PNC TN8100 91－030

第10回ＰＮＣ／ＫｆＫ高レベル廃棄物管理会議発表資料集

（1990年11月18日～22日動燃東海事業所および東京にて開催）

1 9 9 1年4月

動力炉・核燃料開発事業団
東 海 事 業 所

PNC TN8100 91－030
1 9 9 1 年 4 月

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（1990年11月18日～22日動燃東海事業所および東京にて開催）

実施責任者　坪谷隆夫* 高橋武士**

報　告　者　吉岡正弘**五十嵐寛**青野忠純***
江本秀夫**鮎田文夫**遠藤　昇**
正木敏夫**加藤　功**菖蒲康夫**
池上靖志**

要　　旨

本資料集は、第10回ＰＮＣ／ＫｆＫ高レベル廃棄物管理会議（1990年11月18日～22日、動燃事業団の東海事業所および東京にて開催）において、双方から発表されたＯＨＰ資料をとりまとめたものである。

ＫｆＫ側の発表内容は、ＫｆＫ－ＩＮＥでの高レベル廃棄物に関する技術開発の概要、Ｋ－6’メルタの運転結果、オフガス処理設備の特性、メルタ内のシミュレーション結果、耐火物および電極材料と溶融ガラスの反応に関するものである。

ＰＮＣ側の発表内容は、ガラス固化技術開発の現状、モックアップ３号のメルタの運転経験および白金族元素の抜き出し性評価、Ｒｕ、Ｃｓ、Ｓｒおよび粉塵のオフガスへの移行評価ならびに準揮発性元素の模擬廃液仮焼時の揮発率評価、メルタ内のシミュレーション技術開発、新電極材料および新耐火物材料の開発に関するものである。

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*** 東海事業所　環境施設部　技術課

別冊資料目次

1.会議の日程およびプログラム

The 10th KfK-PNC Annual Meeting on High Level Waste Management

AGENDA

Nov. 17th(Sat.) Arrive at Narita
Stay at Shinbashi Daiichi Hotel

18th(Sun.) Meeting at Tokyo Bay Hilton Hotel with Tokyo staffs in the evening
Stay at Tokyo Bay Hilton Hotel

19th(Mon.) General discussion at the Hilton Hotel

AM:
1.Welcome remarks — PNC Yamamoto, M
2.Presentation on the state of the arts on the HLLW conditioning program
(1) State of the arts on the HLLW conditioning program in Japan — PNC Mano, T
(2) State of the arts on the HLLW conditioning program in Germany — KfK Dr.Weisenburger
3.Discussion

PM: Move to Katsuta
Stay at Hotel Crystal Plaza in Katsuta

20th(Tsu.)

8:20	Pick-up at Crystal Plaza in Katsuta	
9:00	Welcome remarks	PNC Yamanouchi, T.
9:15	Opening remarks	PNC Tsuboya, T. KfK Dr.Weisenburger
9:30	Technical Presentation	Chairman : Takahashi, T.
	(1) Evaluation at Mock-up 3 melter operation with noble metals	PNC Yoshioka,M
10:15	(2) Operation results with the K-6' melter	KfK Dr.Grunewald
10:40	(Coffee Break)	
11:05	(3) Off-gas treatmentnt process melter	PNC Igarashi, H.
11:25	(4) Performance of the off-gas treatment system	KfK Dr.Weisenburger
11:55	(5) Development of Three Dimensional Thermal Fluidic analysis Code for Electric Glass Melter	PNC Ayame, Y.
12:20	(Lunch)	

(to be continued)

13:30 Technical Discussion

(6) 3-dimensional melter modelling
KfK Dr.Roth

14:30 (7) Advanced electrode and refractory materials
PNC Masaki, T.

15:00 (8) Corrosion of melter materials
KfK Prof.Pentinghaus

15:30 (Coffee Break)

15:50 Technical Discussion

17:00 (Dinner)
Transfer from PNC to Mito

21st(Wed.) 8:30 Pick-up at Crystal Plaza

9:00 Technical Discussion

10:00 (Coffee Break)

10:15 Technical Discussion

11:55 (Lunch)

13:20 Technical Discussion

14:50 (Coffee Break)

15:00 Transfer from Head Bulding to TVF construction site

15:10 Facility tour
TVF construction site
PNC (Construction & Maintenace Office)
PNC Karino, M Ueno, T
EDF- III
PNC (CMS)
PNC Kawada, T Kawatsuma, S

16:45 Transfer from EDF- III to Head Bulding

16:50 Discussion

17:05 (Dinner)
Transfer from PNC to Katsuta

(to be continued)

22th(Thu.)	8:30	Pick-up at Crystal Plaza
	9:00	Technical Discussion
	10:00	(Coffee Break)
	10:15	Technical Discussion
	11:55	(Lunch)
	13:00	Technical Discussion
	15:00	(Coffee Break)
	15:15	Summarize the meeting records
	15:45	Closing remarks PNC Tsuboya, T.
	17:00	(Dinner) Transfer from PNC to Katsuta
23th(Fri.)		National Holiday (Labor thanks giving day) Move to Osaka Stay at Hotel Consolt in Osaka
24th(Sat.)		Free
25th(Sun.)		Free Move to Kyoto Stay at Kyoto Century Hotel in Kyoto
26th(Mon.)		Visit to Nippon Electric Glass Co.,Ltd. and Discussion Stay at Kyoto Century Hotel in Kyoto
27th(tsu.)		Move to Osaka Leave Osaka for Frankfurt

2. 動燃側の発表資料

2-1 Overview and Strategy for Radioactive Waste Management in PNC

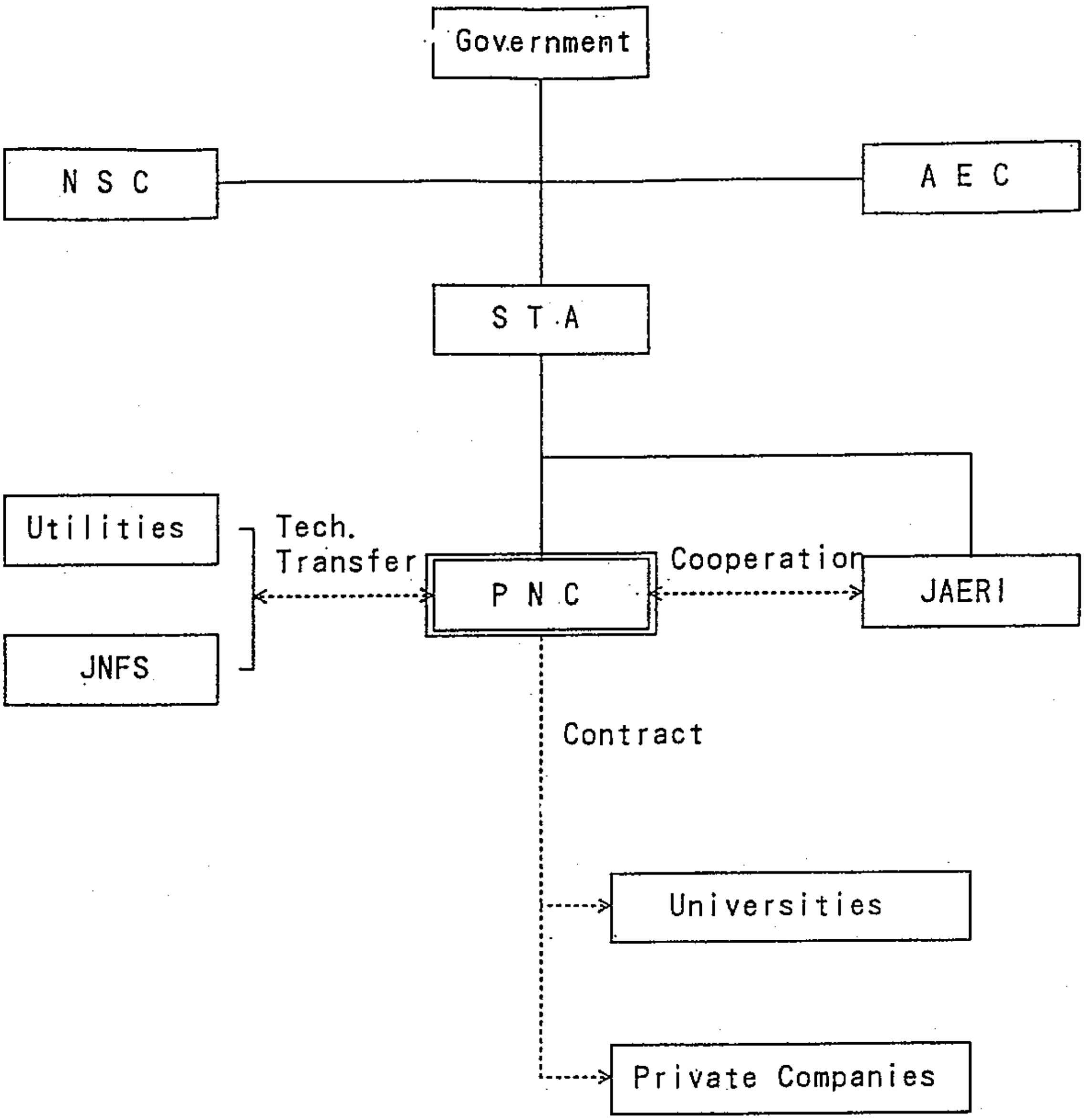
PNC TN8100 91-030

Overview and Strategy for

Radioactive Waste Management in PNC

November, 1990

POWER REACTOR AND NUCLEAR FUEL
DEVELOPMENT CORPORATION (PNC)



TRU Waste Management Organization in Japan

Oct. 1, 1990

Waste Management Organization of PNC
(PNC Works Concerned)

Chubu Works		Waste Isolation Reserch Section	General Manager : M. Yamakawa Deputy : M. Iketani Senior Staff : T. Takeda
		Exploration and Mining Technology Development Section	General Manager : T. Tomishige Deputy : K. Tsubota
Ningyo-toge Works	Waste Isolation and Ore Processing Division Director : T. Otake	Waste Isolation and Ore Processing Technology Development Section	General Manager : K. Otomura Deputy : Y. Komoto Senior Staff : S. Masumoto
Tokai Works	Waste Plants Operation Division Director : Y. Asakura Senior Staff : T. Omori	Coordination Section	General Manager : E. Inada Deputy : T. Asami
		Low-Level Waste Conditioning Section	General Manager : K. Ochiai Deputy : K. Takeda Senior Staff : T. Mizutani
		TRU Waste Conditioning Section	General Manager : T. Nagasaki Deputy : H. Miyo
	Waste Technology Developmet Division Director : T. Tsuboya Senior Staff : M. Horie	High-Level Waste Technology Section	General Manager : T. Takahashi Senior Staff : R. Mizuno
		Low-Level Waste Technology Section	General Manager : Y. Nakanishi
		Geological Isolation Technology Section	General Manager : H. Sasaki Deputy : Y. Yusa
O-arai Engineering Center		Waste Management Section	General Manager : H. Miyao Deputy : H. Omori

Oct. 1, 1990

Waste Management Organization of PNC (PNC H/Q)

Radioactive Waste Management Project

Senior Director : Y. Hashimoto (Executive Director)
Deputy Senior Director : M. Yamamoto
Deputy Director : H. Ando
Deputy Director : A. Yamato

Senior Staff :
K. Hanada
K. Kikuchi
H. Sato
Y. Ando

Coordination Section

General Manager : K. Tsurumaki
Deputy : S. Tajima

- Project Coordination
- Waste Management Implementation Control

Conditioning Research Program

General Manager : T. Mano

Research Management on Conditioning, D&D Technologies

Isolation Reserch Program

General Manager : S. Masuda
Deputy : M. Ohsawa

Management on Geological Isolation Research and Technologies

Geosciences Research Program

General Manager : T. Shiota
Deputy : M. Kinugasa

Management on URL and Storage Engineering Center Program

Presentation Management Research Program

General Manager : Y. Kimura
Senior Staff : H. Komoto

Management and Implementation of Presentation Management Research and Geotopia

Facilities in PNC Concerned Waste Management

(*) including TRU bearing waste

Waste Classification		HLW	LLW: from MOX (*)	LLW: from Reprocessing (*)	LLW: Hull (FBR)	LLW: U	LLW: Joyo	LLW: FMF
CONDITIONING	Cold R&D	ETF, MTF, EDF III	—	—	—	—	—	—
CONDITIONING	Hot R&D	CPF	—	—	WDF	—	—	—
CONDITIONING	Demonstration (in operation)	—	PWTF	AsF ST	—	—	＊1	—
CONDITIONING	Demonstration (to be operated)	TVF	—	LWTF	HWTF	UWTF	＊2	—
Storage (in operation)		—	PWSF	LASWS I LASWS II As I, As II	HASWS I HASWS II	UWSF	＊3	—
D & D (R & D)		WDF					—	—

PNC TN8100 91—030

＊1 : Joyo Radioactive Waste Treatment Facility
＊2 : New Joyo Radioactive Waste Treatment Facility
＊3 : Radioactive Waste Treatment Plant of the O-arai Site

Tokai Reprocessing Plant (TRP)

Hot operation	since 1977
Reprocessed LWR fuel	416 tons
Recovered Plutonium	2.8 tons
Separated HLW	365 m^3

(Mar. 31, 1990)

JNFS Reprocessing Plant

Start operation	around 1997
Nominal Capacity	800 tons/ye[illegible]

PNC TN8100 91—030

Waste Arising and Conditioning in PNC Tokai Works

(Waste Containing TRU Nuclides)

	Waste	Conditioning Methods ( ) : Under planning	Cumulative-Waste-after conditioning (drums)
Takai Reprocessing Plant	Hull and Hardware	(HIP)	1,300
	LLLW	Bitumen	17,000
	Spent Solvent	Plastics	1,900
	Solid Waste -Combustible -Non-combustible	 (Cement) (Cement)	 200 700
Mox Fuel Fablication Facilities	Solid Waste -Combustible -Non-combustible	Incineration-Melting with Microwave Melting with ESR	200 600

Long Term Program on HLW Management

(1987, AEC)

- Utilization of uranium and plutonium recovered from reprocessing of spent fuel shall be promoted;

- Plutonium utilization in LWRs and in advanced thermal reactors (ATRs) shall be promoted;

- Safe and appropriate treatment and disposal of radioactive waste shall be implemented.

HLW Treatment Program

(1987, AEC)

- Demonstrate the Vitrification Technology based on LFCM Method Using Borosilicate Glass in the Early 1990's

- Reflect the Vitrification Technology Developed by PNC to the private Sector's Reprocessing Plant

The Basic Policy on HLW Management

(1987, AEC)

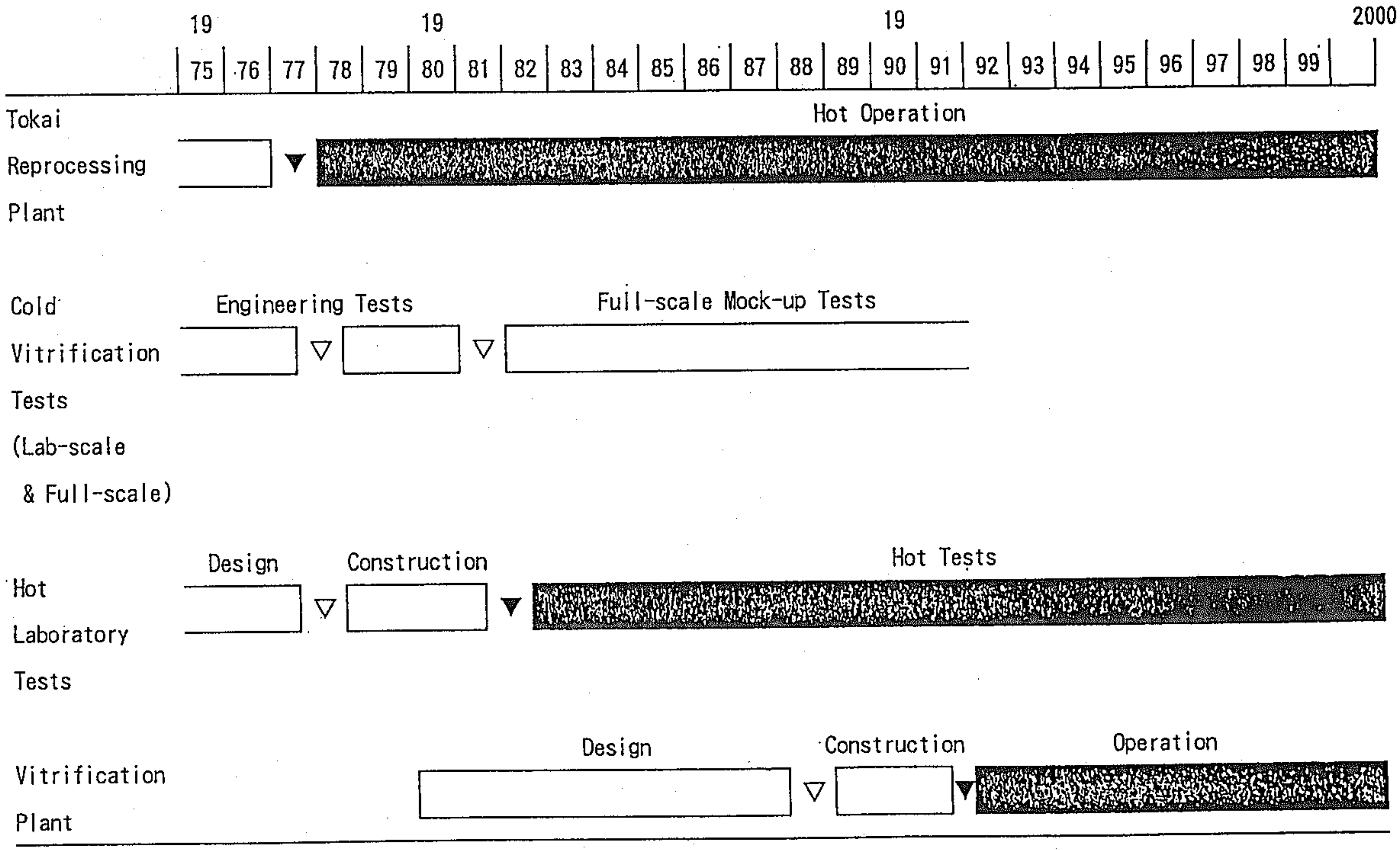
(1) Vitrification of HLW → (2) Storage 30～50Yrs → (3) Geological Disposal

(1) Vitrification of HLW
(borosilicate glass)

(2) Storage 30～50Yrs
(cooling)

(3) Geological Disposal
(several hundred meters underground)

