Marangoni Convection of Molten Silicon under Microgravity

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1. Introduction

One of the important phenomena for a heat and mass transport process during melt crystal growth is Marangoni flow, which takes place on the surface of the melt. Marangoni flow might be useful in discussing the formation of the network pattern[1) and asymmetric temperature distribution[2) in the Czochralski melt, but the Marangoni flow of molten silicon has not yet been clearly explained due to the difficulty of performing experiments. For example, the existence of a strong buoyancy force on earth makes it difficult to distinguish Marangoni flow from buoyancy flow. The temperature oscillation due to Marangoni convection in a silicon melt column was precisely measured under microgravity on board the rocket TR-IA #4 launched by the National Space Development Agency of Japan (NASDA).

2. Experimental

A molten silicon column 10 mm in diameter and 10 mm high was formed under microgravity using carbon rods, as shown in Fig. 1. The specimen was set into a silica glass ampoule through which s-grade argon gas was flowed to prevent oxidization of the sample. The specimen was heated in an infrared image furnace, High Temperature Furnace Type II (HTF-II), as shown in Fig.2. Two sets of thermocouples were set near the upper and lower solid-liquid interfaces to monitor the temperature difference between the two interfaces. Four sets of thermocouples 0.1 mm in diameter within a quartz glass sheath 1.0 mm in diameter were immersed into the silicon melt 2 mm above the lower solid-liquid interface at the periphery of the melt to measure temperature oscillation. The temperature sampling speed was 40Hz. The melt shape and surface oscillation were recorded using a video camera. Microgravity of $10^{-4}g_0$ was obtained for six minutes on board the NASDA TR-IA #4 rocket launched on August 25, 1995 from Tanegashima Space Center of NASDA.

3. Results and Discussion

Figure 3 shows the temperature oscillation detected by the thermocouples in the silicon melt column (T3-T6). Note that the temperature measured in the solid part did not show any oscillation (T1-T2). Figures 4a and 4b show the results of Fourier-transform analysis of the oscillation frequency. A particular frequency of 0.1 Hz was observed while the column was formed. After the column had finished forming, the temperature oscillation did not show any particular frequency. These results suggest that the flow mode is periodic-oscillatory during column formation and non-periodic-oscillatory after formation of the liquid column. In order to explain the flow mode of molten silicon, one must discuss the temperature coefficient of surface tension for molten silicon, i.e. the driving force for the Marangoni flow. As reported recently, the temperature coefficient of surface tension of molten silicon shows various values depending on the oxygen partial pressure of the ambient atmosphere, suggesting that it would depend on oxygen concentration of the melt[3-5]. Oxygen concentration analyzed for a flight sample was 7 x 10^{18}atoms/cm^3, which was one order of magnitude higher than those measured for samples processed in a containerless condition or under finely controlled oxygen partial pressure[4,5]. The high measured oxygen concentration was probably due to the use of a quartz sheath for the thermocouples which touched the silicon melt column as this would have dissolved slightly into molten silicon. Although the relationship between temperature coefficient and oxygen concentration has not yet been quantitatively clarified for samples with such a high concentration of oxygen, a flight sample probably has a rather small temperature coefficient. However, the Marangoni number for the present experimental system would have been sufficiently larger than the critical value of 100-200[6] for the steady to periodic-oscillatory flow and also larger than the expected critical value for the periodic- to non-periodic-oscillatory flow.

1) H. Yamagishi and I. Fusegawa: J. Jpn. Assoc. Crystal Growth, 17 (1990) 304.
2) K.-W. Yi, K. Kakimoto, M. Eguchi, M. Watanabe, T. Shyo and T. Hibiya: J. Crystal Growth 144 (1994) 20.
3) B. J. Keene: Surface Interface Anal., 10 (1987) 367.
4) M. Przyborowski, T. Hibiya, M. Eguchi, I. Egry: J. Crystal Growth, 151 (1995) 60.
5) Z.-G. Niu, K. Mukai, Y. Shiraishi, T. Hibiya and K. Kakimoto: 4th Asian Thermophysical Properties Conf., Tokyo, September 1995, Abstract, pp.73.
6) A. Cröll, W. Muller-Sebert and R.Nitsche: Matar. Res. Bull., 24 ,(1989) 995.

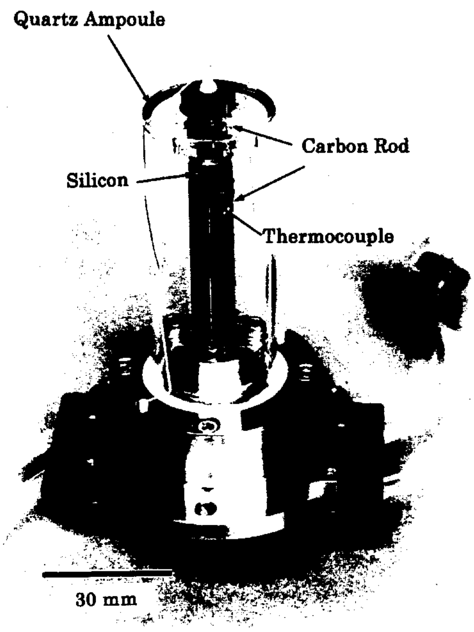

Figure 1 Quartz ampoule containing silicon sample, carbon rods and thermocouples.

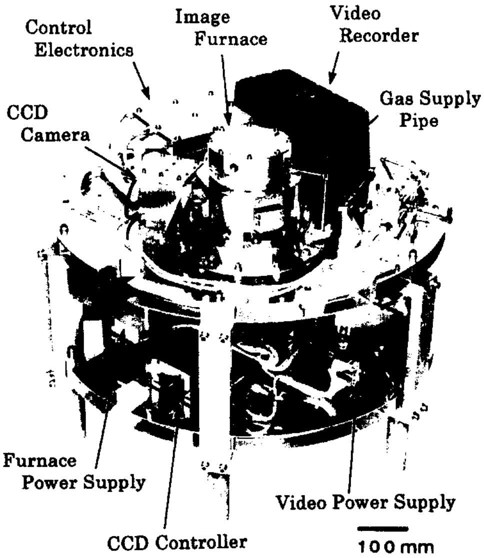

Figure 2 High Temperature Furnace Type Ⅱ (HTF-Ⅱ).

