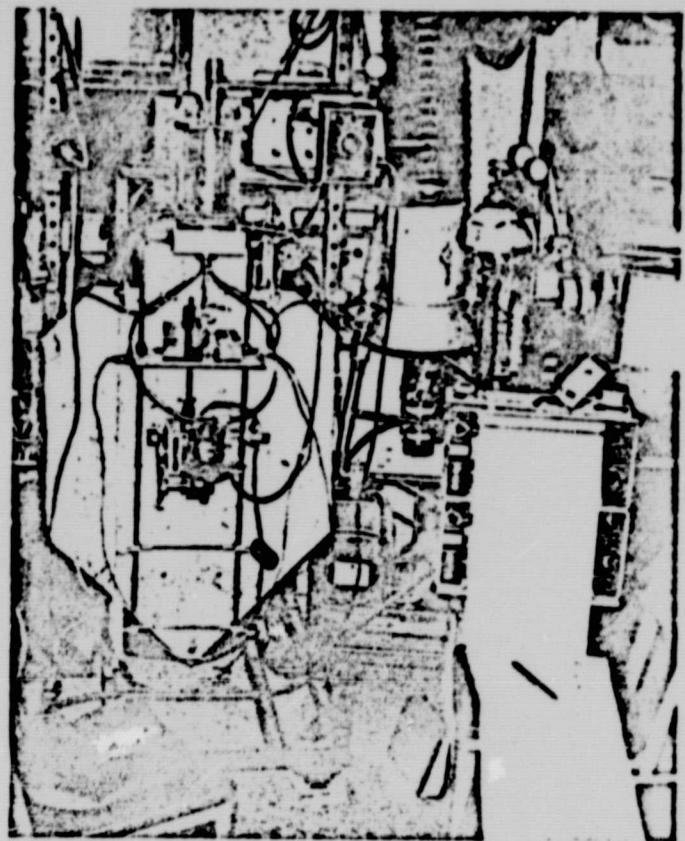
Figure 16: High-temperature HGC in the laboratory.