Schedule for the Development of HLW Vitrification Technology

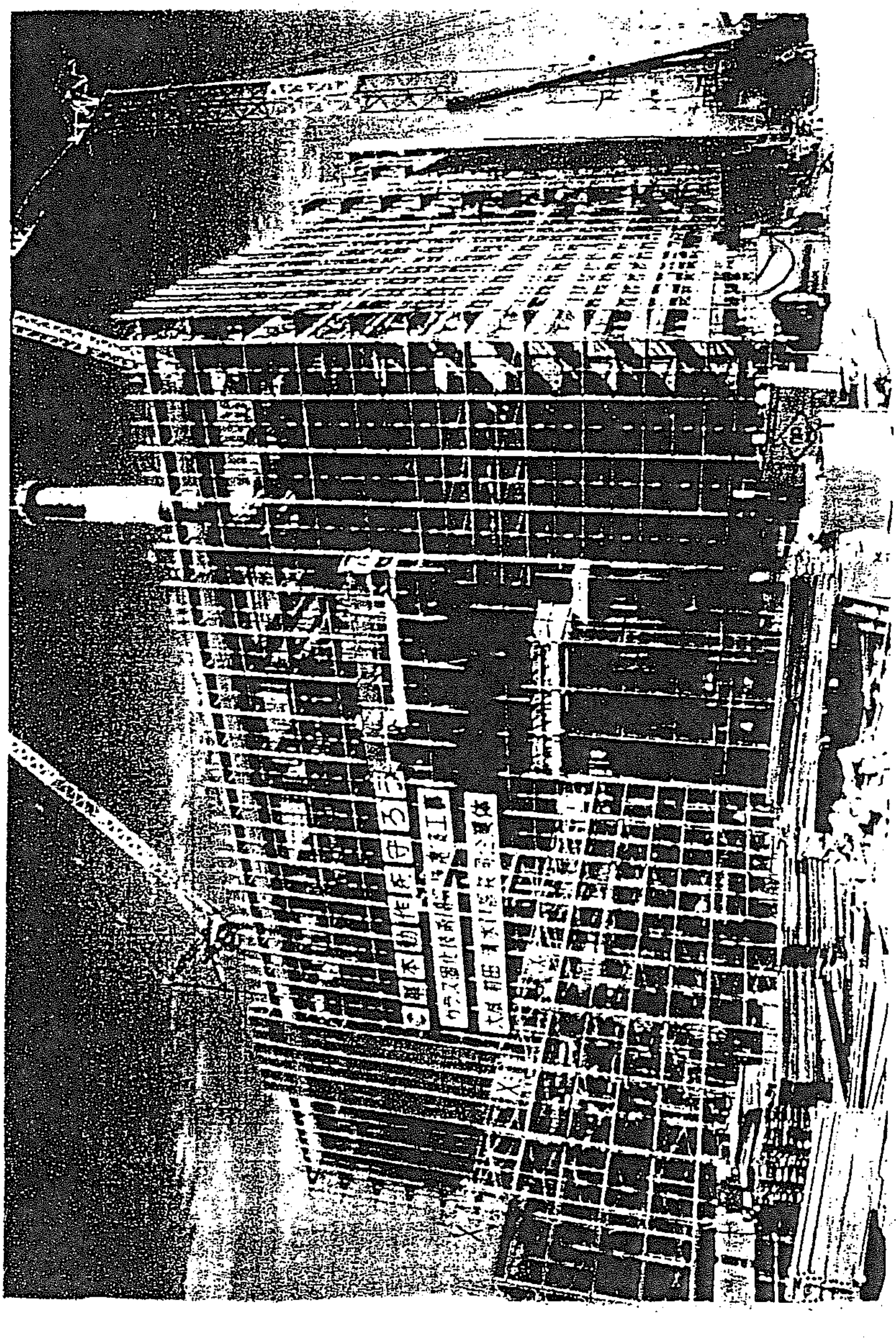


Features of Tokai Vitrification Facility (TVF)

- ○ Process : Liquid-fed joule-heated ceramic melter (LFCM) process
- ○ Glass additive : Glass fiber cartridge
- ○ Maintenance : fully remote maintenance
- ○ Ventilation : Low Flow Ventilation System
- ○ Cell Arrangement : Underground
- ○ Geseous Waste : Activity 1/1000 of TRP (Tokai Reprocessing Plant)
- ○ Independent Utility Equipment and Secondary Waste Treatment Equipment.

(Bird's-eye View of TVF)

	Administration Building	Vitrification Building
Length	28.8m	59.8m
Width	22.0m	44.3m
Height(above ground)	18.9m	26.0m
(Under Ground)	——	18.8m
Area of IF	700m^3	2,600m^3
Total Area	2,500m^3	12,000m^3
Total Volume of Building	11,000m^3	98,000m^3



(Package to be used in the TVF)

Contents	Borosilicate Glass
Weight	approx. 300kg
Volume	approx. 110 ℓ
Radioactivity	approx. 4×10^{5}Ci/package
Heat Generation Rate	approx. 1.4kW/package
Canister	Vertical Cylinder Type
Material	SUS304L/SUS F 304L
O.D.	430mm
Height	1,040mm

Technologies to be Developed

	Wastes	Technologies
from Reprocessing	Hull and hardware	• Compaction • HIP • Electro Slag Re-melting
	Spent Solvent	• Decomposition • Solidification(Hydro Thermal Solid., Ceramic)
	Low-Level Liquid Waste	• Solidification(Hydro Thermal Solid., Cement) • TRU Nuclides Separation
	Combustible Solid Waste	• Solidification (Hydro Thermal Solid., Cement, Melting with Microwave)
	Non-Combustible Solid Waste	• Electro Slag Re-melting • Compaction • Immobilization(Cement)
from MOX Fablication	Combustible Solid Waste	• Acid Digestion • Solidification
	Non-Combustible Solid Waste	• Decontamination (Electro Polishing) • Solidification

Japanese Policy on Management of Waste Containing TRU Nuclides

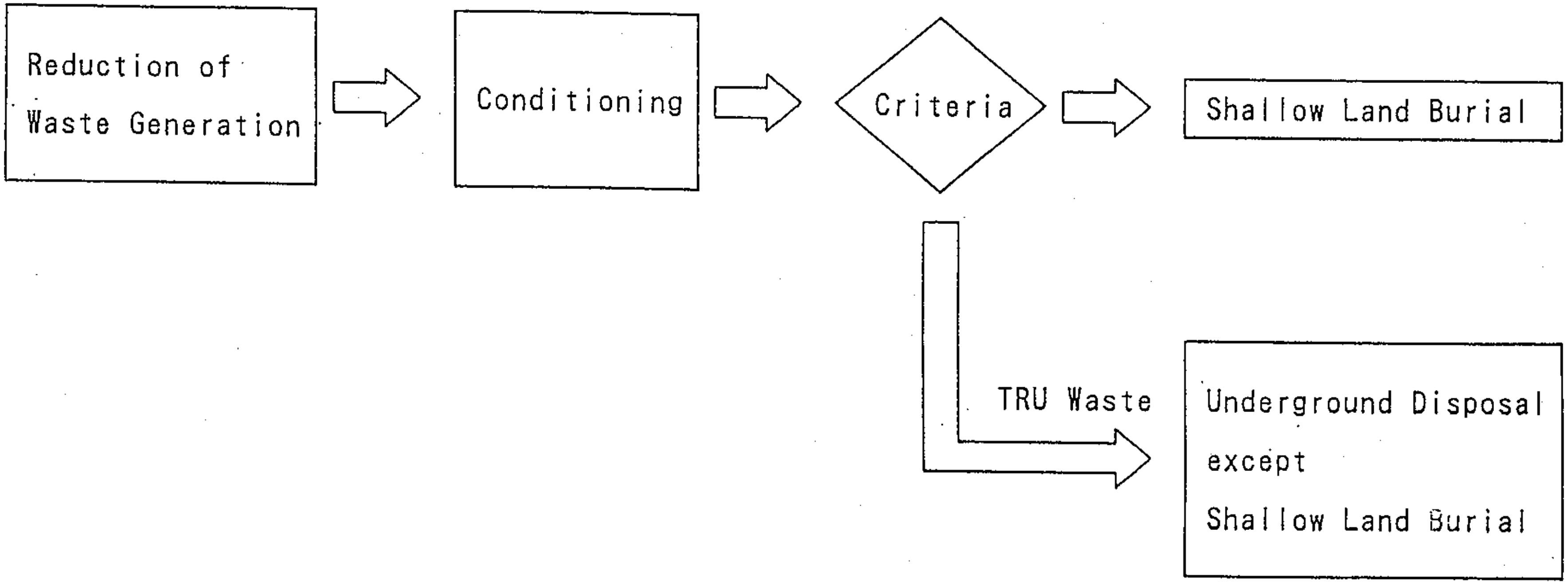
- Establish the Criteria for the Proper Classification and the Methods for the Rational Disposal

- Carry Out R&D on Disposal Technology by PNC with Cooperation of JAERI, Private Sectors and Universities

- Carry Out Study on the Entity to Take Charge of the Disposal and the Measures To Raise the Required Financial Resources

〔1987, AEC〕

Waste Management Strategy
(Waste Containing TRU Nuclides)



Objectives of Disposal Technologies to be Developed

- Identification of Source Term
- Development of Rational Disposal Methods in accordance with Each Waste Classification
- Optimization of Disposal System through Integration of Each Disposal Methods

Objectives of Conditioning Tecnologies to be Developed

- Reduction of Waste Generation
- Reduction of Waste Volume
- Stabilization
- Classification

R&D on TRU Waste Management

- Demonstration
  - -Bituminization
  - -Plastic Solidification
  - -Conditioning Waste from MOX Facilities

- Research and Development
  - -Conditioning Hull and Hardware
  - -Nuclide Separation from TRU Waste
  - -D&D
  - -TRU Waste Isolation Research

National Program for Geological Isolation of HLW

1st
(1976-1984)
Selection of Potential Geological Formations
- Research and Survey of Geological Formations
- Performance Assessment

⇒

2nd Stage
(1985-)
Selection of Candidate · Disposal Sites
- R&D Aiming at the Establishment of Geological Isolation Technology
- Investigation on Geological Environment

⇒ • Siting

3rd Stage
Demonstration of Geological Isolation Technology at the Candidate Site

⇒

4th Stage
Construction, Operation and Closure of the Repository

← PNC [Leading Organization] (JAERI, GS of JAPAN, etc.) →|← Implementation Body →

PNC Budget on Waste Management (FY '90)

130Yen=1$
M=million

	Conditioning	Storage	Disposal	Total
HLW	¥ 1.4B ($81M)	¥ 0.5M ($ 3.5M)	¥ 4.3M ($33M)	23B (171M)
LLW (Reprocessing)	¥ 3.2B ($25M)	¥0.02B ($ 0.1M)	¥ 0.1B ($ 0.8M)	
LLW (MOX, FBR)	¥ 3.2B ($25M)	¥ 0.1M ($ 1M)	――	
LLW (U)	¥ 0.1B ($ 0.9M)	――	¥0.04B ($ 0.3M)	

No.	Alloy	addition	Attack Depth (μm)	Solidus (°C)
1	F 8 Fe-40Ni-30Cr	no add.	219	1384
2		+0.3Ti	187	1377
3		+0.3Nb	152	1372
4		+0.3Mg	245	1383
5		+0.1Y	91	1385
6		+0.6Ti	140	1366
7		+1.0Ti	84	1360
8		+1.5Ti	120	1338
9		+1.0Nb	307	1357
10		+1.5Nb	282	1345
11	F 10 Fe-45Ni-35Cr	no add.	108	1380

Corrosion depth in molten glass and soidus for experimental alloys

2-2 Progress in Vitrification Technology in PNC Over a Past Year

PNC TN8100 91-030

Progress in Vitrification Technology in PNC Over a Past Year

The Tenth Annual KfK-PNC Meeting on Cooperation
in High-Level Waste Management

November 18 - 22, 1990

POWER REACTOR AND NUCLEAR FUEL
DEVELOPMENT CORPORATION

PNC

PROGRESS IN VITRIFICATION TECHNOLOGY
IN PNC OVER A PAST YEAR (Nov. 1990)

Mock-up III melter operation test in MTF

- establishing operational procedure of TVF process
- reliability test for overall system of process
- noble metal feed tests
  - -effect of optimizing temperature distribution in the melter on noble metal draining
  - -effect of bubbling on noble metal draining
  - -confirm the design and operational strategy to noble metal for long-term operation

SUMMARY OF MOCK-UP III MELTER OPERATION

ITEM \ TIME	1989								1990							
	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8
MELTER RUNS			20th				21st				22nd		23rd			
OPERATION OBJECTS	•Operation Test through a method Controlling Melter Bottom Temperature				•Confirmation of -Bubbling Effect -Operation Method Effect during Melter Idling				•Rinsing Test of the Noble Metals left in Melter				•Evaluation of Operation Method for Noble Metals during Longer Periods			
PRODUCED CANISTERS	19				15				—				35			

PROGRESS IN VITRIFICATION TECHNOLOGY IN PNC OVER A PAST YEAR (continued)

Design of large-scale melter

- schedule
  - -major structural design was finished
  - -detailed design of components is now underway
  - -operation is expected in Oct. 1991
- design features
  - -design phylosophy is based on TVF melter
  - -design throughput ; nominal : 44.3 kg glass/h
    maximum : 52.1 kg glass/h
  - -melting surface area : 2.2 m^2

PROGRESS IN VITRIFICATION TECHNOLOGY IN PNC OVER A PAST YEAR (continued)

Mathematical modelling

- comparison with the operational data
- modification of mathematical model
  - consider temperature dependance of resistivety for electric potential equation
  - consider buoyancy by bubbling in molten glass for momentum equation
- simulation of melter operation
  - effect of noble metal sedimentation
  - effect of bubbling

PROGRESS IN VITRIFICATION TECHNOLOGY IN PNC OVER A PAST YEAR (continued)

Fabrication of physical model
- now under test operation

Melter dismantling test
- preliminary test for dismantling metallic casing by plasma torch
- development of positionning tool for torch
- fabrication of remote dismantling test installations(underway)
- remote dismantling of radioactive melter in CPF

PROGRESS IN VITRIFICATION TECHNOLOGY IN PNC OVER A PAST YEAR (continued)

Study on semi-volatile elements

- volatization of Ru, Cs and Tc in calcination
- absorption of Tc into water in scrubbing

Study on off-gas entrainment in Mock-up operation

- effect of bubbling or plenum heating in melter on off gas entrainment (Ru, Cs and aerosol)

PROGRESS IN VITRIFICATION TECHNOLOGY IN PNC OVER A PAST YEAR (continued)

Glass characterization

- product characterization for Mock-up melter operation
  - noble metal element analyses for mass balance evaluation
- study on long-term thermal stability of glass product

Effect of noble metal elements on properties

- electric resistivity, - viscosity
- crystal growth of ruthenium dioxide

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- crystal growth of ruthenium dioxide

2-3 Evaluation of Mock-up ⅢMelter Operation with Noble Metals

PNC TN8100 91-030

EVALUATION OF MOCK-UP III MELTER OPERATION WITH NOBLE METALS

The Tenth Annual KfK-PNC Meeting on Cooperation
in High-Level Waste Management

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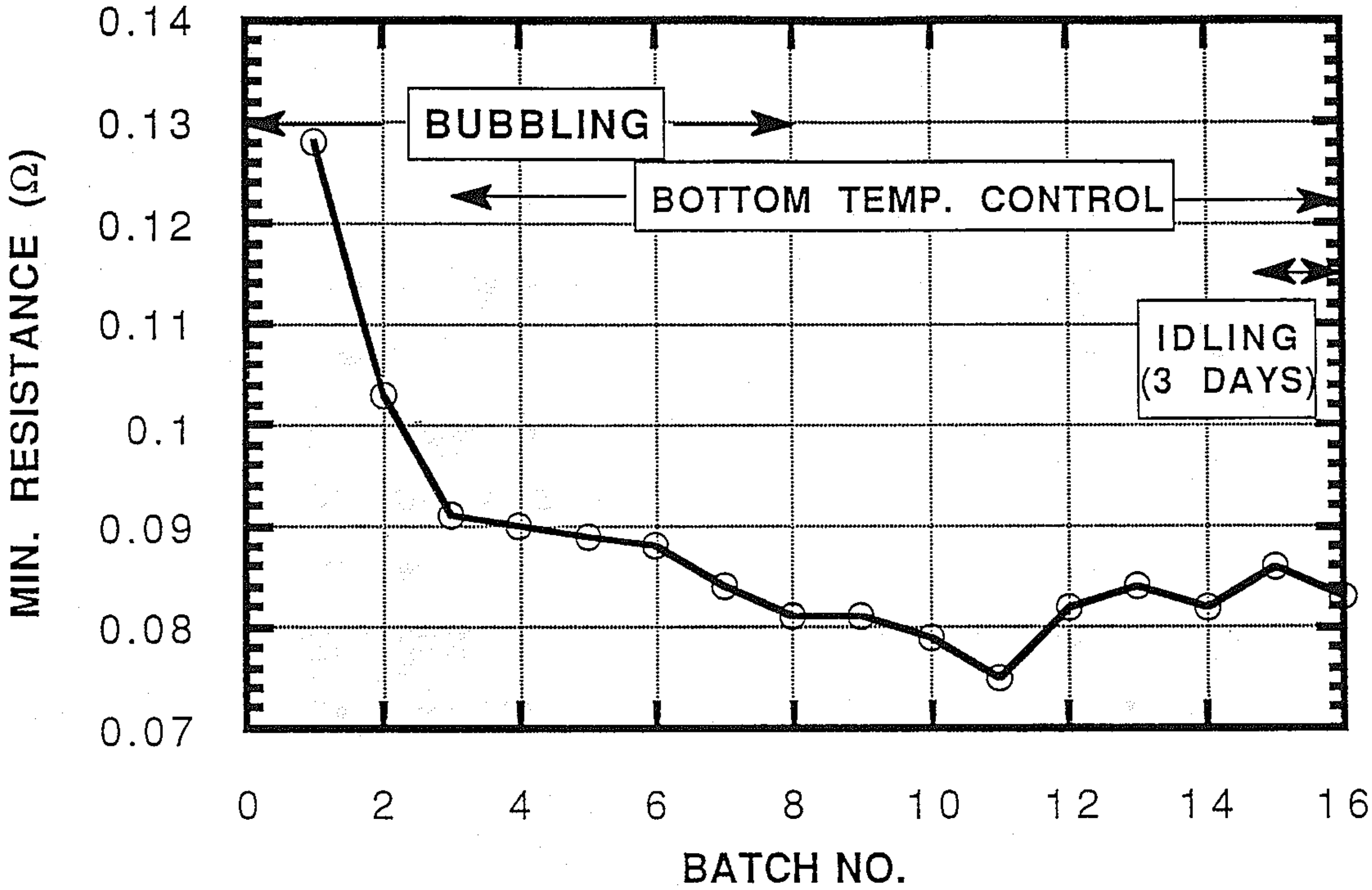
POWER REACTOR AND NUCLEAR FUEL
DEVELOPMENT CORPORATION