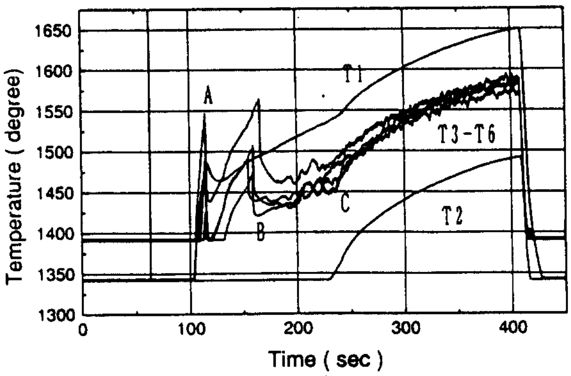

Figure 3 Temperature oscillation detected by thermocouples in the molten silicon column (T3 - T6) and at the solid-liquid interface (T1-T2). At point A, input power was changed from 1300W to 900W. Thermocouples touched the melt at point B. The molten silicon column was completely formed at point C.

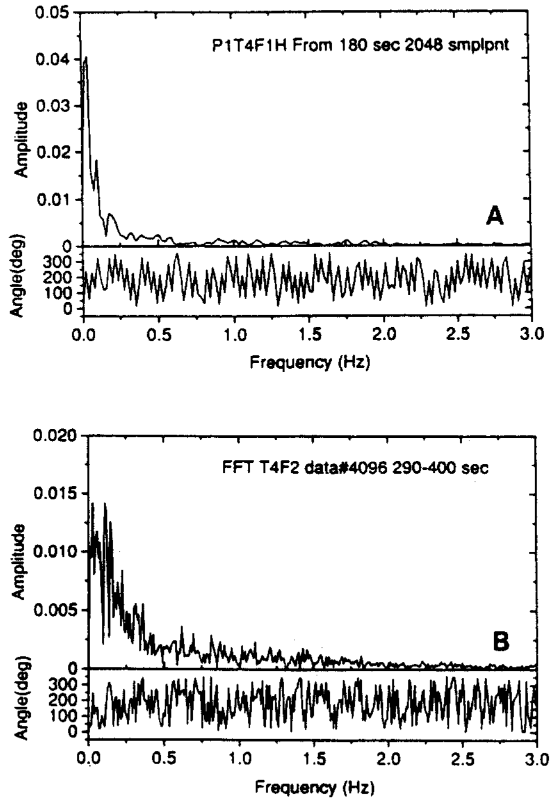

Figure 4 Power spectrum for temperature oscillation (a) during and (b) after column formation. Note that the scales of the ordinate are magnified in (b). Peak at 0.05 Hz cannot be judged if it really exists or not.

미소중력 상태의 용융 실리콘에서 일어나는 마랑고니 대류

日比谷 孟俊
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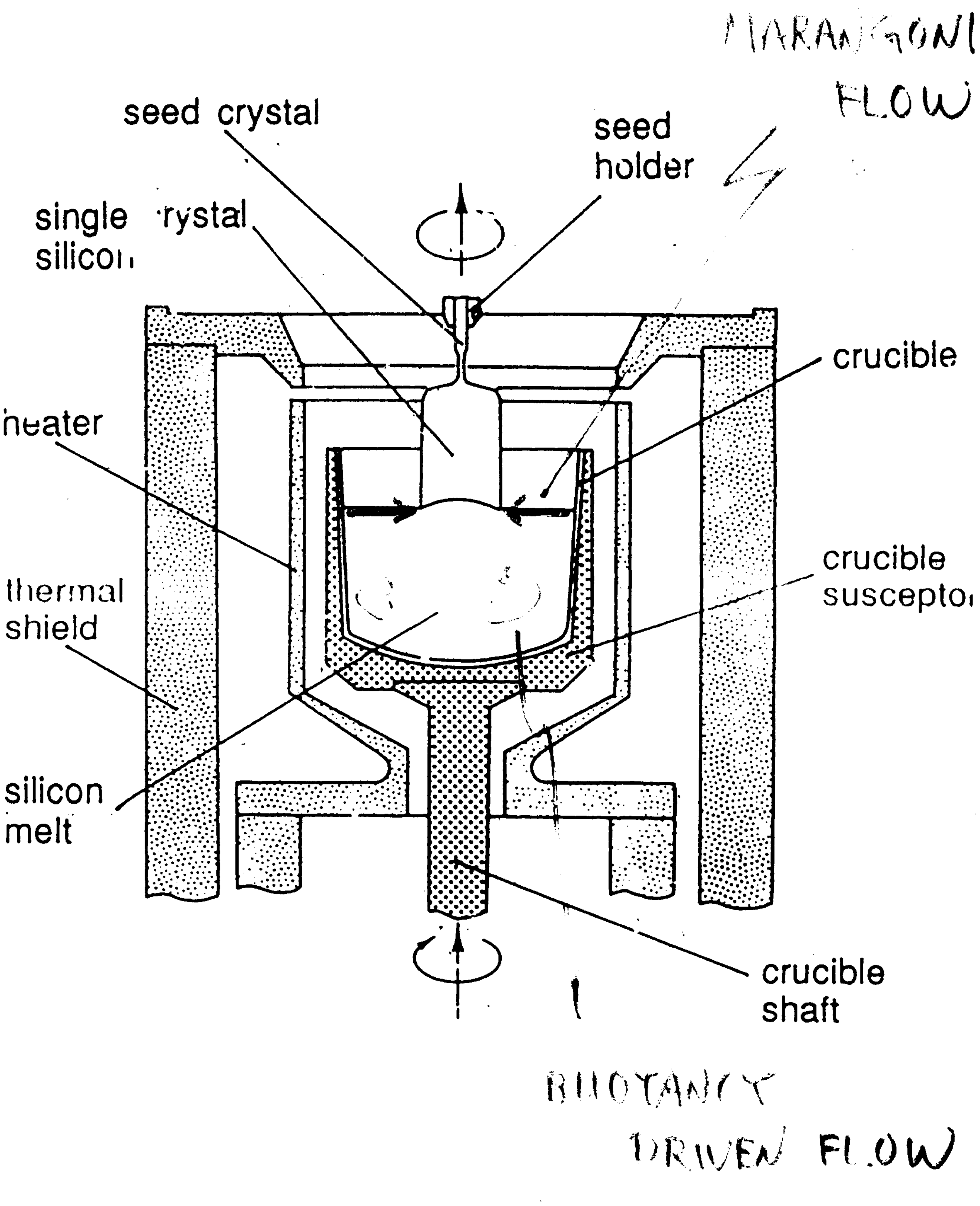
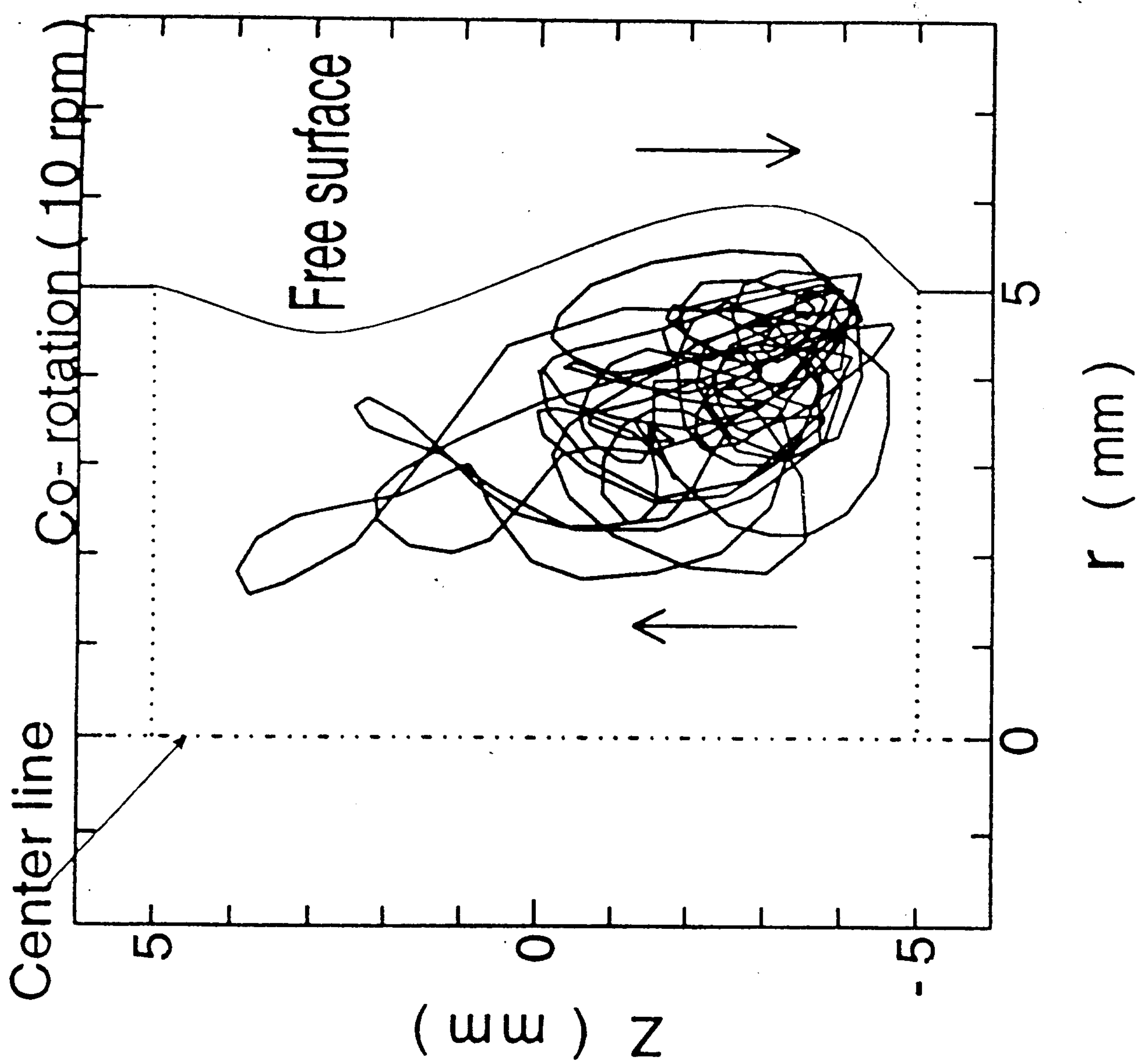
- CRYSTAL GROWTH AND MARANGONI FLOW
- PROBLEMS TO BE SOLVED
- APPARATUS AND SAMPLE
- RESULTS: ROCKET, EARTH
- DISCUSSION
- CONCLUSION