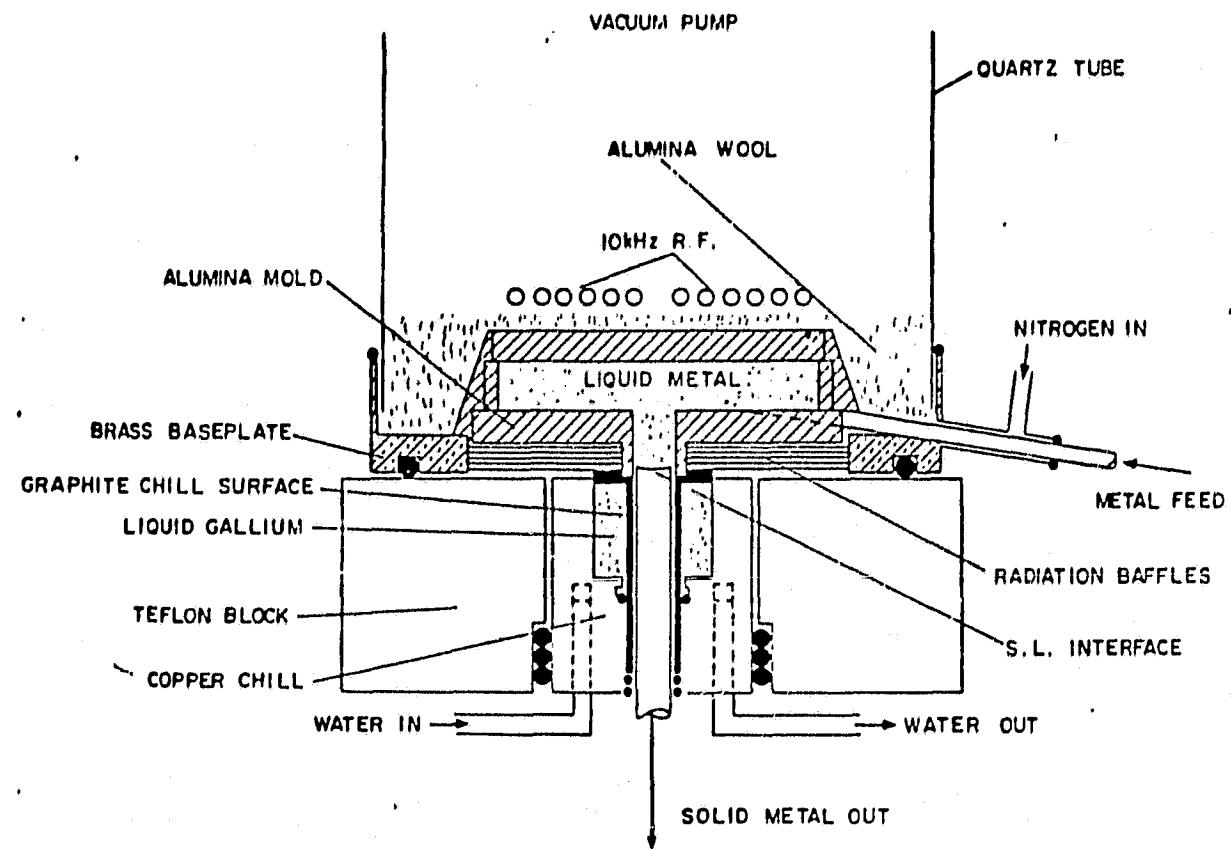
**Figure 17.** Schematic diagram of the high temperature furnace.
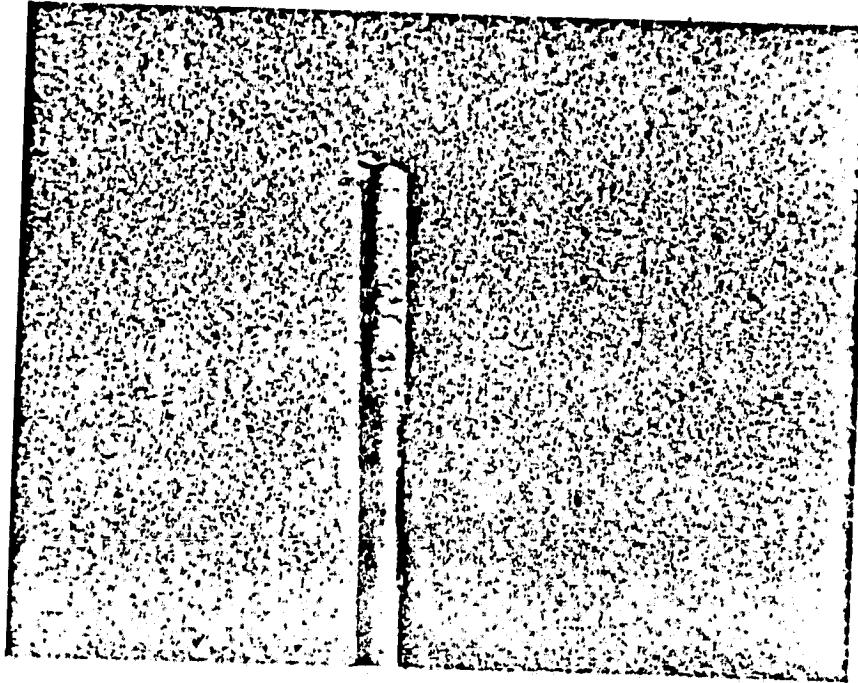
Figure 18: Iron casting produced at 1 cm/hr in a 400°C/cm
thermal gradient. Length of growth section = 9 cm.
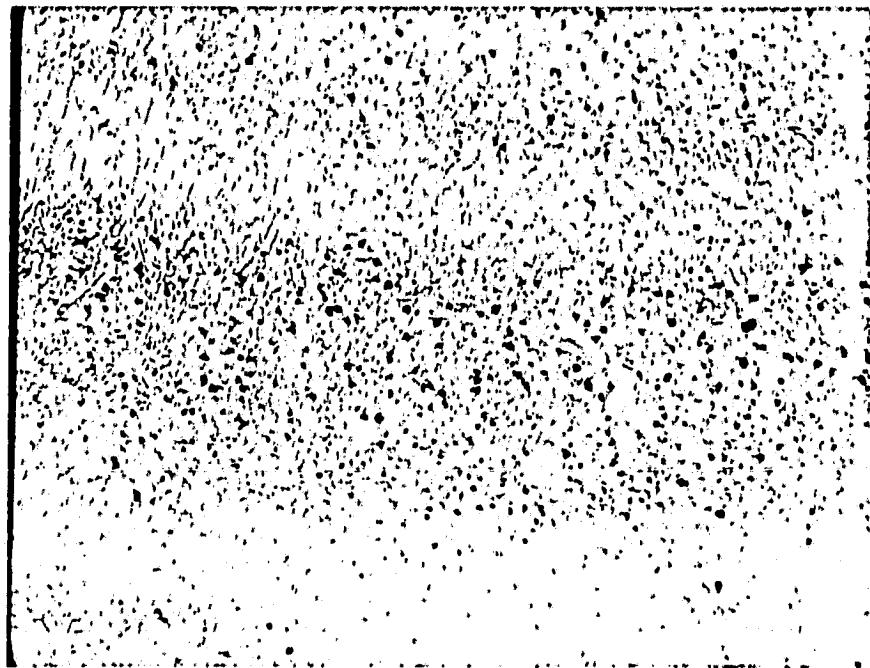
$1000 \mu m$

Figure 19: Microstructure of iron casting showing formation of white iron and carbide precipitation.