PNC

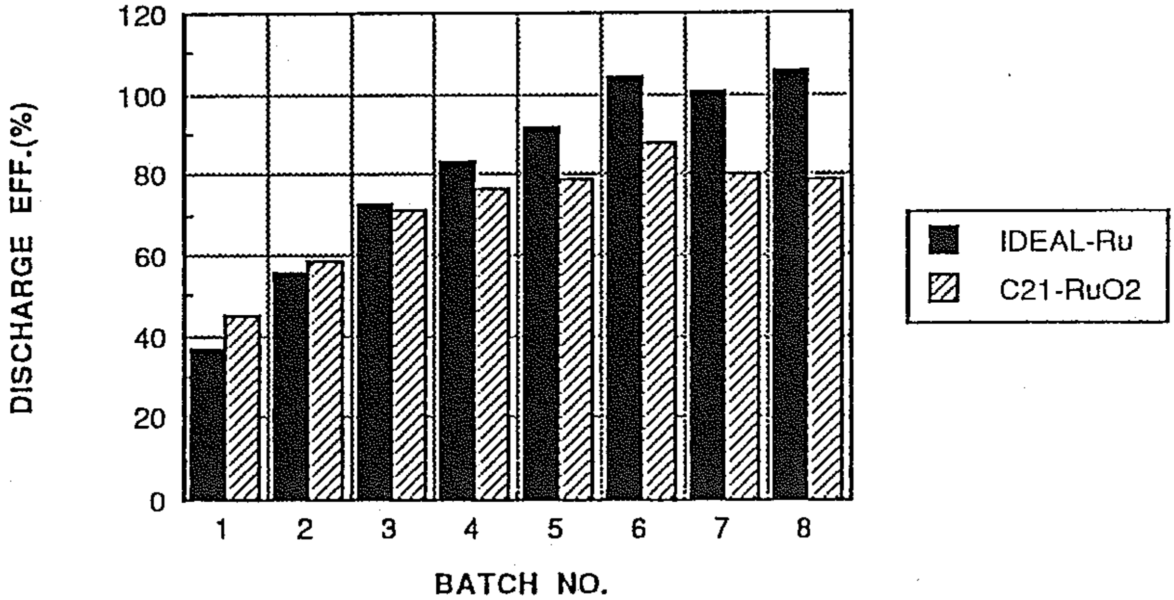
SUMMARY OF MOCK-UP III MELTER OPERATION WITH NOBLE METALS

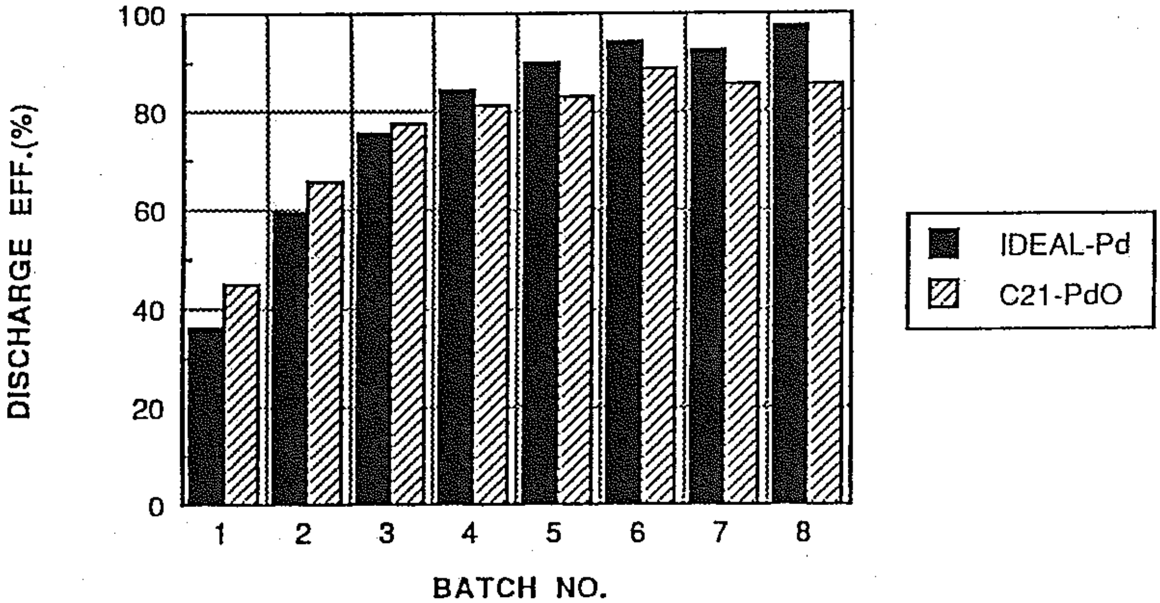
TIME / ITEM	1989				1990			
	5 6	7 8	9 10	11 12	1 2	3 4	5 6	7 8
MELTER RUNS	20th			21st		22nd	23rd	

ITEM	20th	21st	22nd	23rd
OPERATION OBJECTS	•Operation Test through a method Controlling Melter Bottom Temperature	•Confirmation of -Bubbling Effect -Operation Method Effect during Melter Idling	•Rinsing Test of the Noble Metals left in Melter	•Evaluation of Operation Method for Noble Metals during Longer Periods
AMOUNT	19 canisters	15 canisters		35 canisters
RuO_2 (kg)	40.8	41.6	—	93.6
PdO (kg)	22.9	17.7		37.3
Rh_2O_3 (kg)	0.7	1.3		9.3
OPERATION RESULTS	•Big Effect for Discharge of Noble Metals	•Cold Bottom Operation Superior to Bubbling •Effective in Idling	•Rinsing Effect by Na	•Discharge Over 99% of Noble Metals during 70 days Operation



RESISTANCE BETWEEN MAIN ELECTRODES DURING OPERATION

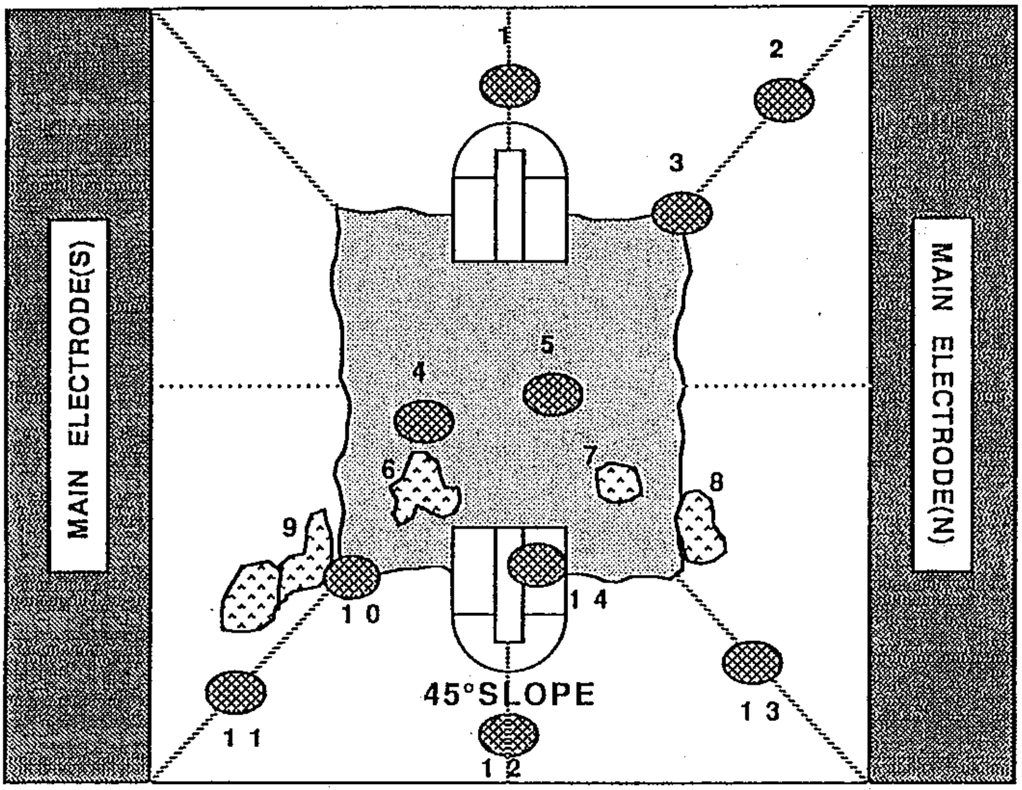




COMPARISON OF DISCHARGE EXPECTED AND RESULTED THROUGH BUBBLING

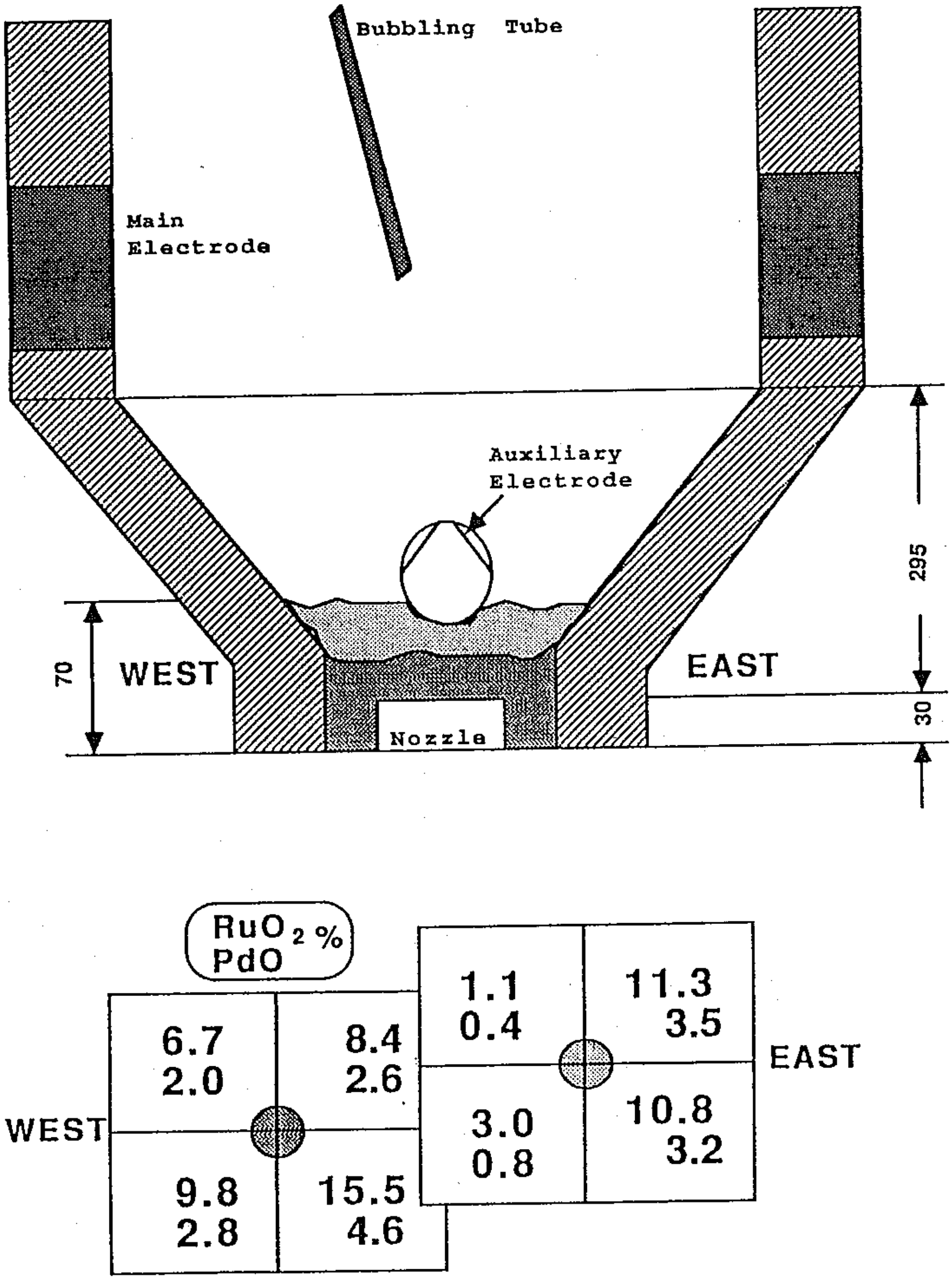
BALANCE OF NOBLE METALS IN MELTER (21st CAMPAIGN)

OPERATION METHOD / ITEM	BUBBLING RuO_2	BUBBLING PdO	COLD BOTTOM RuO_2	COLD BOTTOM PdO	COLD BOTTOM Rh_2O_3
FEED AMOUNT (kg)	18.7	8.7	17.5	7.9	2.5
DISCHARGE AMOUNT (kg)	16.6 (13.1)	8.2 (6.5)	16.8	7.4	2.6
AMOUNT HELD IN GLASS (kg)	(5.6)	(2.2)	-	-	-
ACCUMULATED AMOUNT (kg)	2.1	0.5	0.7	0.5	- 0.1
DISCHARGE EFFICIENCY (%)	88.8	94.3	96.0	93.7	102.4

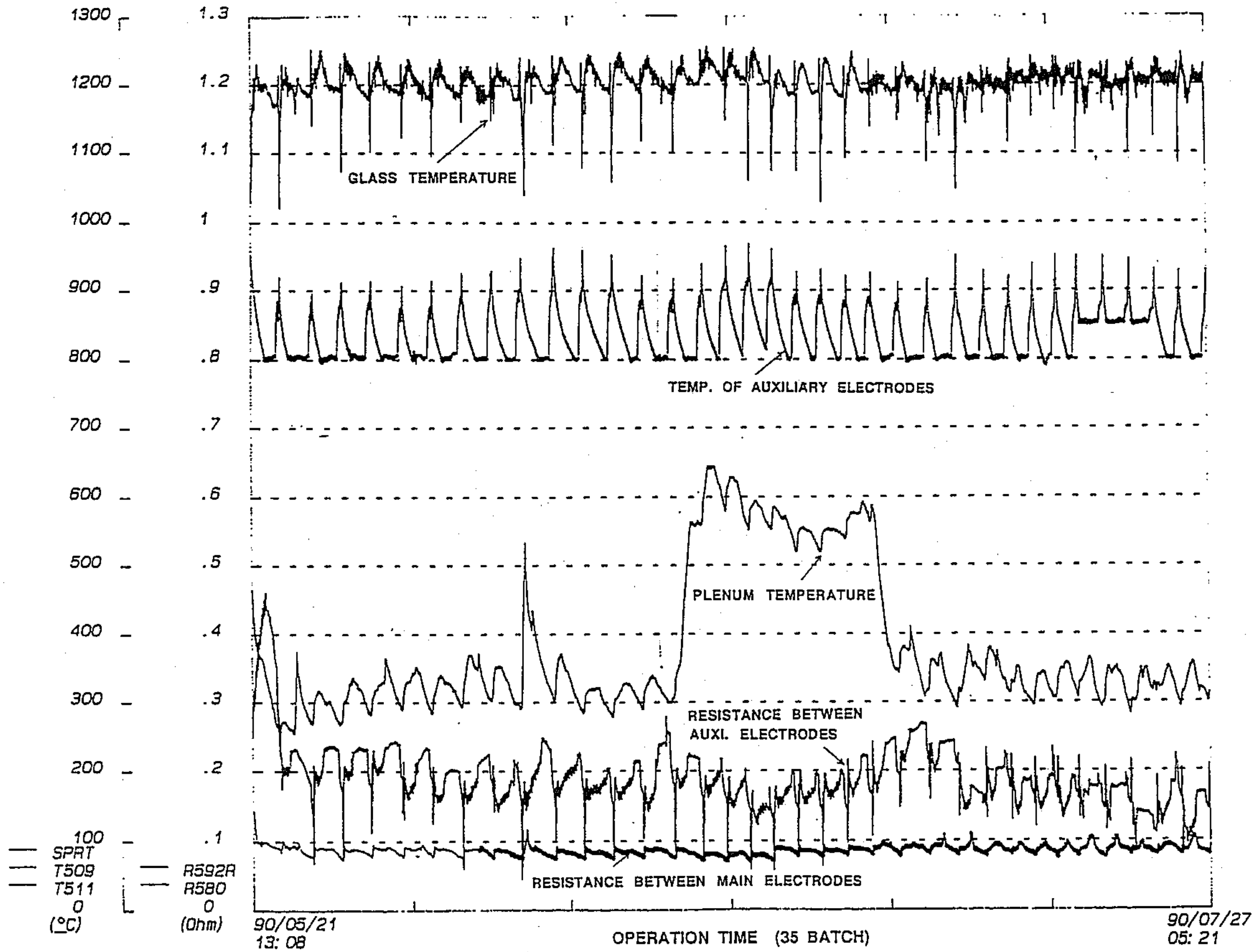


SAMPLE NO.	CONTENTS(%) RuO_2	CONTENTS(%) PdO	SAMPLE NO.	CONTENTS(%) RuO_2	CONTENTS(%) PdO
1	3.9	1.7	8	13.8	3.9
2	1.1	0.5	9	14.4	4.1
3	1.0	0.4	10	9.4	2.8
4	9.6	3.0	11	2.2	0.5
5	1.6	0.5	12	4.5	1.9
6	5.8	2.3	13	0.4	0.3
7	5.2	1.8	14	14.7	5.3