LESS INFORMATION ON MARANGONI FLOW OF MOLTEN SILICON

- **Mac1 for**
  "STEADY TO PERIODIC-OSCILLATORY FLOW"
  → 100 - 200
- **Mac2 for**
  "PERIODIC TO NON-PERIODIC-OSCILLATORY FLOW" → ?
- **EFFECT OF DEFORMATION**
  STRAIGHT OR WAIST-SHAPED ?
- **EFFECT OF HEAT TRANSFER THROUGH**
  LIQUID/GAS-INTERFACE ?

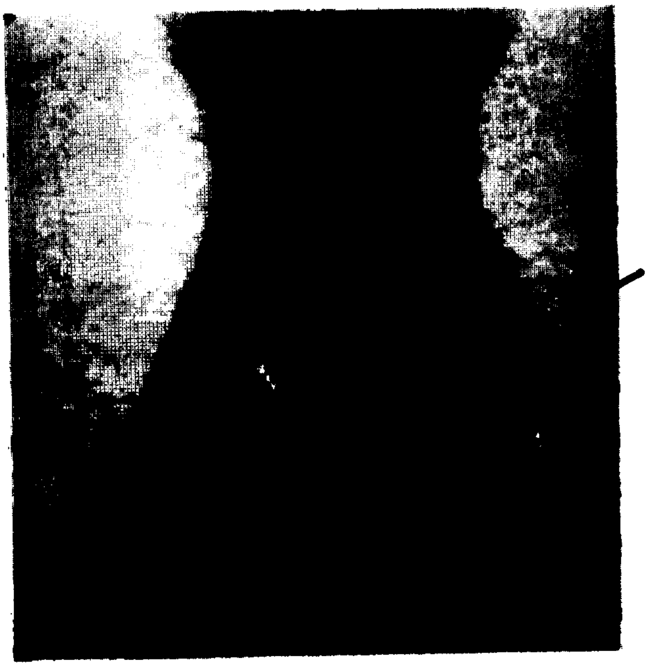

MARANGONI
FLOW
seed crystal
seed
holder
single crystal
silicon
crucible
heater
crucible
susceptor
thermal
shield
silicon
melt
crucible
shaft
BUOYANCY
DRIVEN FLOW