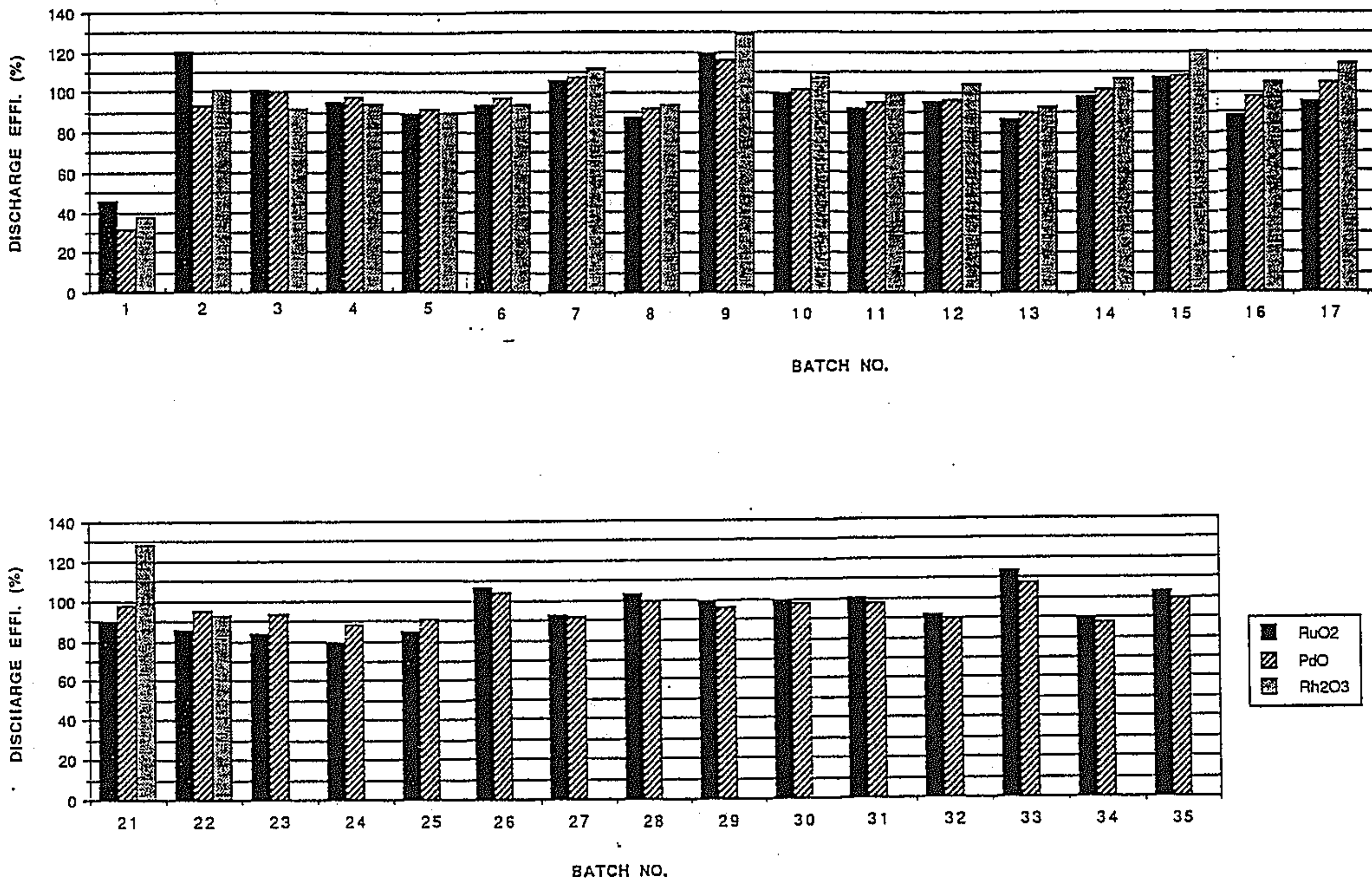
ACCUMULATION OF NOBLE METALS IN MELTER



NOBLE METAL CONTENTS IN ACCUMULATED GLASS



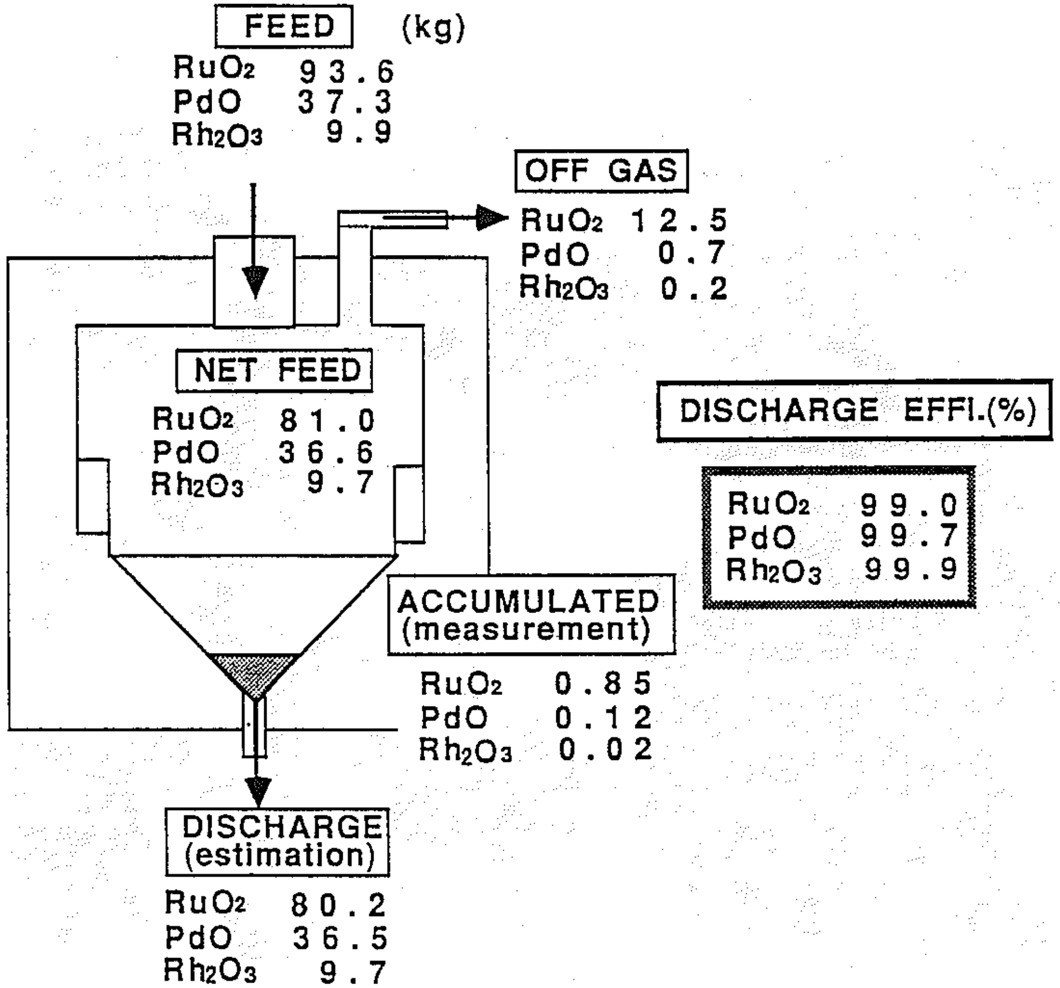
MELTER OPERATION DURING THE FEED OF NOBLE METALS



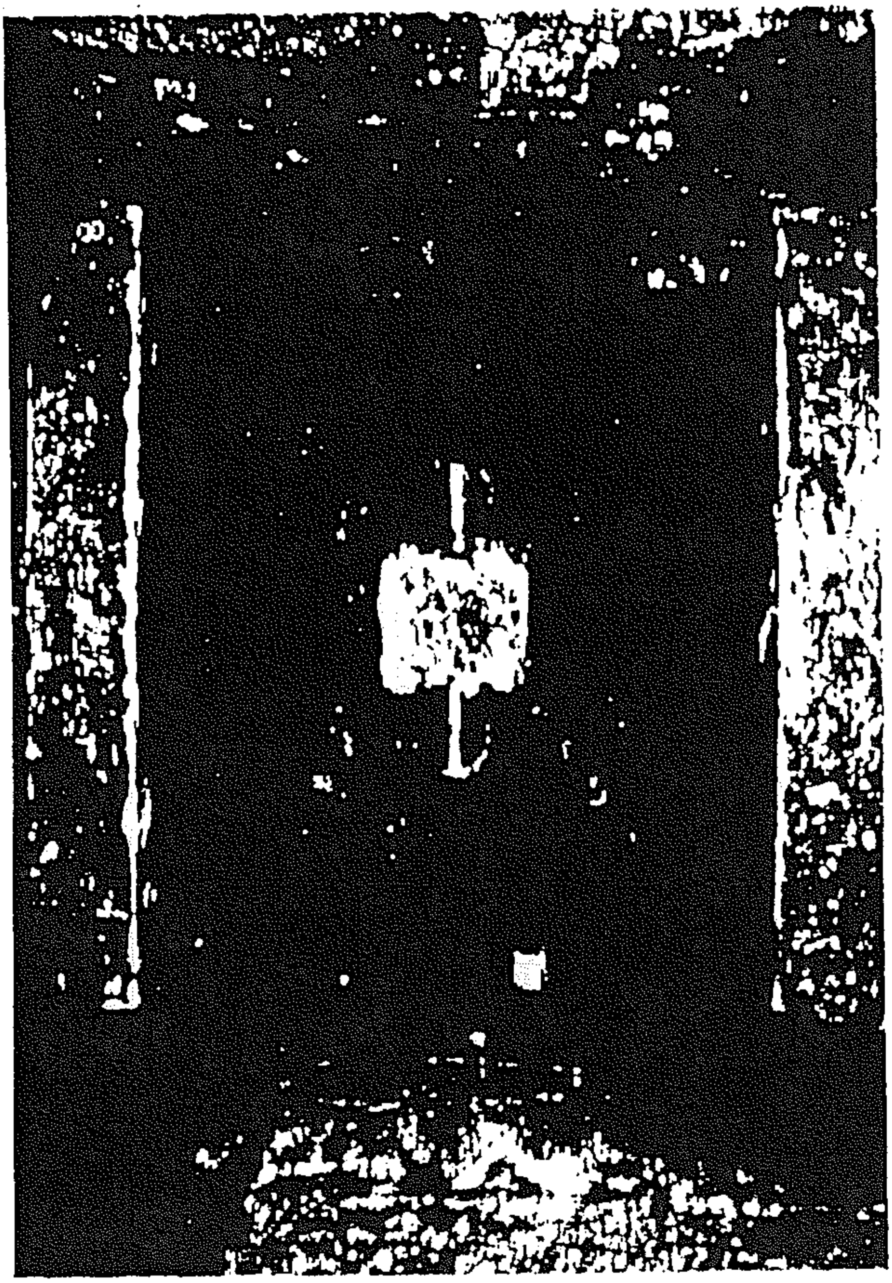
DISCHARGE EFFICIENCY OF NOBLE METALS IN EACH POURING

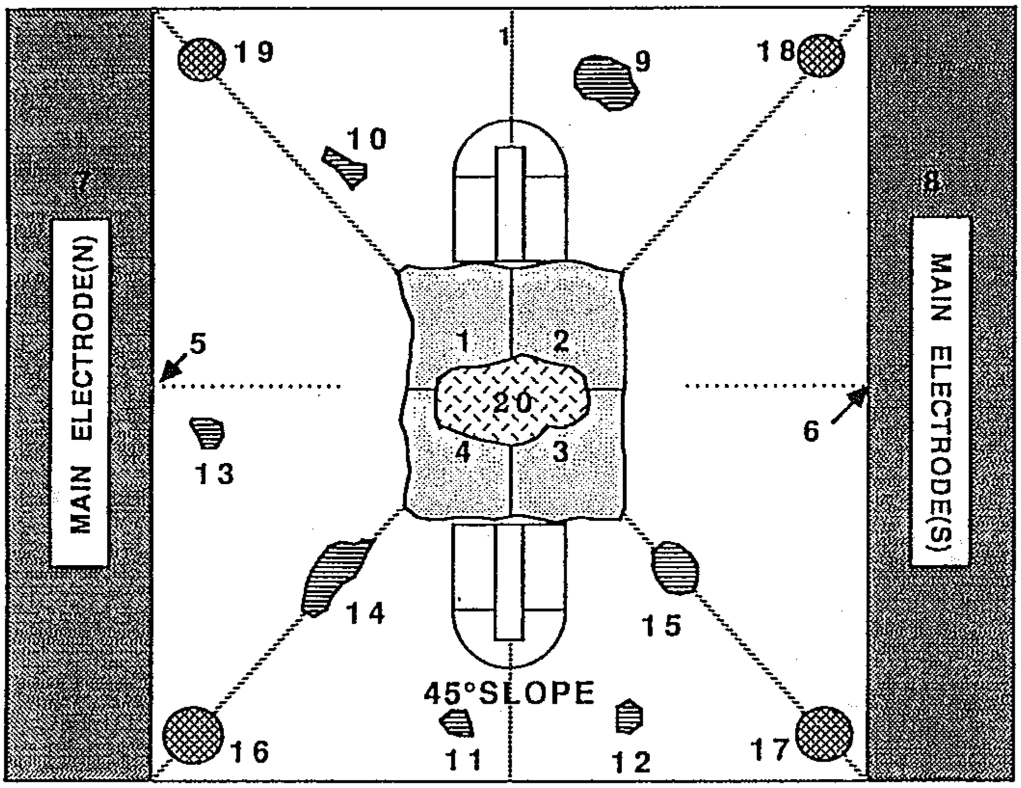
BALANCE OF NOBLE METALS IN MELTER (23rd CAMPAIGN)

NOBLE METALS / ITEM	RuO_2	PdO	Rh_2O_3
FEED AMOUNT (kg)	81.0	36.6	9.7
DISCHARGE AMOUNT (kg)	79.9	36.4	9.9
ACCUMULATED AMOUNT (kg)	1.2	0.1	-0.2
DISCHARGE EFFICIENCY (%)	98.6	99.5	102.3



BALANCE OF NOBLE METALS IN MELTER

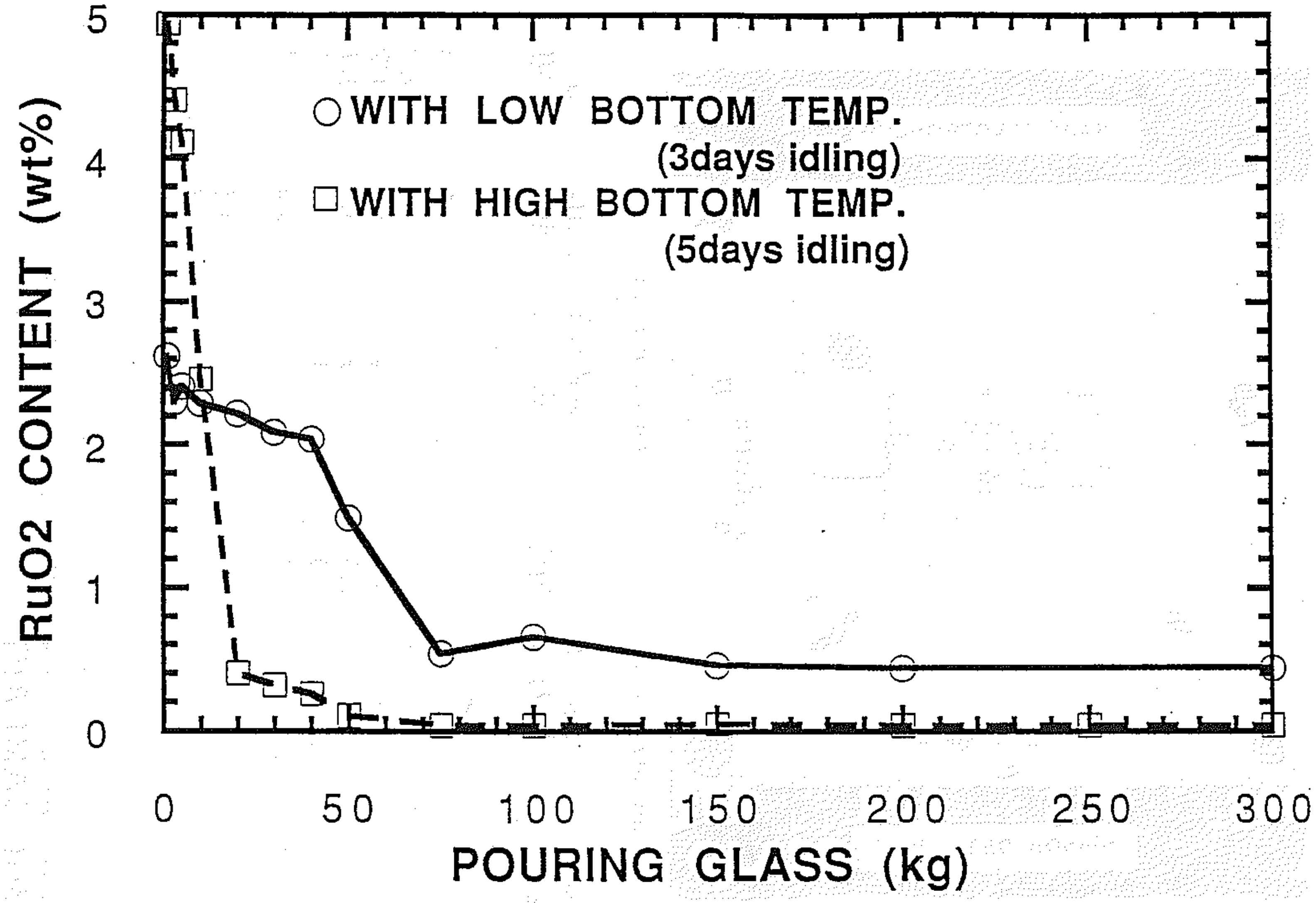




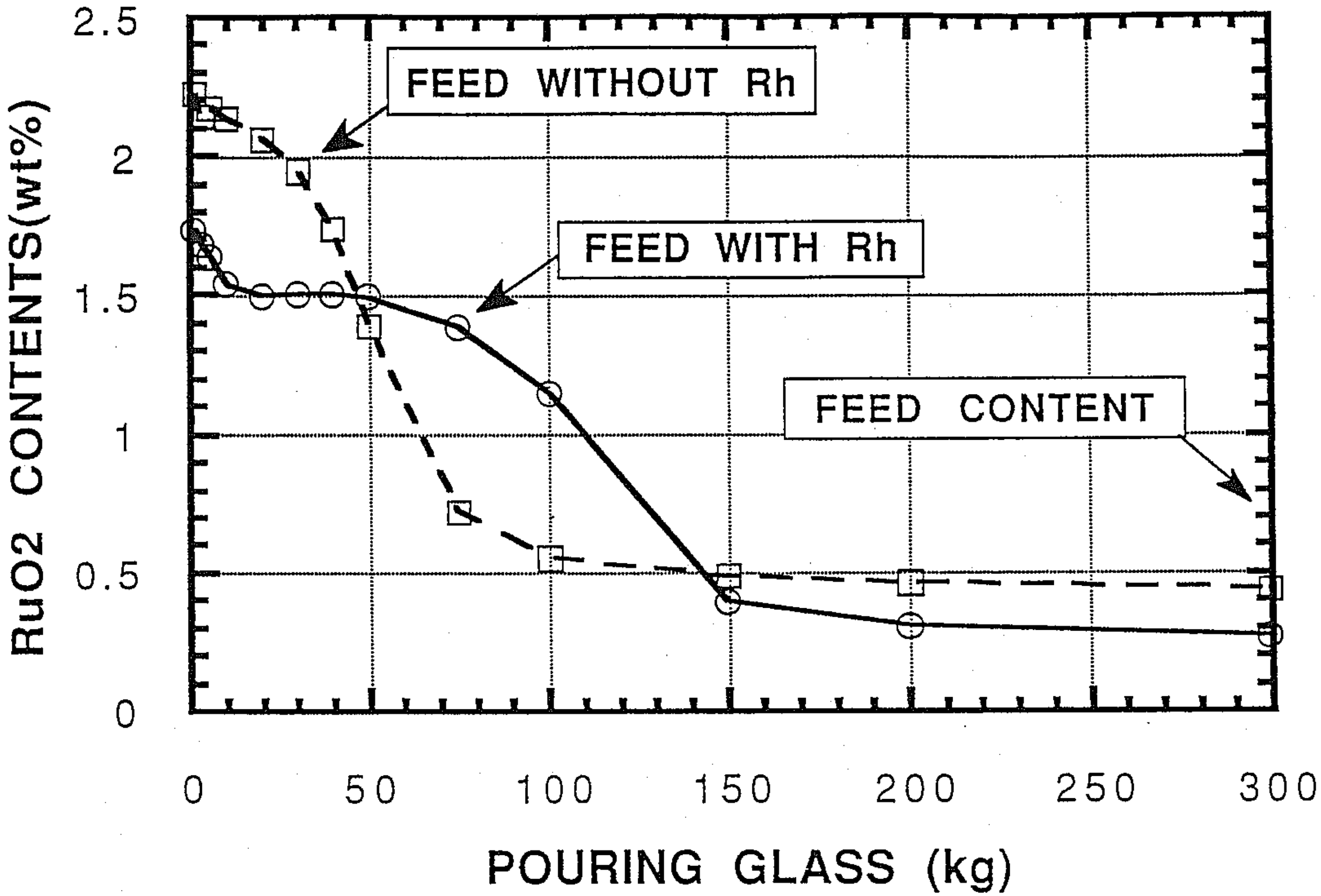
* 20 show the glass dropped from the vertical side of main electrodes

SAMPLE NO.	CONTENTS(%) RuO_2	PdO	SAMPLE NO.	CONTENTS(%) RuO_2	PdO
1	2.9	1.0	11	3.5	1.3
2	4.0	1.1	12	3.3	1.3
3	3.7	1.2	13	1.8	0.2
4	3.6	0.9	14	5.7	0.2
5	6.2	2.7	15	3.5	0.7
6	6.6	3.0	16	1.2	0.3
7	1.4	0.5	17	0.8	0.3
8	1.7	0.5	18	1.1	0.4
9	4.6	1.7	19	0.6	0.3
10	1.7	0.7	20	16.8	0.6

ACCUMULATION OF NOBLE METALS IN MELTER (23rd CAMPAIGN)



EFFECT OF OPERATION METHOD ON POURING AFTER IDLING



EFFECT OF Rh ON DISCHARGE CONTENTS

CONCLUSIONS

- Evaluated operational method controlling bottom glass temperature be most reliable for TVF
  - in normal operation
  - in idling operation

- Should keep melter bottom at low temperature to avoid accumulation of noble metals and to facilitate discharge

- Evaluated Rh would not have influence on discharge of other noble metals in this operation method

2-4 Off-gas Treatment Process

PNC TN8100 91-030

OFF-GAS TREATMENT PROCESS

The Tenth Annual KfK-PNC Meeting on Cooperation
in High-Level Waste Management

November 18 - 22, 1990

POWER REACTOR AND NUCLEAR FUEL
DEVELOPMENT CORPORATION

PNC

Off-gas entrainment in bubbling operation of melter

objectives of bubblling operation ;

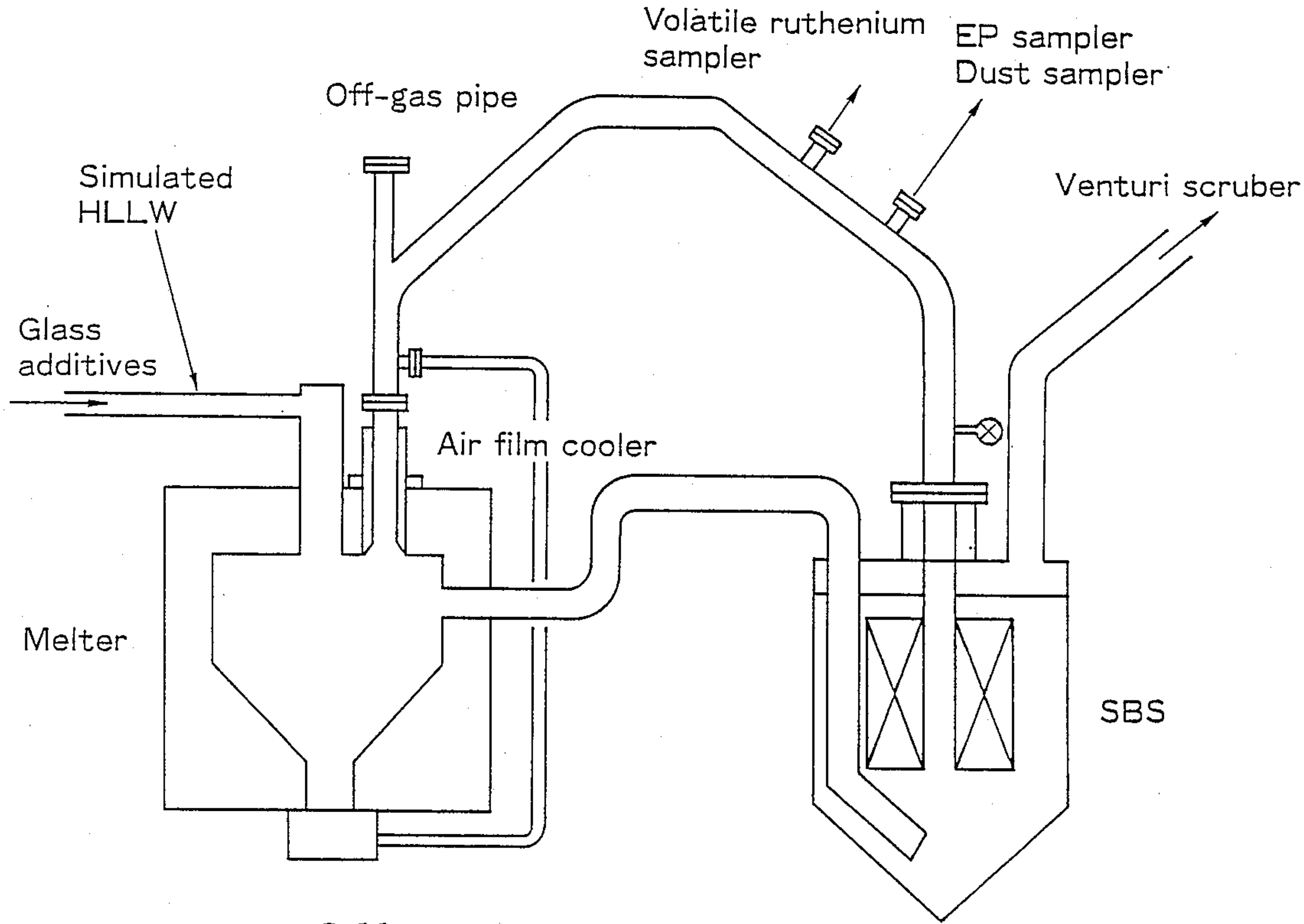
- increase of melting rate
- agitate the noble metal precipitation
- study the possibility of adopting bubbling in TVF operation within the license taken

anticipated effect on off-gas treatment;

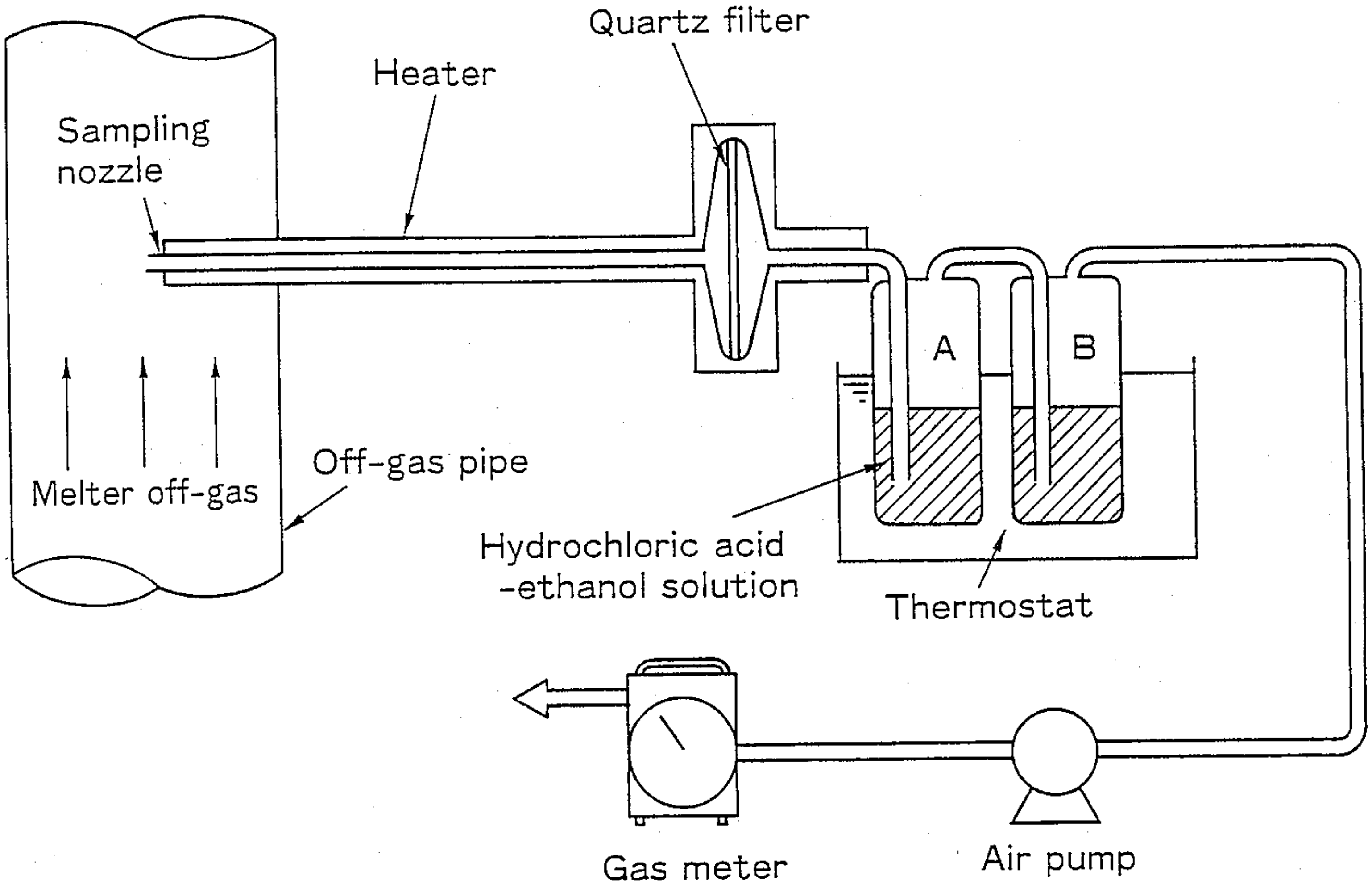
- increase in off-gas entrainment from melter for Ru, Cs, aerosols

Off-gas treating experiment (the 21st campain)

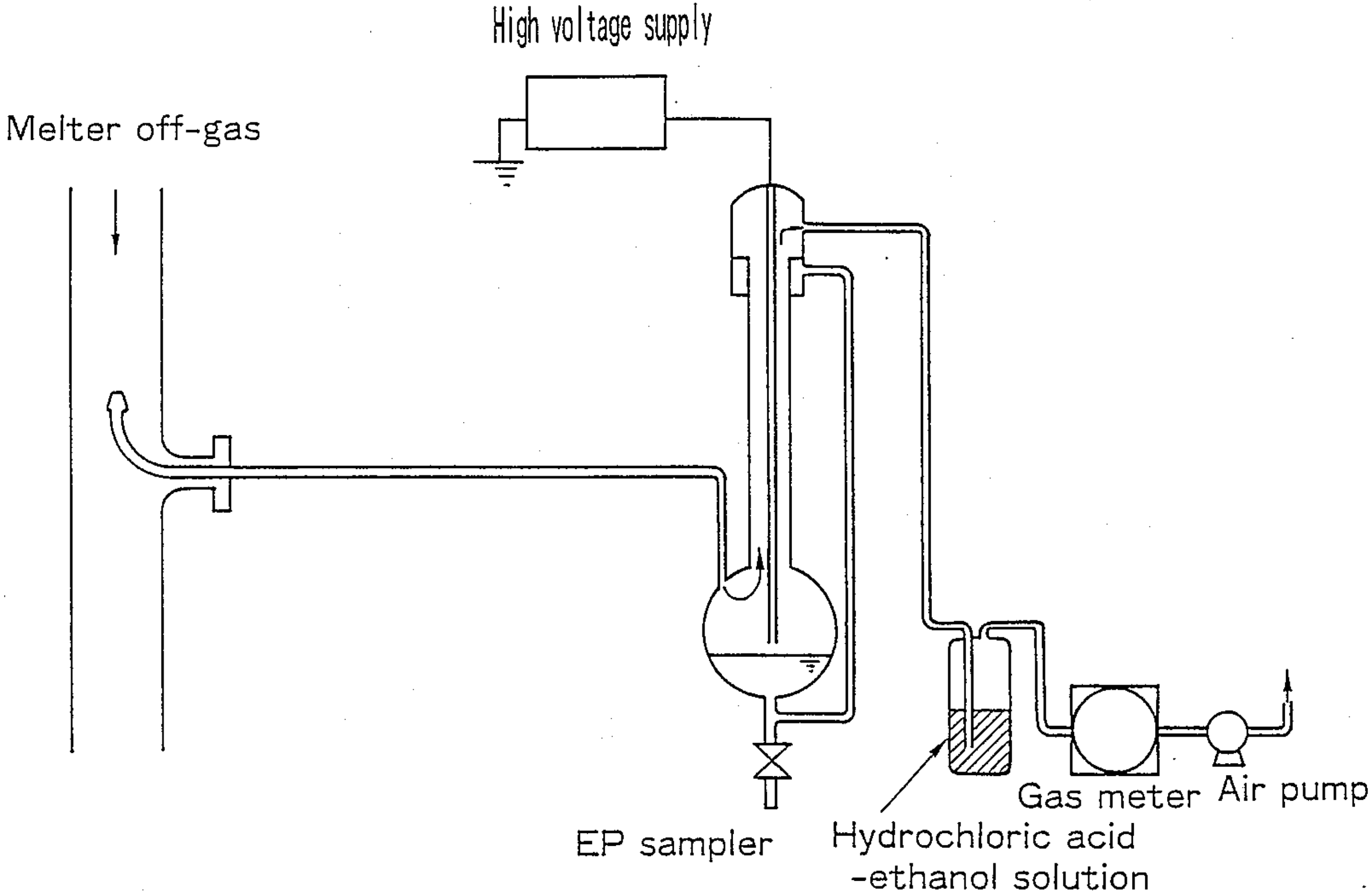
objective	Evaluation on the influence of the bubbling in the melter on the off-gas entrainment
objective element	1. ruthenium 2. cesium 3. strontium 4. dust
evaluated items	1. Influence by the composition of the bubbling gas Air or N_2 gas 2. Influence by the flow rate of the bubbling gas 400 ℓ/h or 200 ℓ/h
sampling method	Off-gas sampling at the outlet of the melter 1. volatile ruthenium sampler 2. electrostatic precipitator (EP) sampler 3. dust sampler
evaluation method	Measurement of the entrainment ratio or volatilizing ratio entrainment ratio = (entrained quantity into the off-gas) /(feed into the melter)×100 = (captured amount by each sampler × melter off-gas) /(feed into the melter × sampling gas flow) ×100