Co- rotation (10 rpm)
Free surface
Center line
r (mm)
Z (mm)
5
0
-5
5
0

FZシリコンメルト対流可視化

0秒後



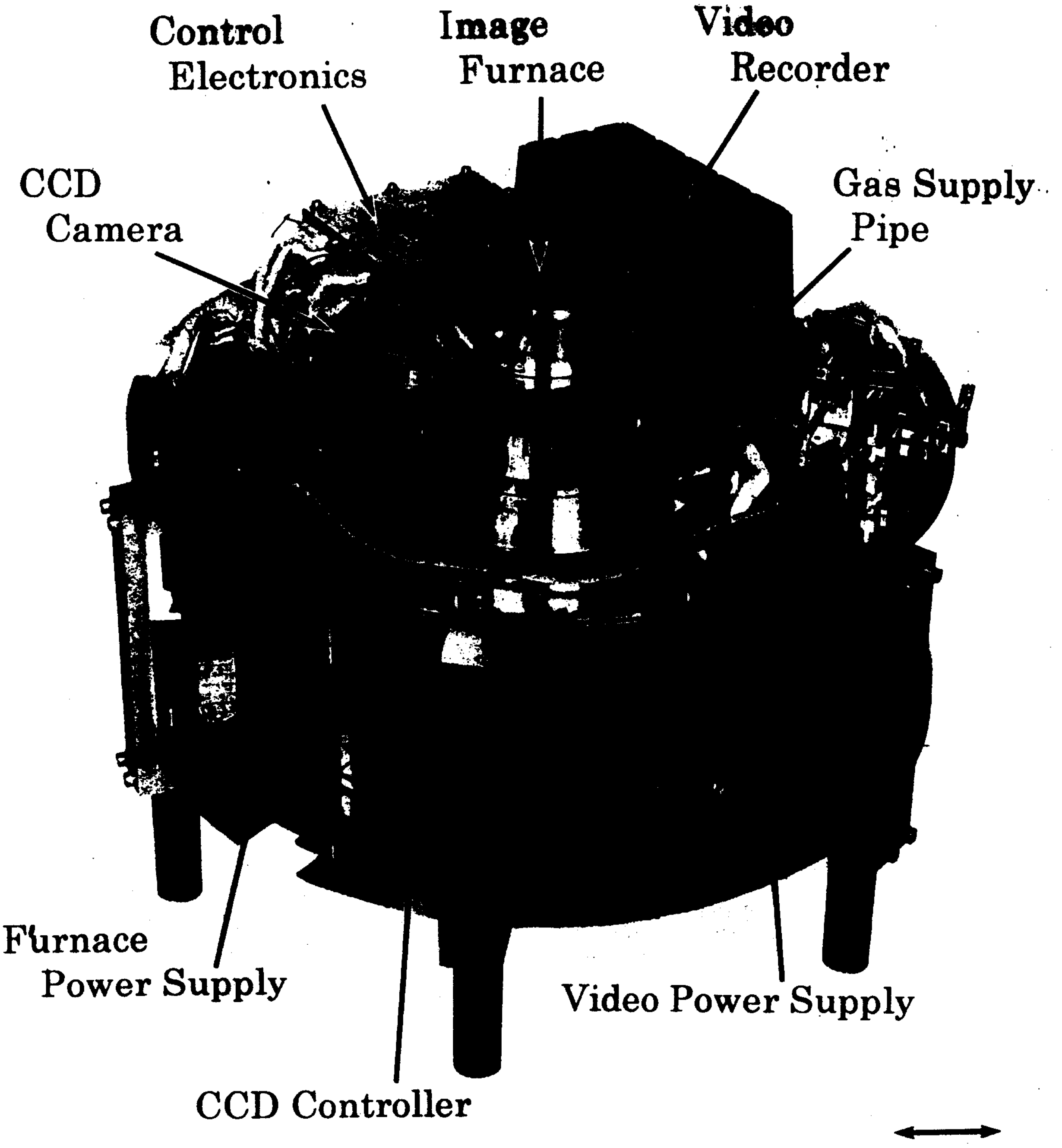
0.5秒後

MARANGONI FLOW

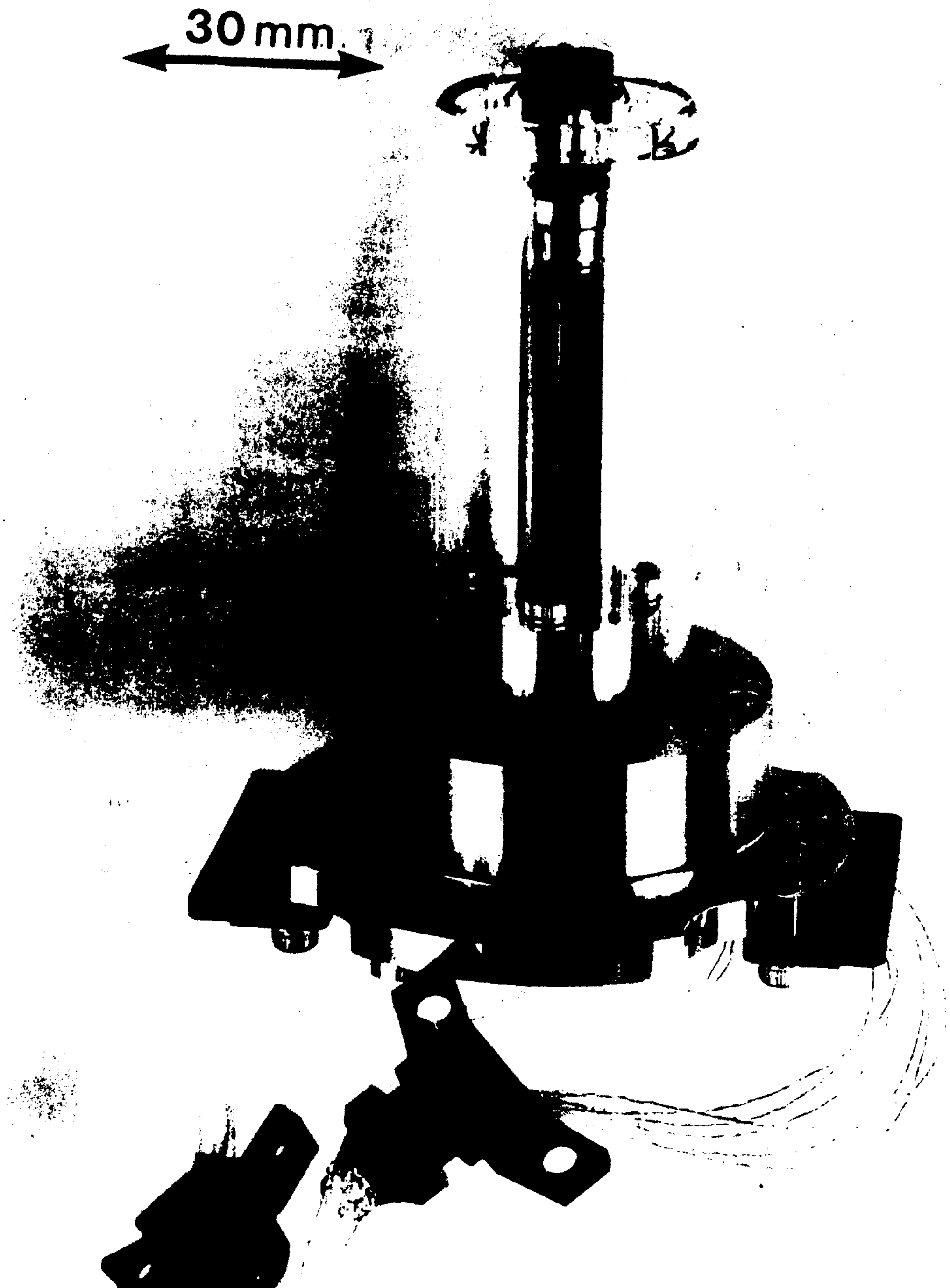
- ◆ NO CLEAR EXPLANATION ON MARANGONI FLOW
- ◆ DIFFICULTY IN EXPERIMENTS

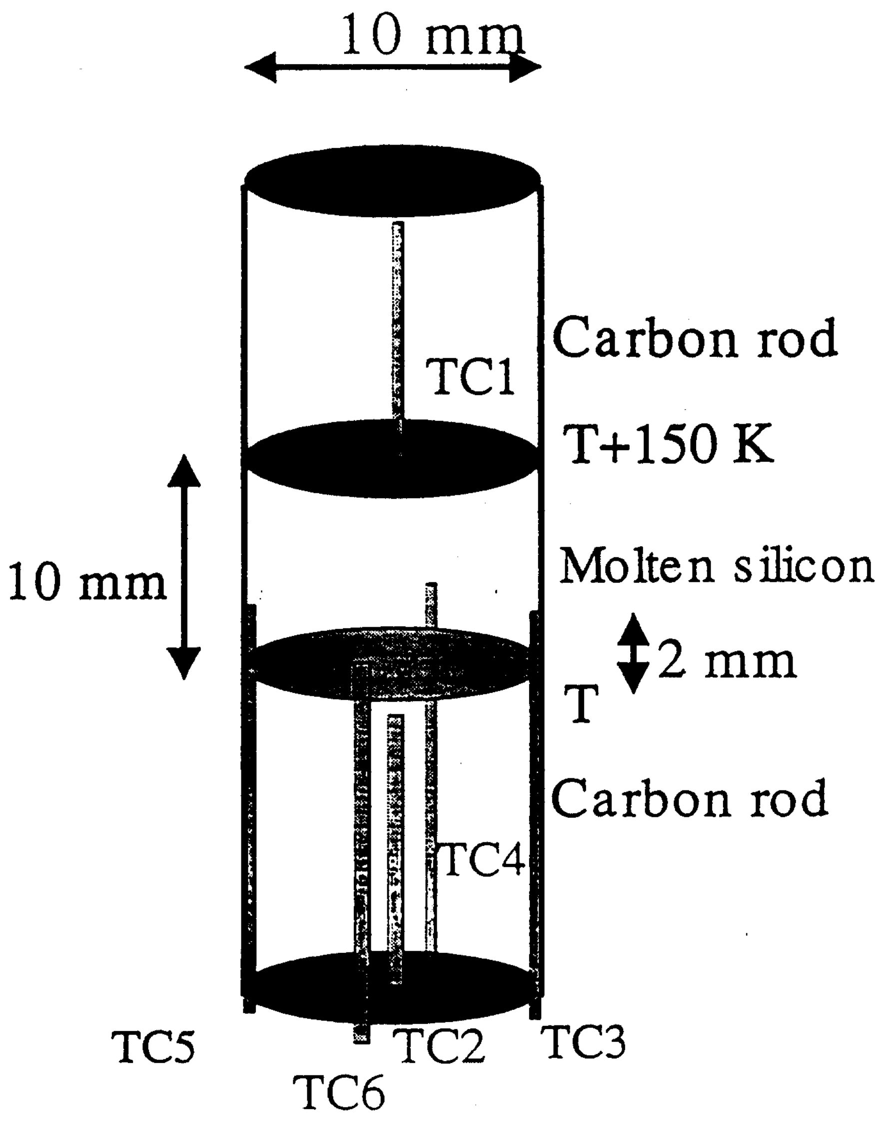
  DISTINGUISHING MARANOGNI FLOW FROM BUOYANCY FLOW

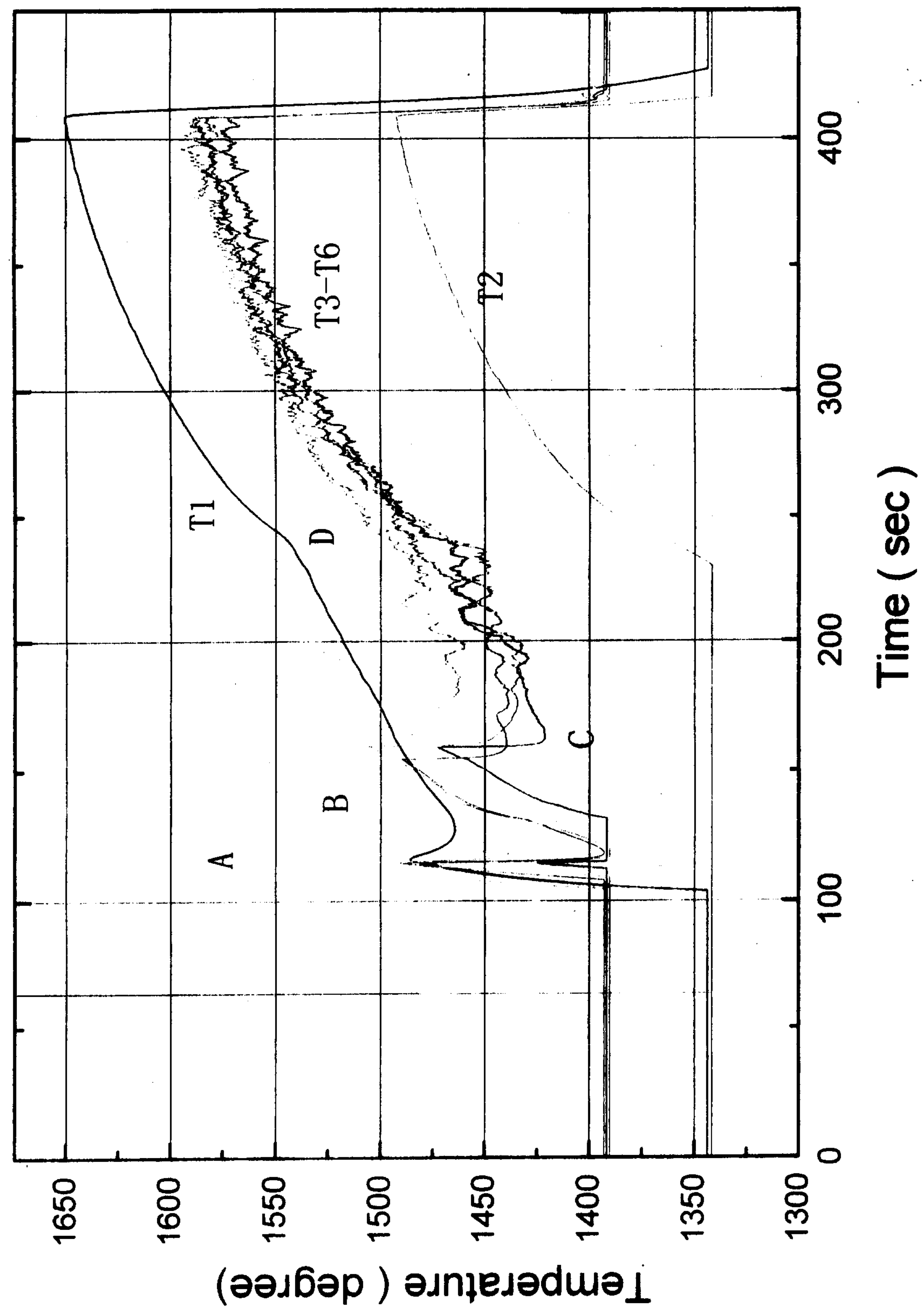
⇨ UTILIZATION OF MICROGRAVITY


Control Electronics
Image Furnace
Video Recorder
CCD Camera
Gas Supply Pipe
Furnace Power Supply
Video Power Supply
CCD Controller
100 mm