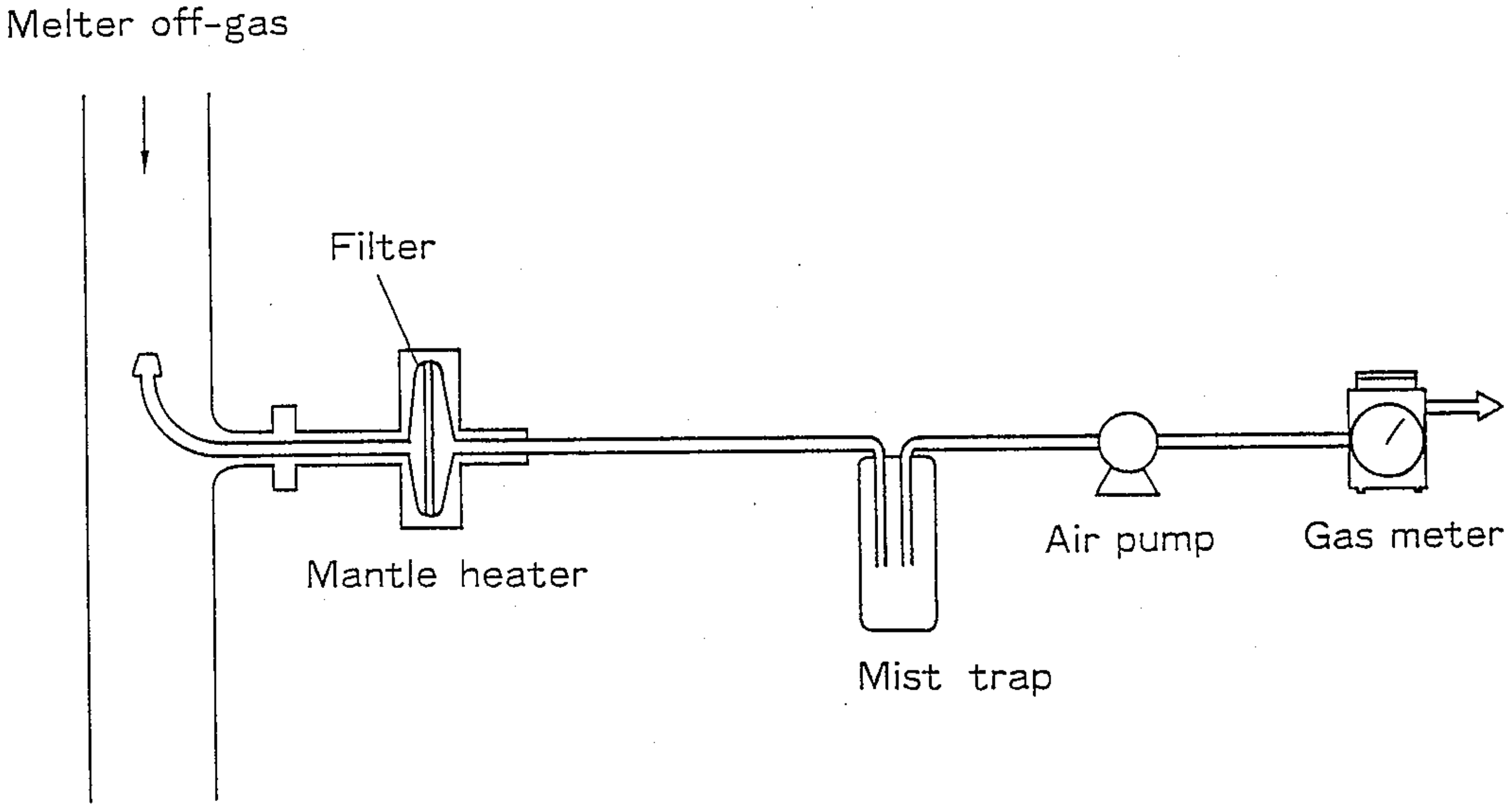
Off-gas sampling points



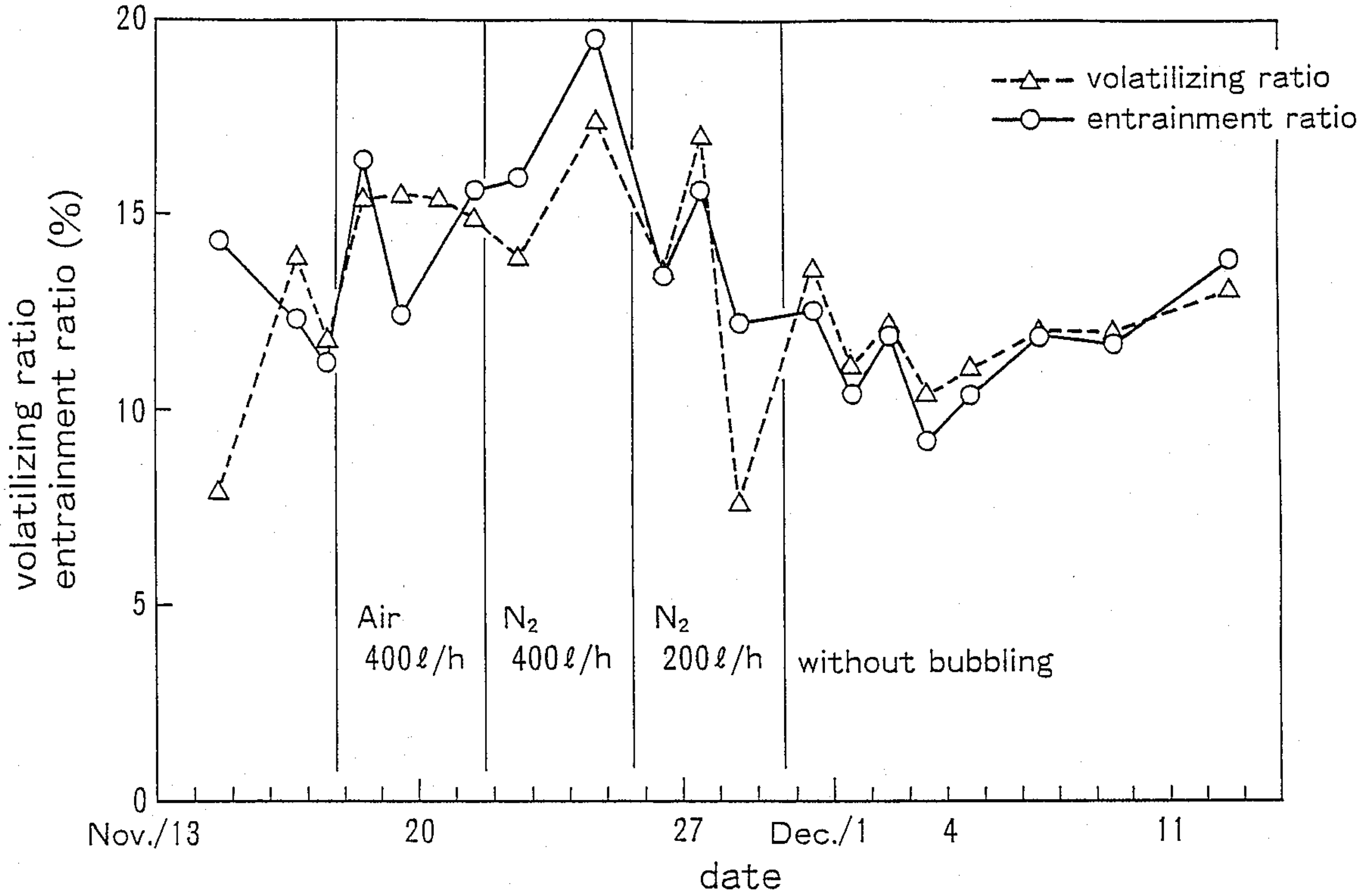
Volatile ruthenium sampler



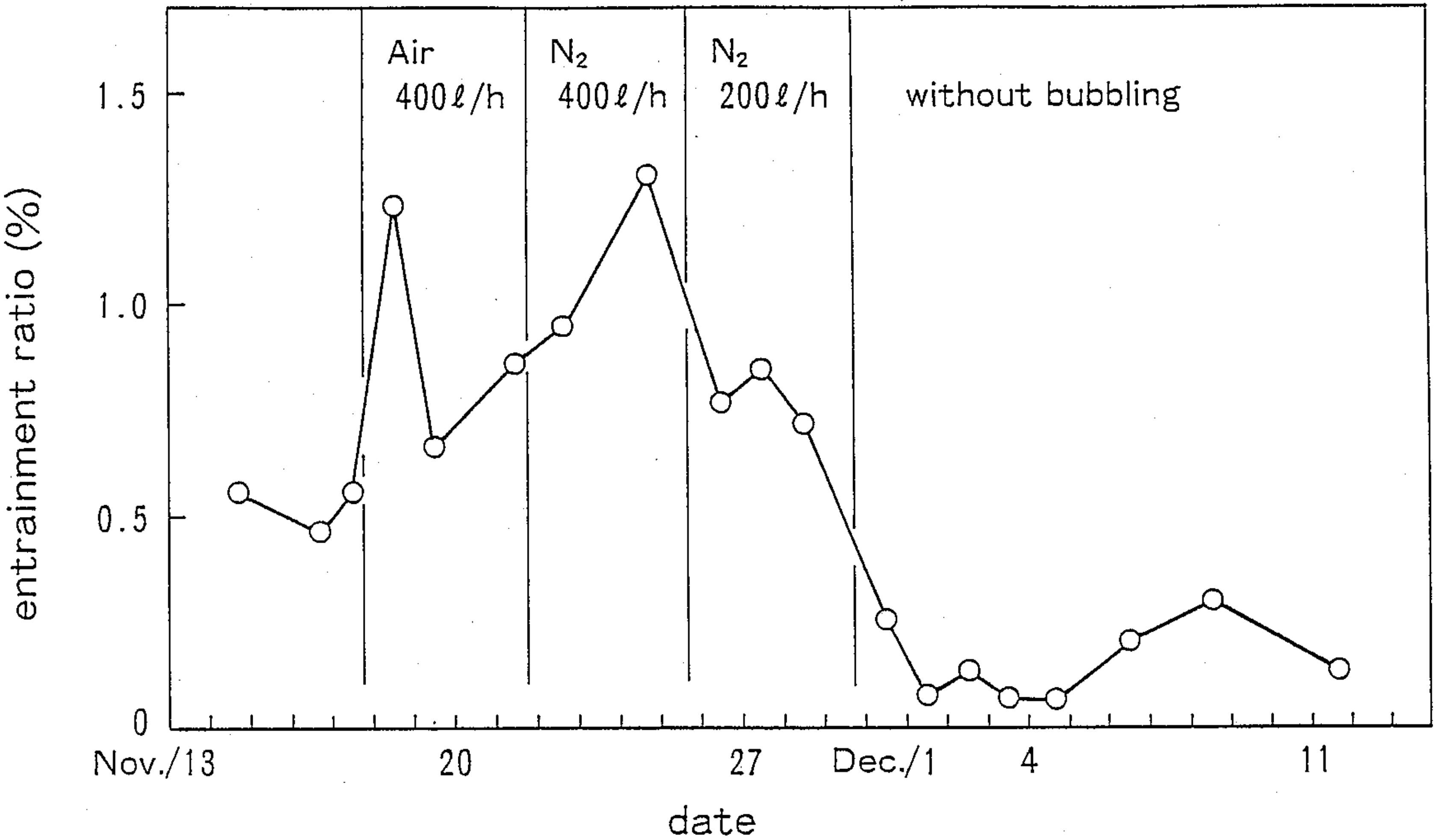
Electrostatic Precipitator (EP) Sampler



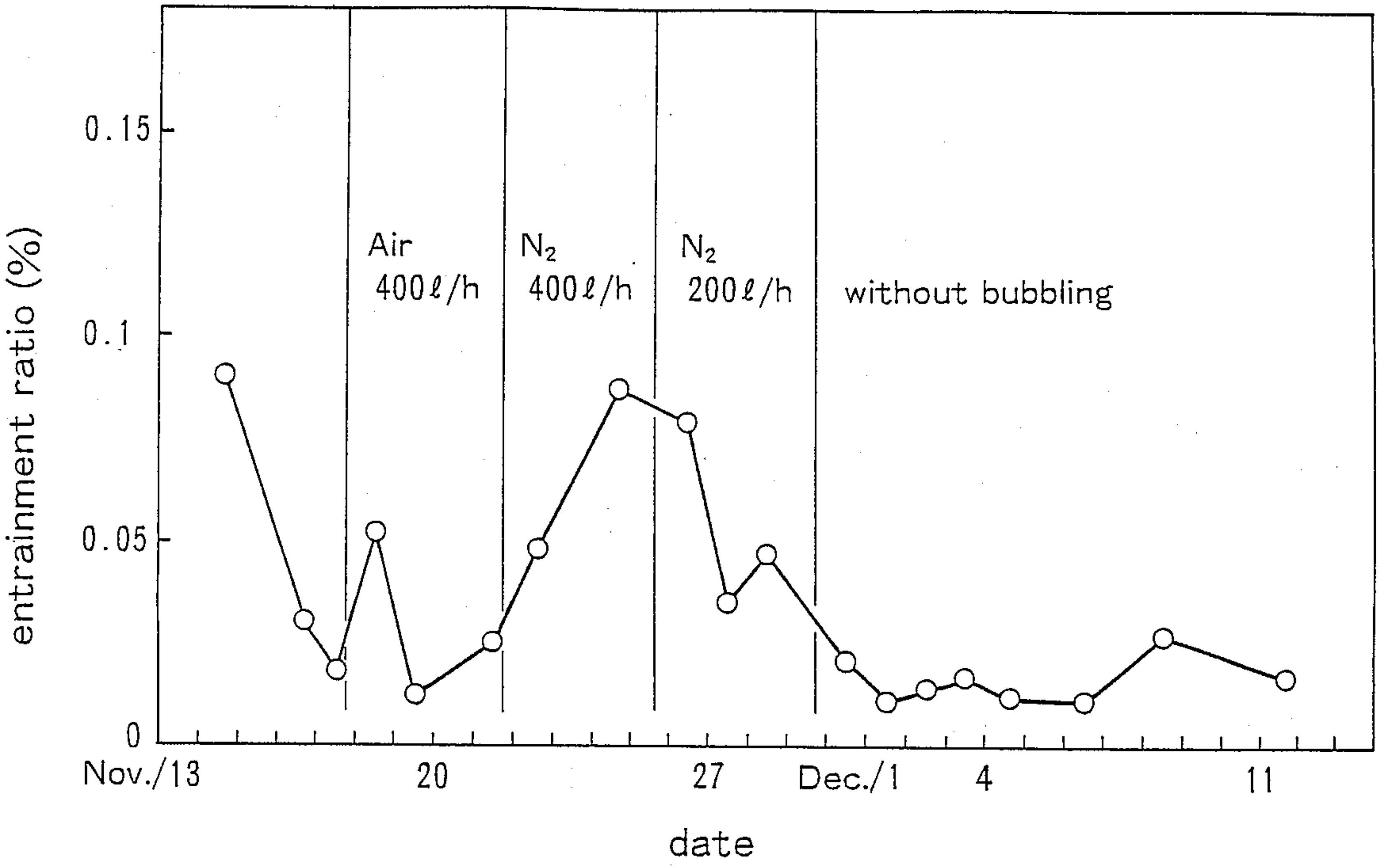
Dust sampler



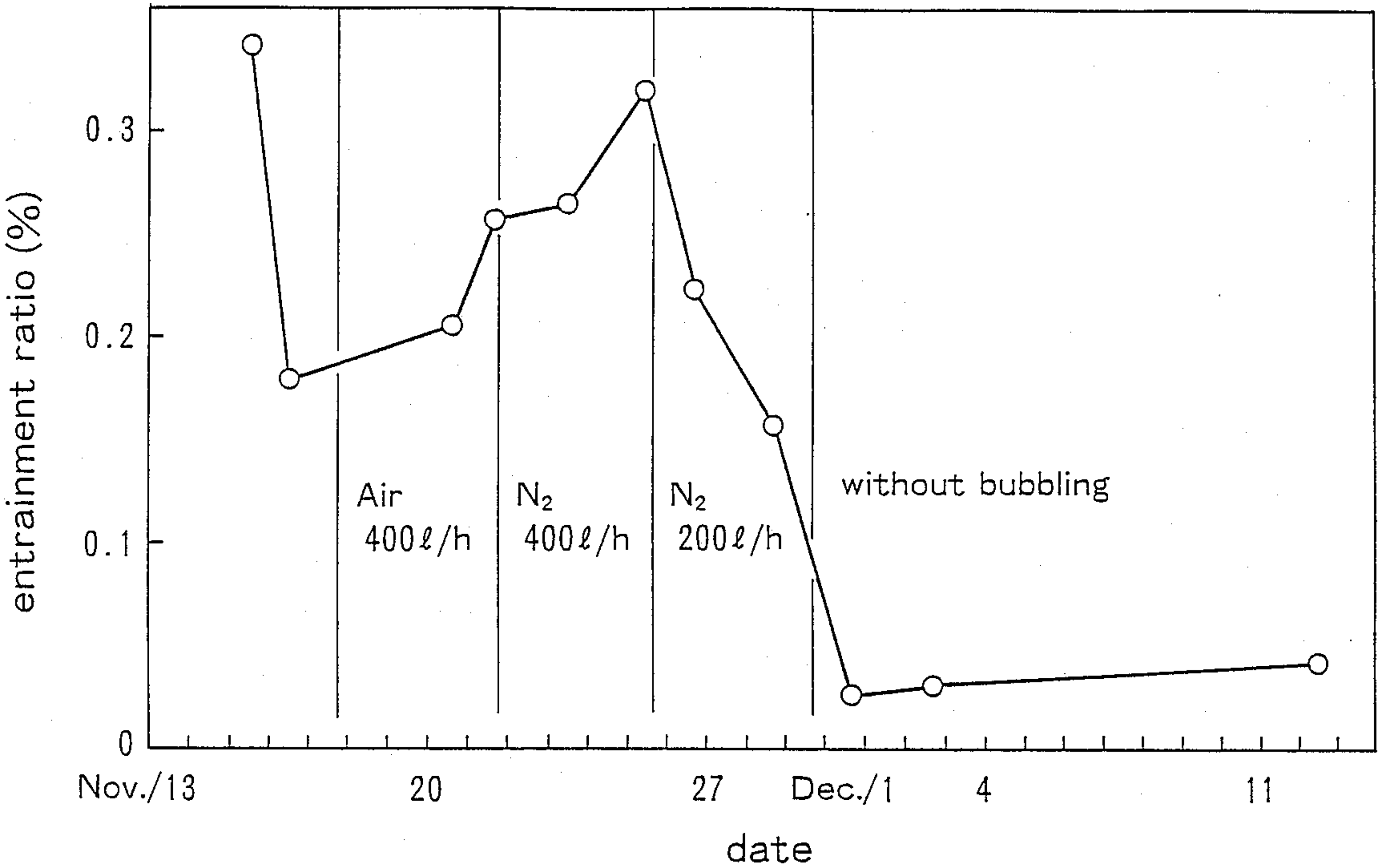
Entrainment ratio and volatilizing ratio for ruthenium



Entrainment ratio for cesium



Entrainment ratio for strontium

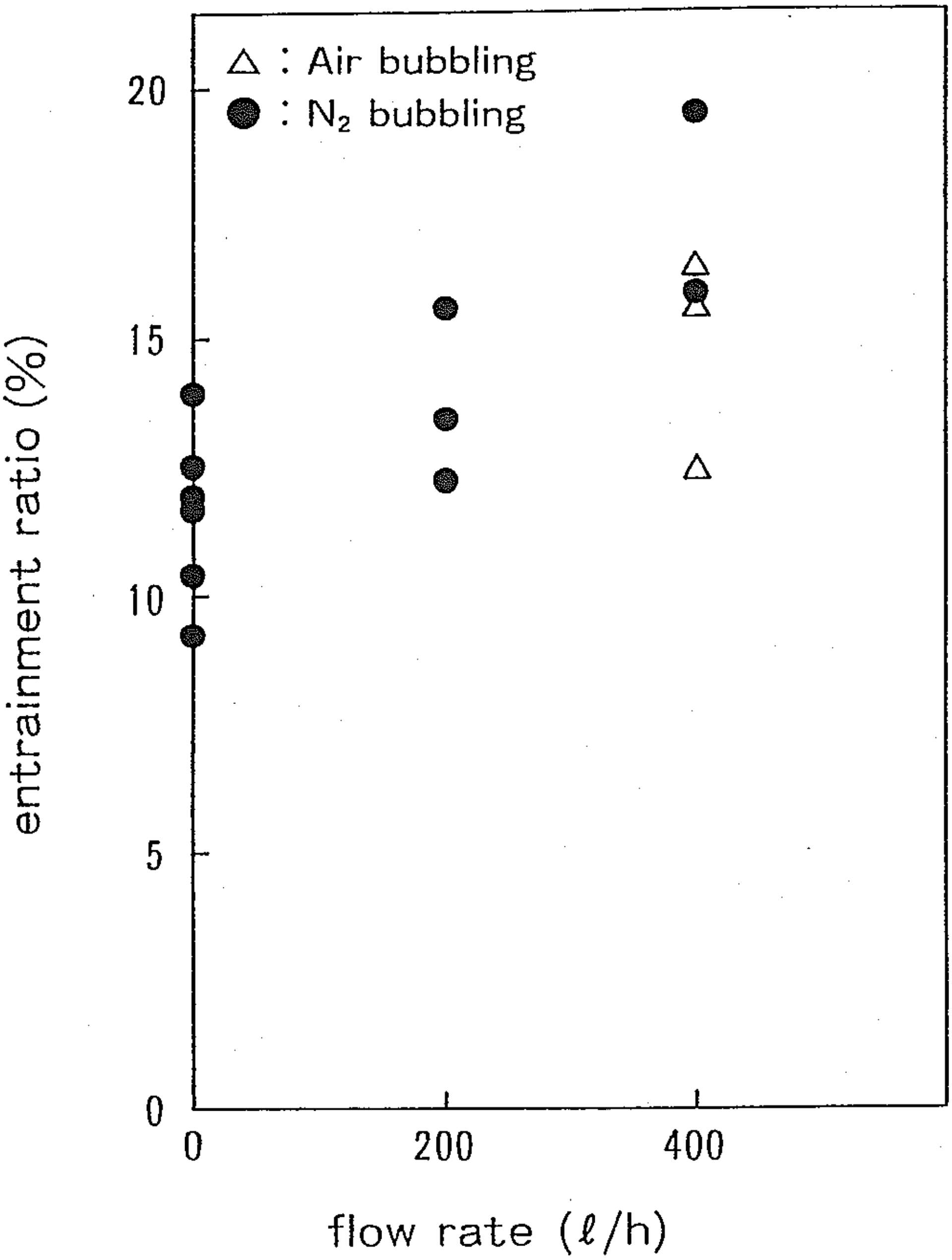


Entrainment ratio for dust

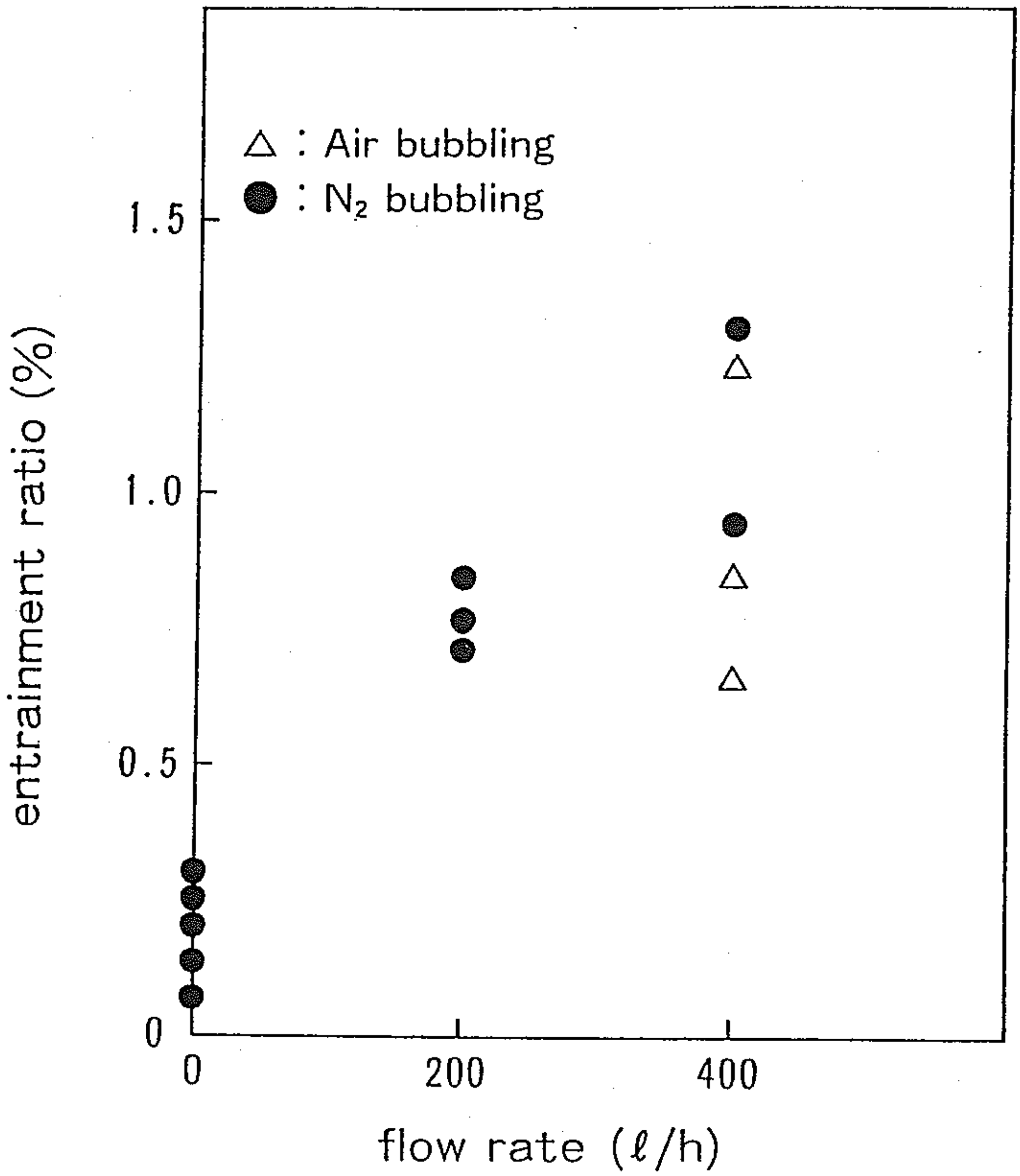
Composition of bubbling gas and entrainment ratio