YCP-85730

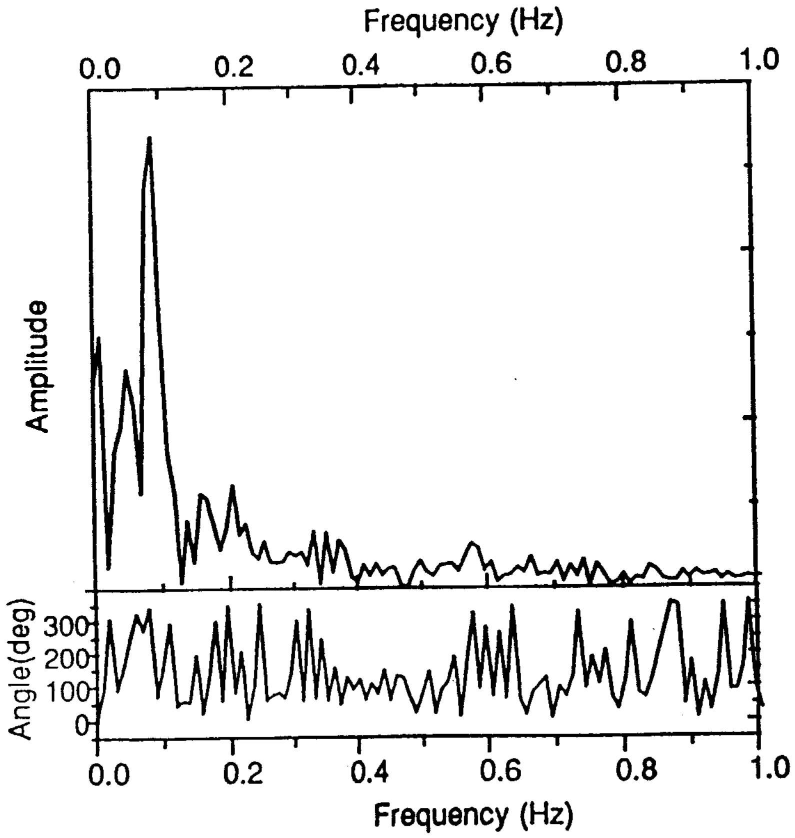



10 mm
Carbon rod
TC1
T+150 K
10 mm
Molten silicon
2 mm
T
Carbon rod
TC4
TC5
TC2
TC3
TC6

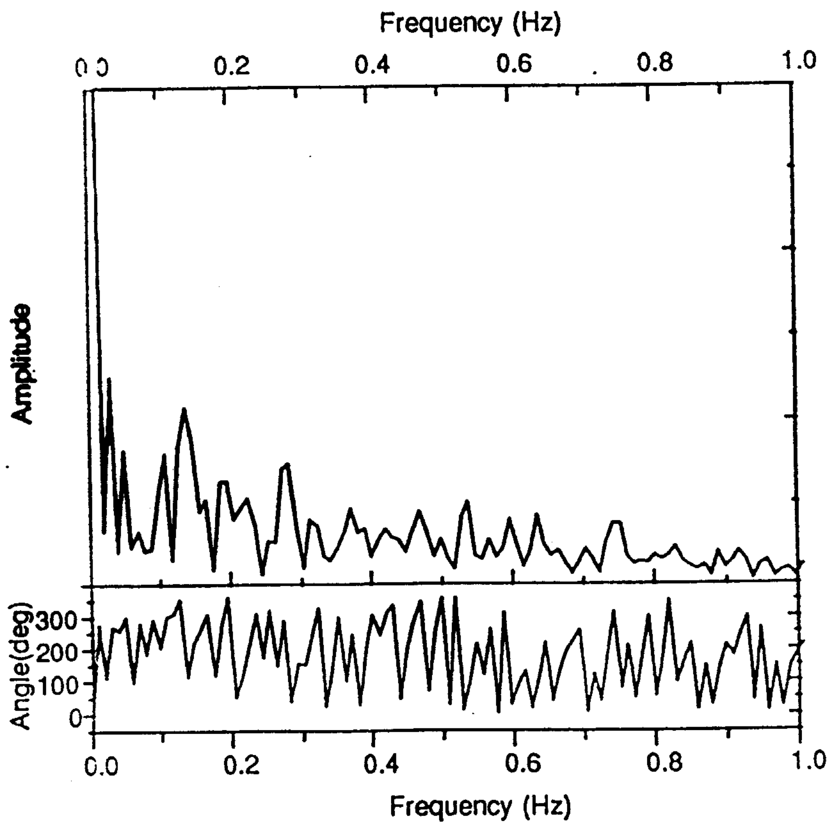


Temperature (degree)
1650
1600
1550
1500
1450
1400
1350
1300
Time (sec)
0
100
200
300
400
T1
T2
T3–T6
A
B
C
D