Sampling No.	Composition of bubbling gas	entrainment ratio (%)			
		Ru	Cs	Sr	dust
EP-4	Air	16.4	1.23	0.052	
-5		12.4	0.66	0.012	
-6		15.6	0.85	0.025	
D-3					0.21
-4					0.26
EP-7	N_2	15.9	0.94	0.048	
-8		19.5	1.31	0.087	
D-5					0.26
-6					0.32

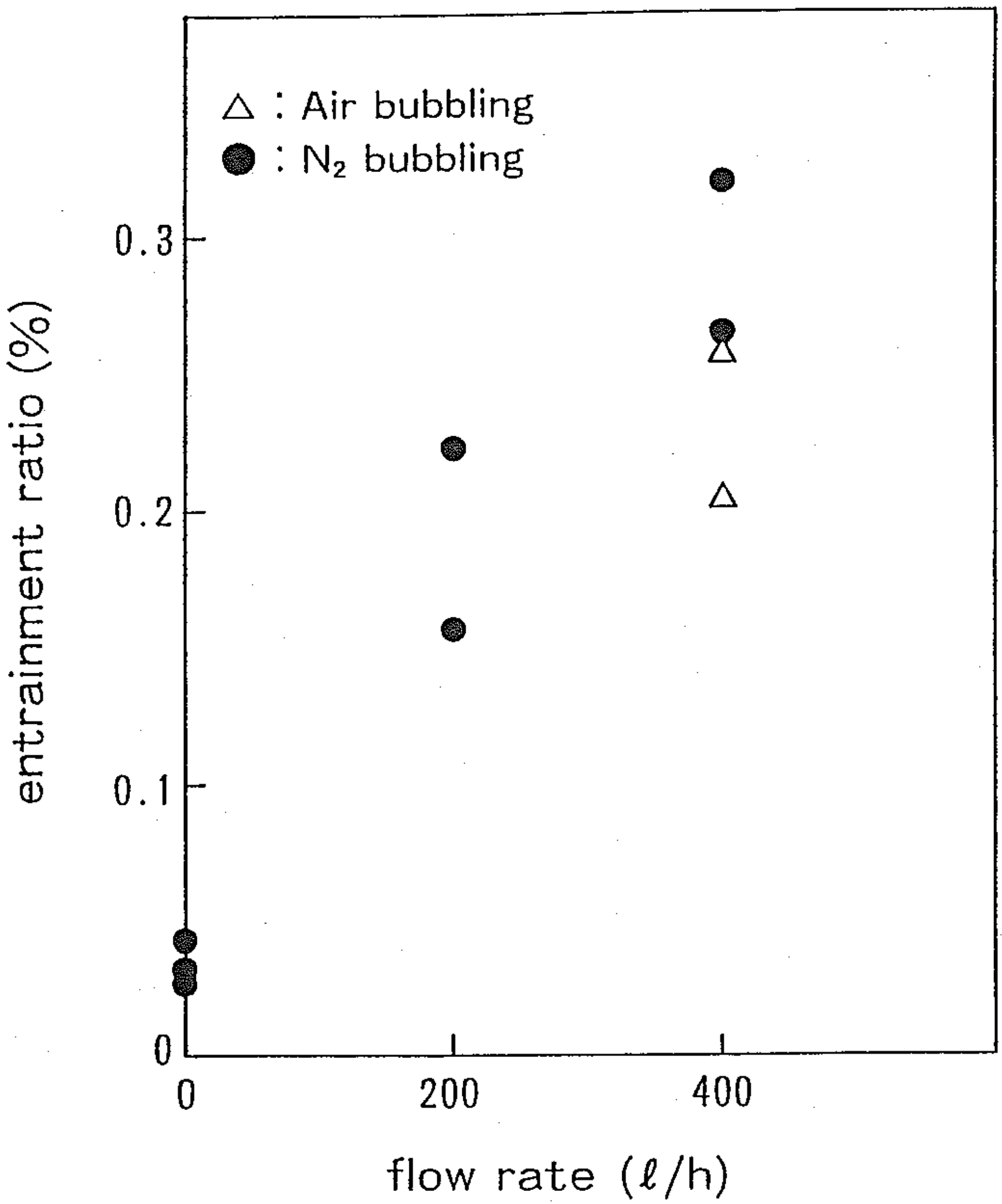
EP — EP sampler
D — dust sampler



Effect of bubbling gas flow rate on Ru entrainment ratio



Effect of bubbling gas flow rate on Cs entrainment ratio



Effect of bubbling gas flow rate on dust entrainment ratio

Conclusion

1. The entrainment ratios of Ru, Cs, Sr and dust increased with the bubbling flow rate. No data, however, was beyond the design set value of TVF.

2. Significant difference was not recognized for entrainment ratio between air and N_2-gas used in bubbling.

3. Ru entrainment ratio with bubbling was close to the design set value of TVF. To adopt the bubbling, it is necessary to consider the fluctuation of the flow rate of the bubbling gas in safty analysis.

Entrainment test for semi-volatile elements

objectives;

- to study effect of temperature on volatilization
- to make sure the variation in melter operation does not increase entrainment from the melter

elements to study ; Tc, Ru, Se and Sr

radioactive tracers; ^{103}Ru, ^{99m}Tc, ^{75}Se, ^{85}Sr

temperature ; 300～800 ℃

simulated HLLW ; SW-30 waste

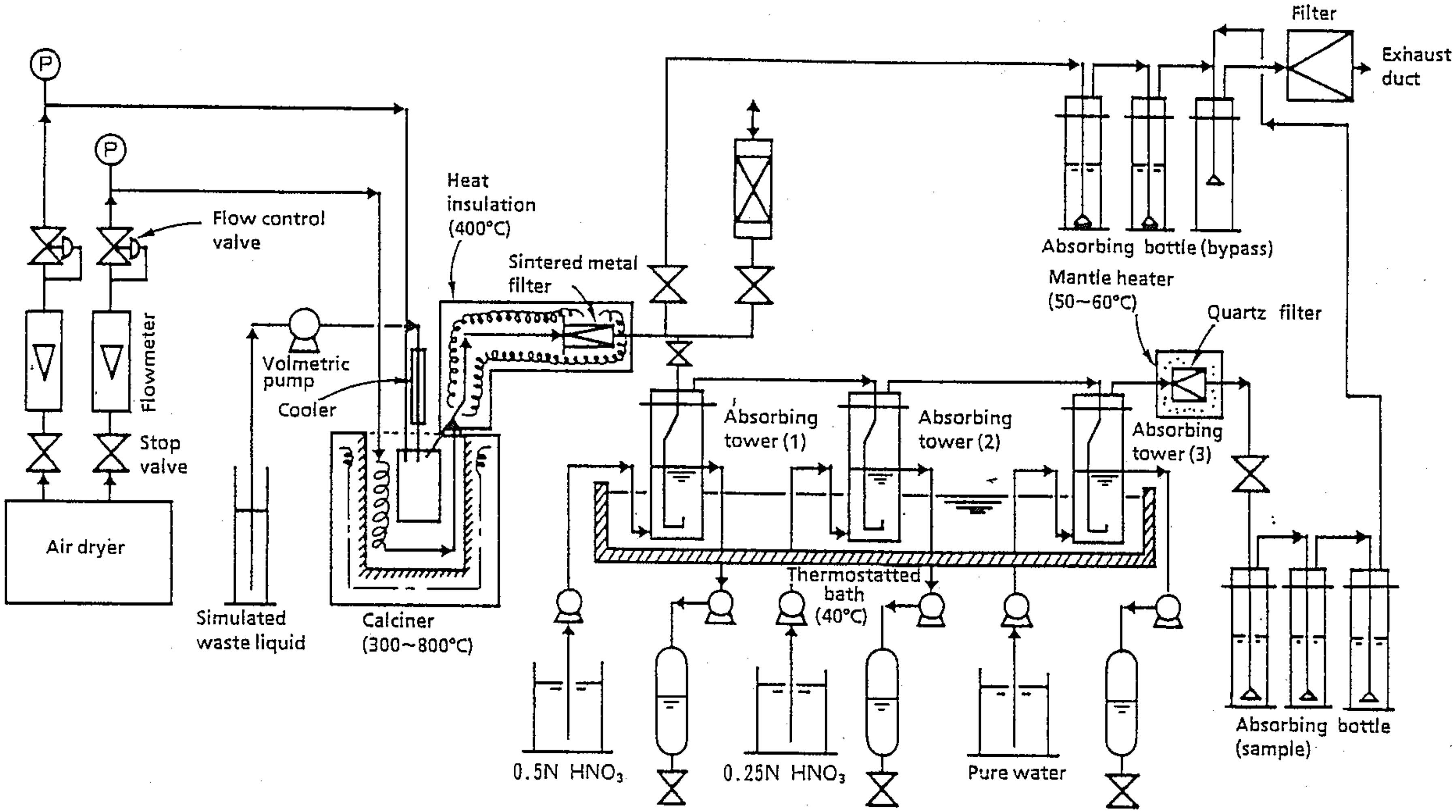


Fig. 1 Outline of testing system for behavior of quasi-volatile elements

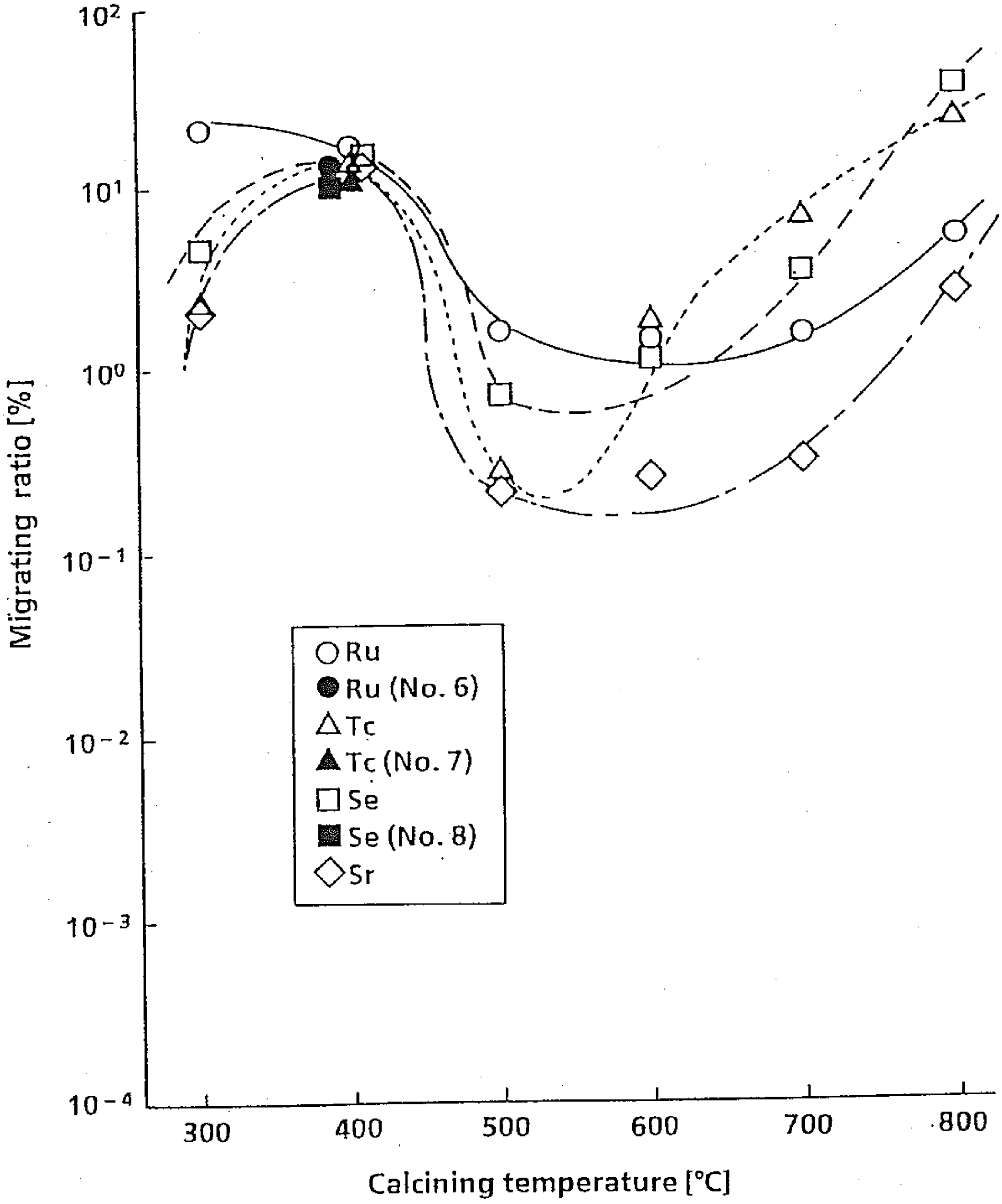


Fig. 5 Migrating ratio of Ru, Tc, Se, and Sr versus calcining temperature

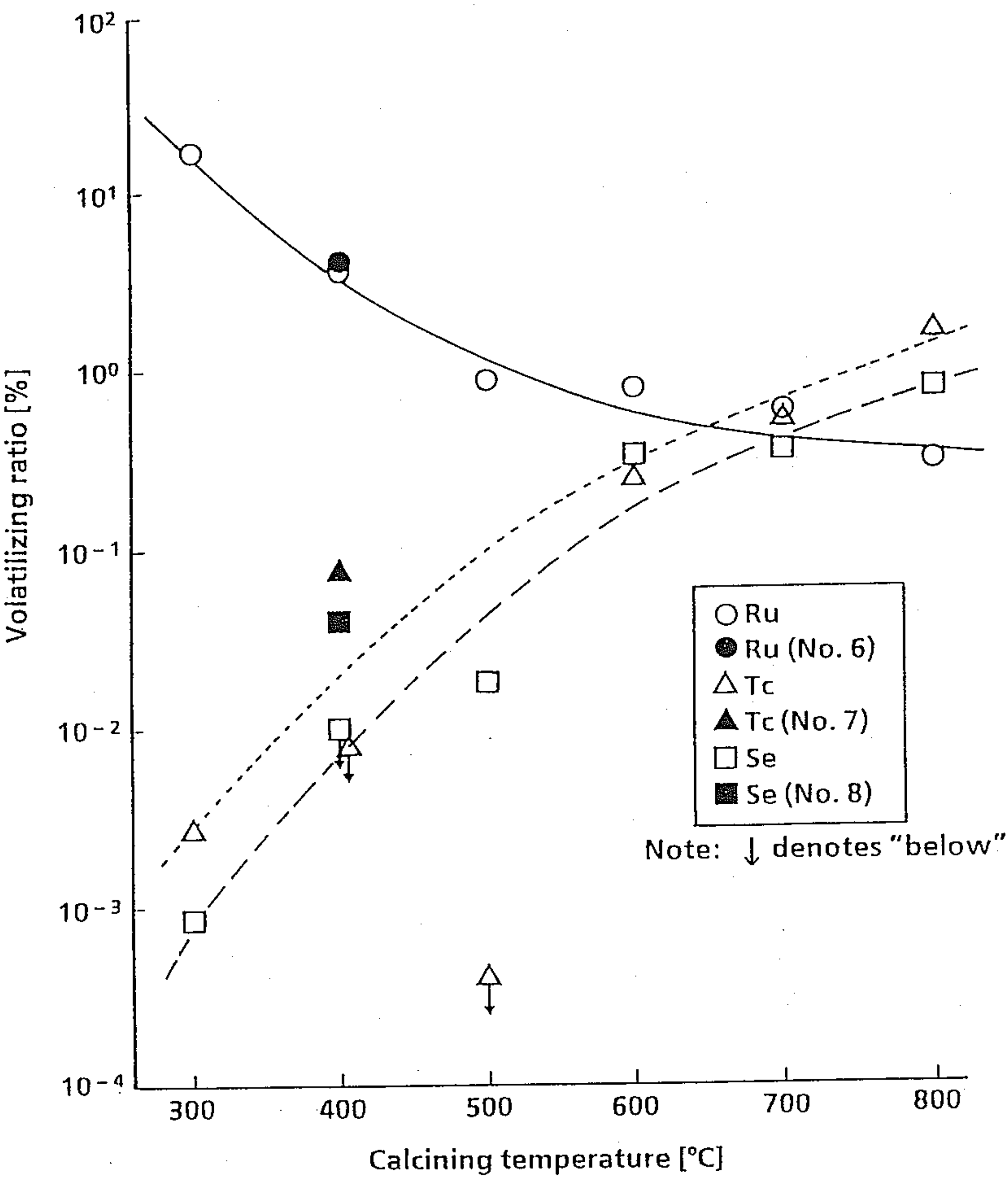


Fig. 6　Volatilizing ratio of Ru, Tc and Se versus calcining temperature

Future work

- labo-scale test to study bubbling effect
  - fabrication of apparatus was finished
  - effect of bubbling in calicination process and for molten glass in details
  - dependence of entrainment or volatilization on temperature and on flow rate

- absorption behavior of Tc into water

2-5 Development of Three Dimensional Thermal Fluidic Analysis Code for Electric Glass Melte

PNC TN8100 91-030

DEVELOPMENT OF THREE DIMENSIONAL THERMAL FLUIDIC ANALYSIS CODE FOR ELECTRIC GLASS MELTER

The Tenth Annual KfK-PNC Meeting on Cooperation in High-Level Waste Management

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POWER REACTOR AND NUCLEAR FUEL DEVELOPMENT CORPORATION

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Introduction

① Outline of analysis code.

② Presentation of calculation example.

③ Comparison between calculation and operation.

④ Modification of analysis code.

⑤ Plane of future.