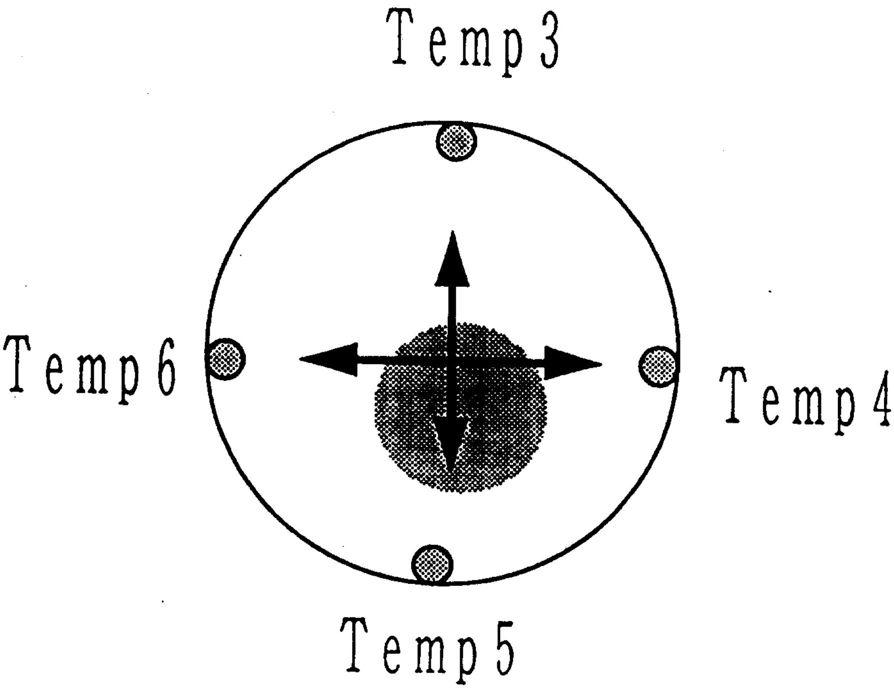
FFT-ANALYSIS

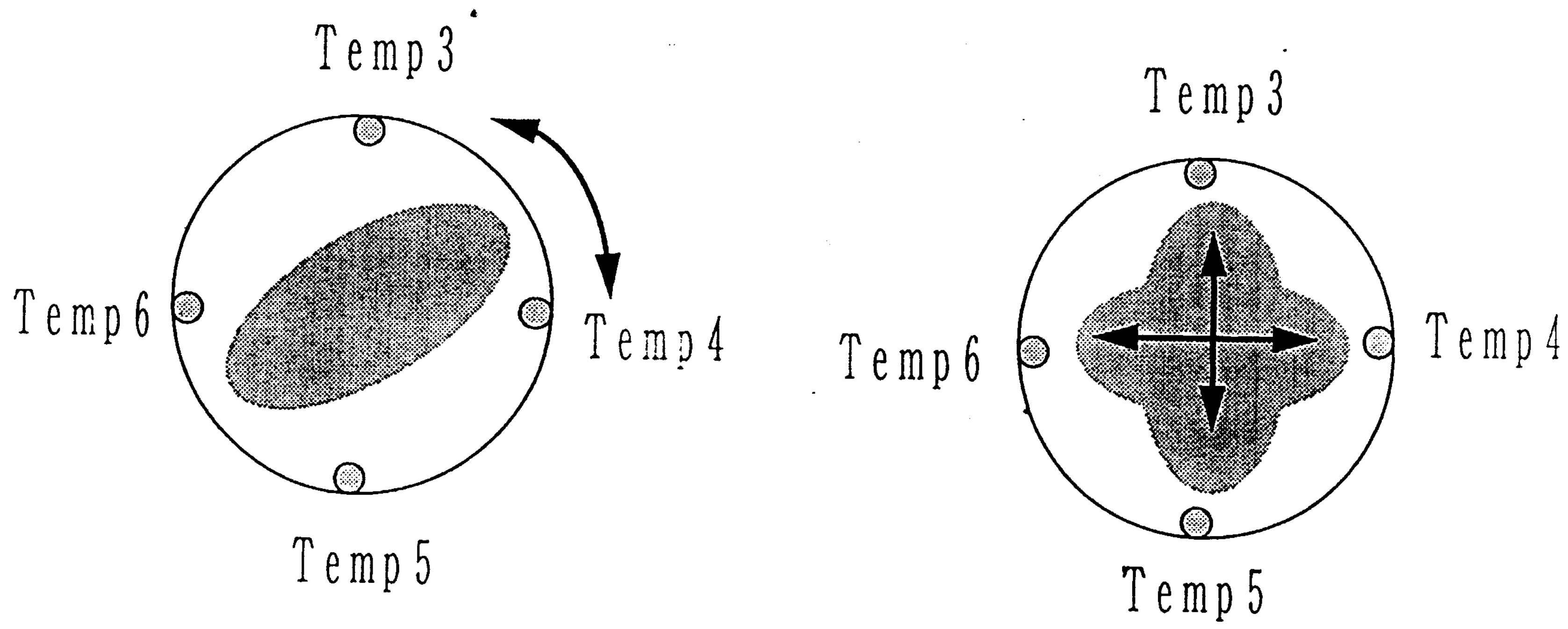


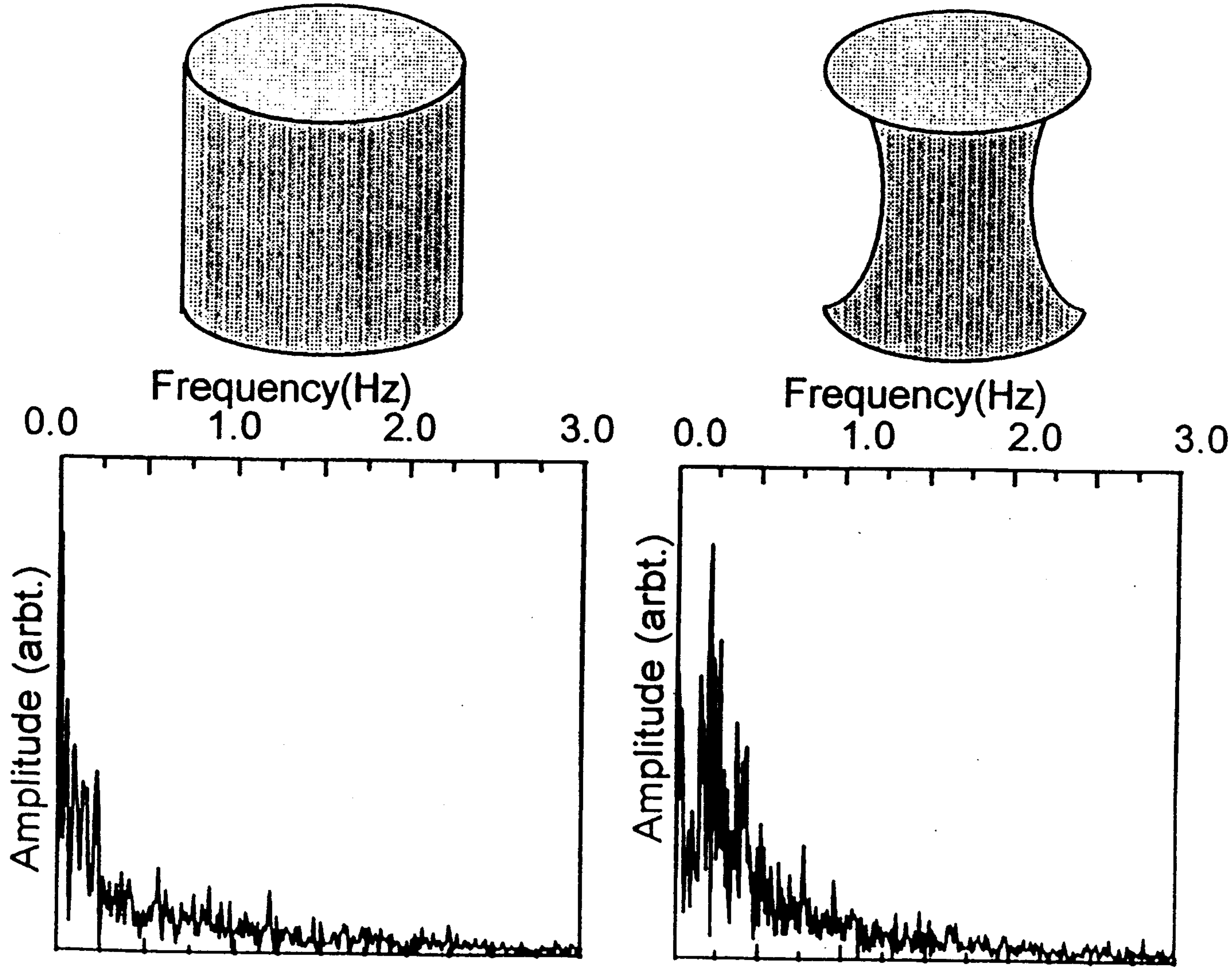
DURING COLUMN FORMATION



AFTER COLUMN FORMATION

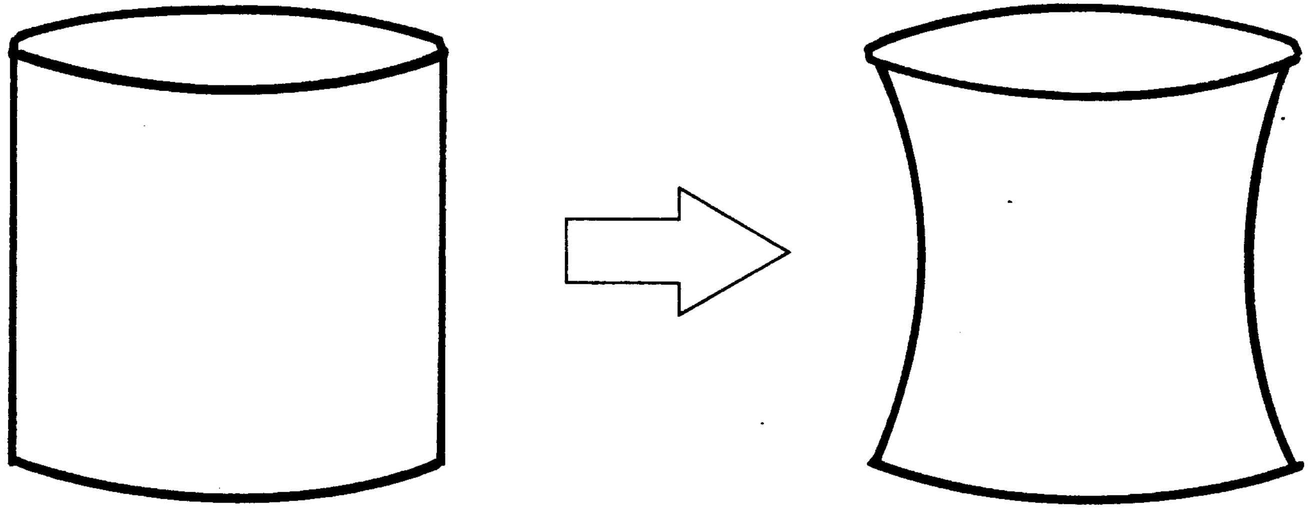

Temp3
Temp6
Temp4
Temp5


Temp3
Temp6
Temp4
Temp5
Temp3
Temp6
Temp4
Temp5



STRAIGHT　　　　WAIST-SHAPED

FLOW STABILIZATION



SILICONE OIL Pr = 10^{1}

PERIODIC-OSCILLATORY ⇨ STEADY

MOLTEN SILICON Pr = 10^{-2}

NON-PERIODIC-OSCILLATORY ⇨ PERIODIC-OSCILLATORY

	1g		μg	
	DRUM	STRAIGHT	DURING COLUMN FORMATION	AFTER COLUMN FORMATION (STRAIGHT)
TEMPERATURE OSCILLATION	~0.2 Hz	NPF	0.1 Hz	NPF
FLOW MODE	POF	NPOF	POF	NPOF
PHASE	1-FOLD	?	2-FOLD	1-FOLD

※ NPF: NO PARTICULAR FREQUENCY
POF: PERIODIC-OSCILLATORY FLOW
NPOF: NON-PERIODIC-OSCILLATORY FLOW

- $$Ma = \frac{\left|\partial\gamma / \partial T\right| \cdot \Delta T \cdot L}{\mu \cdot \kappa}$$

  ~30,000

- TEMPERATURE ASYMMETRY : HIGHEST AT T3

- HEAT BALANCE AT L/G-INTERFACE : Q > 0

  Bi

CONCLUSION

- FIRST MARANGONI FLOW EXPERIMENT
  FOR MOLTEN SILICON FROM THE VIEW
  POINT OF HEAT AND MASS TRANSFER
- NON-PERIODIC-OSCILLATORY FLOW
  AFTER FORMATION OF LIQUID COLUMN
- PERIODIC-OSCILLATORY FLOW
  DURING COLUMN FORMATION
- STABILIZATION OF MARANGONI FLOW
  DUE TO DEFORMATION