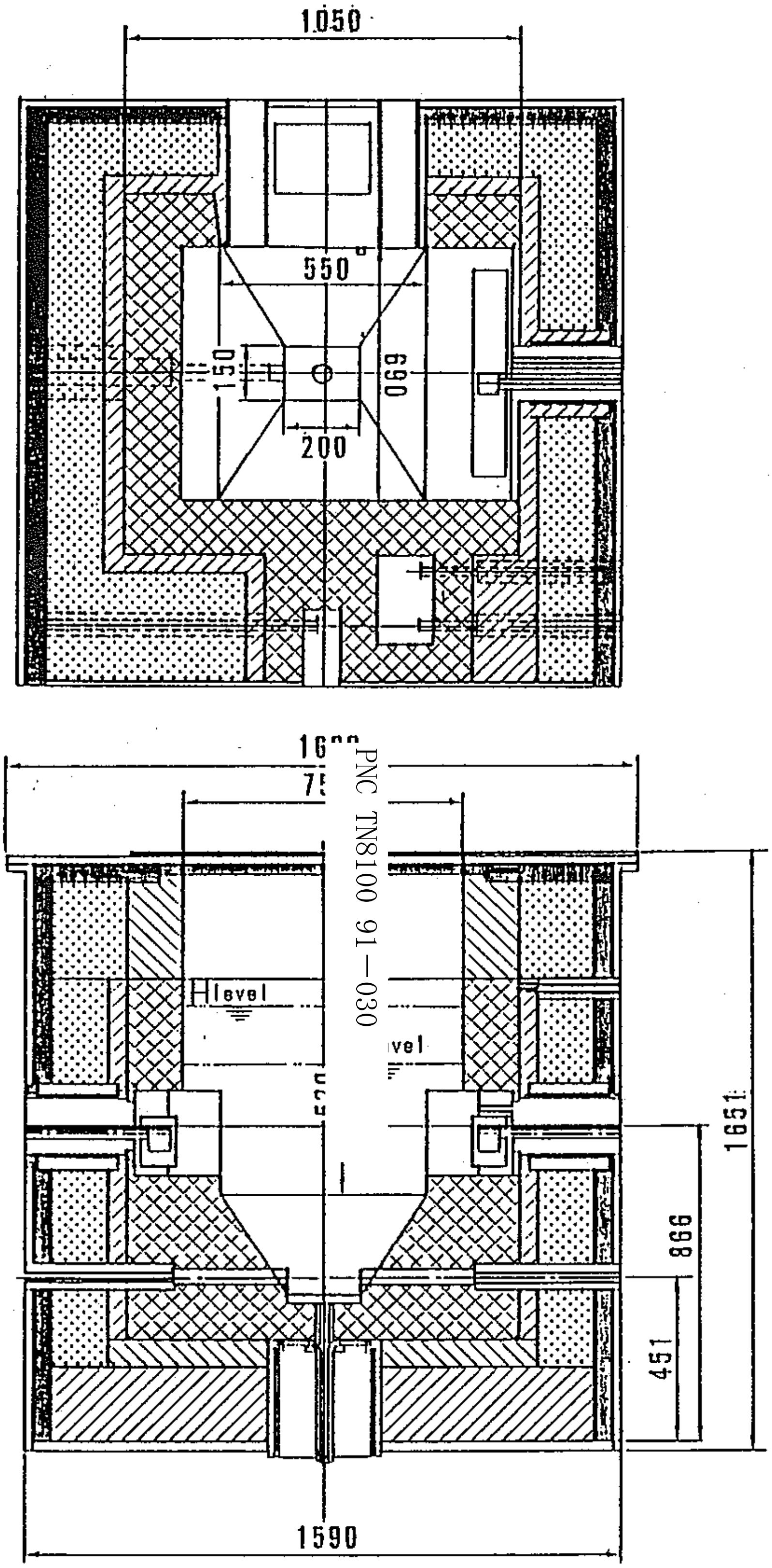
The development objectives of analysis code are :

① To establish the proper operational condition of the melter

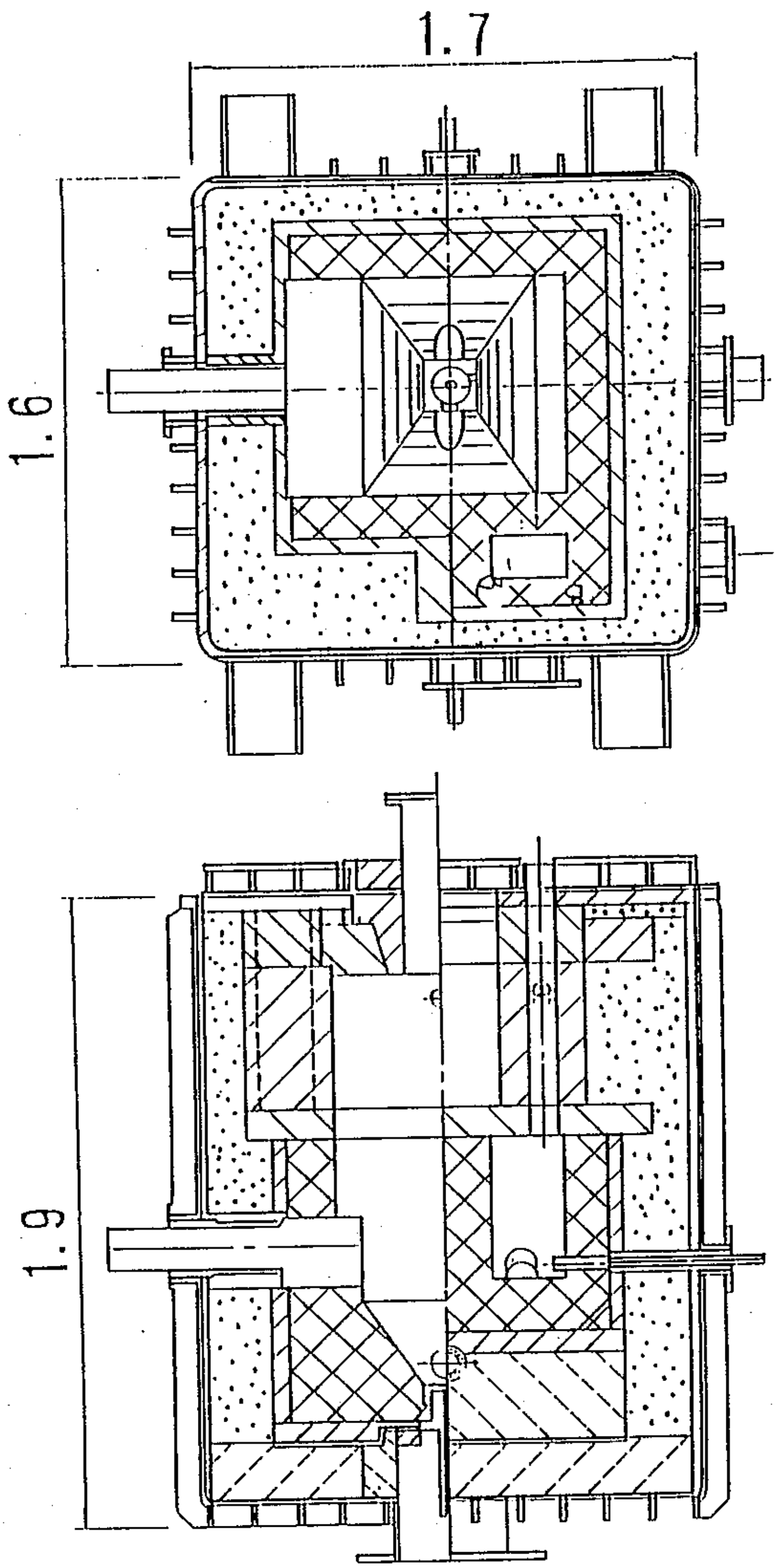
② To understand the thermal and flow profile of melter .

Therefore, we need to calculate the temperature distribution in the melter and flow profile of molten glass.

This analysis code can calculate temperaturein melter, electric potential, heat generation etc..



Cross Sectional view of Advanced B Melter [mm]
(Surface area : 0.53 [m^2])



Cross Sectional view of Mock-up 3 Melter [m]
(Surface area : 0.6 [m^2])

The characteristics of analysis code

① This code is applicafle to the complex structure of melter.

② This code can calaulate consistently the temperature in melter, the electric potential and the velocity of molten glass.

③ Computational domaie is divided into internal domain in order to improve the calculated speed.

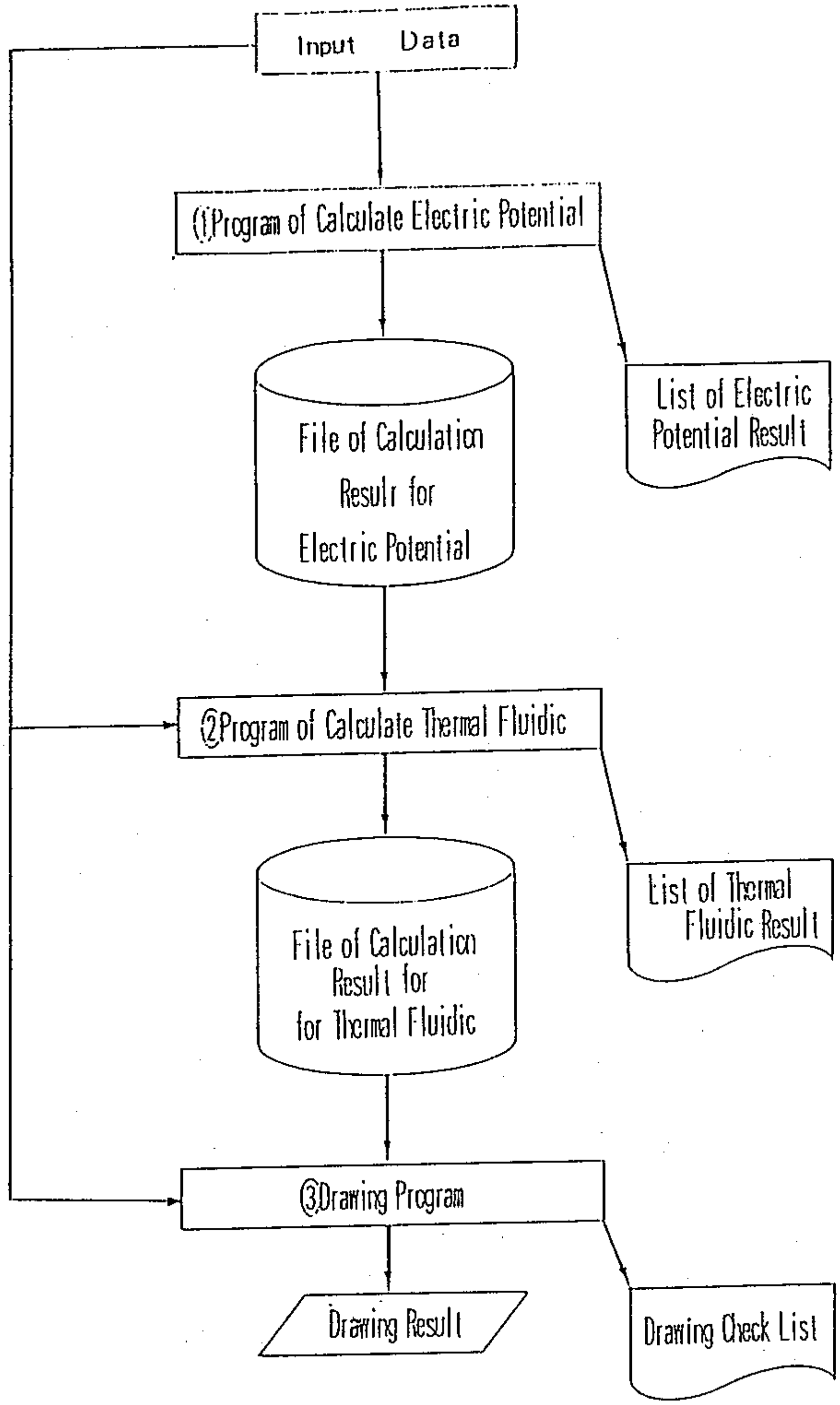
Basic Equation

Internal Domain	Extenal Domain
3 Direction component of Velocity Pressur Temperarure Electric Potential	Temperature
・Electric Potential Distribution $\frac{\partial^2 \phi}{\partial x^2} \div \frac{\partial^2 \phi}{\partial y^2} \div \frac{\partial^2 \phi}{\partial z^2} = 0$; ϕ : Electric Potential	Electric Potential is not Calculated
・Energy Equation(Themal Diffusion Equation) $\rho \cdot C_p \left(\frac{\partial T}{\partial x} \div \frac{\partial T}{\partial y} \div \frac{\partial T}{\partial z} \right) = \lambda \left(\frac{\partial^2 T}{\partial x^2} \div \frac{\partial^2 T}{\partial y^2} \div \frac{\partial^2 T}{\partial z^2} \right) \div Q$ where C_p: Spacific Heat T:Temperature Q:Heat gneration λ:Themal Condition ρ:Density	$\lambda \left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right) \div Q = 0$
・Continuity Equation $\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \div \frac{\partial w}{\partial z} = 0$ ・Momentum Equation $\frac{\partial \psi}{\partial t} \div \frac{\partial \psi}{\partial x} \div \frac{\partial \psi}{\partial y} \div \frac{\partial \psi}{\partial z} = \nu^2 \left(\frac{\partial^2 \psi}{\partial x^2} \div \frac{\partial^2 \psi}{\partial y^2} \div \frac{\partial^2 \psi}{\partial z^2} \right) \div S\psi$ where t : Time ψ: Velocity Component $S\psi$: Generator terms	Flow is not Calculated

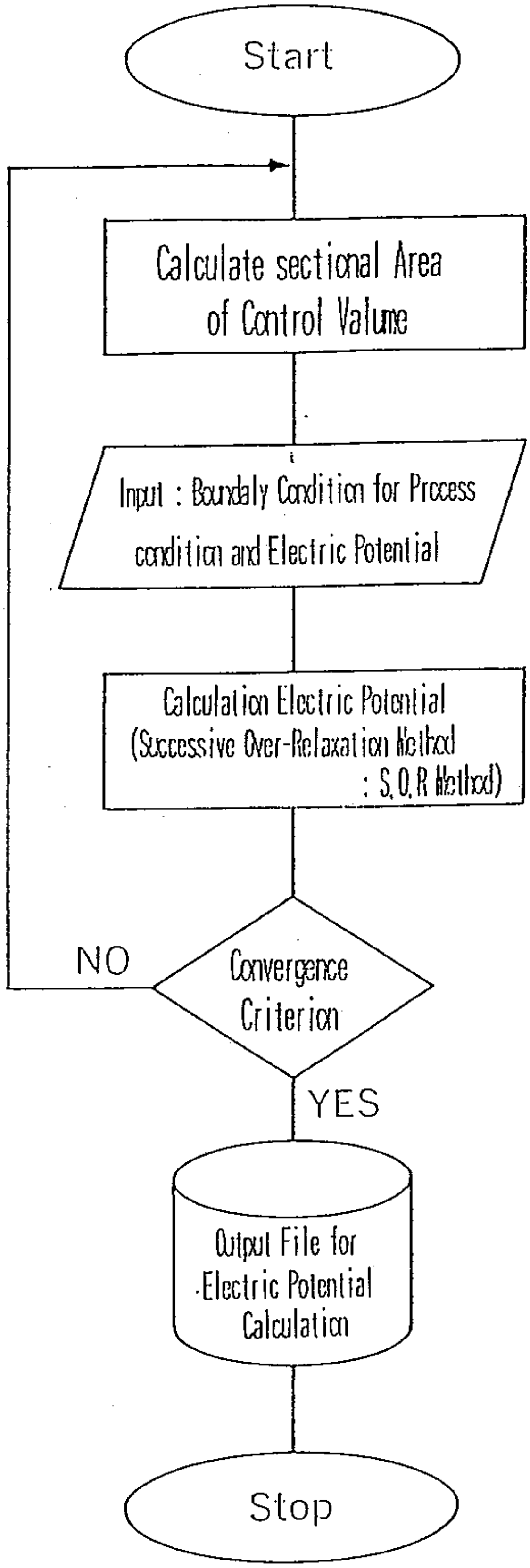
VALVES OF ψ AND $S\psi$

Equation	ψ	$S\psi$
continuty	1	0
x—momentum	u	$\partial p / \partial x$
y—momentum	v	$\partial p / \partial y$
z—momentum	w	$\partial p / \partial z + g \beta \Delta T$

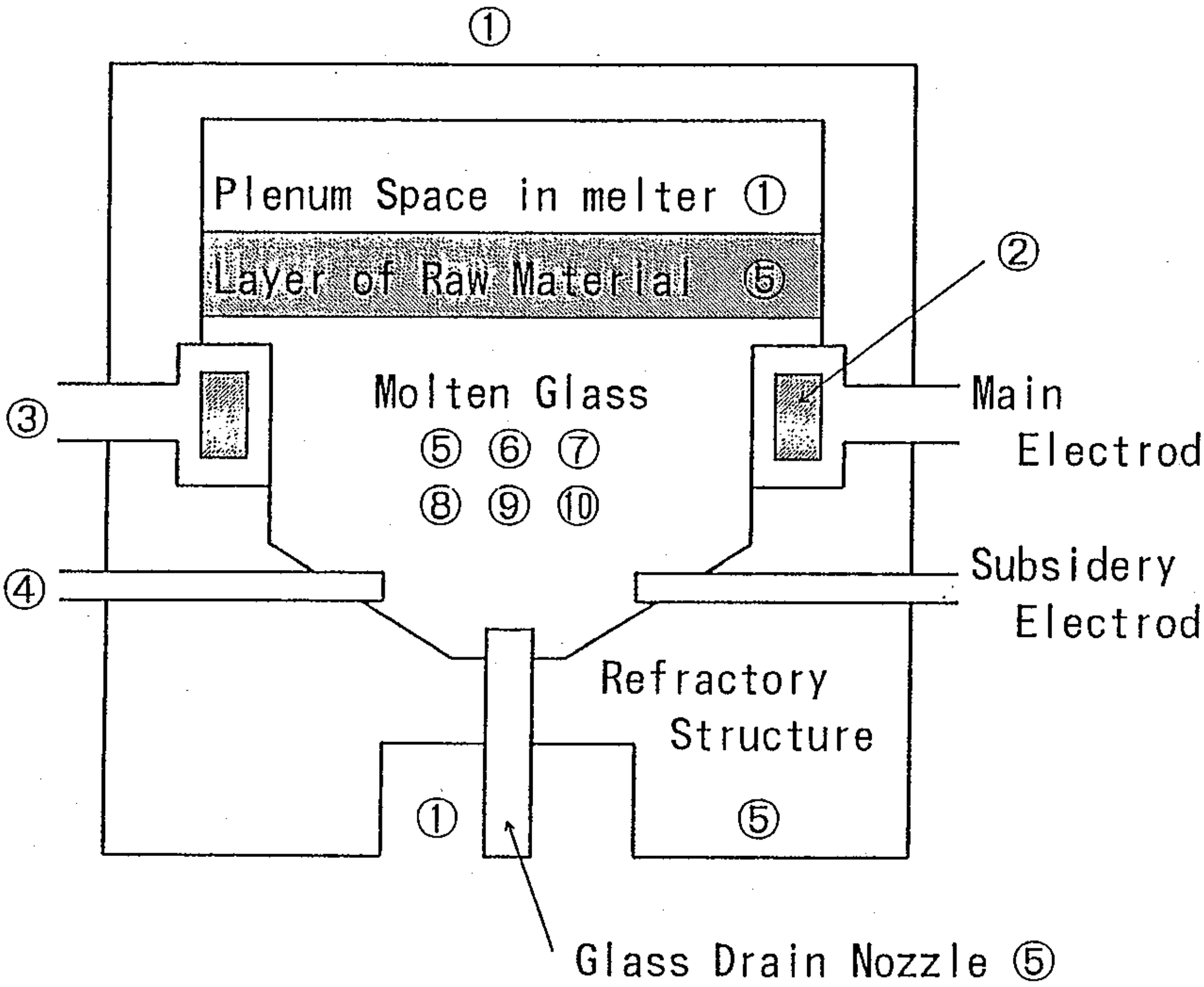
u, v, w : Velocity Component , g : Gravity Acceleration
ΔT : Temperature difference , p : pressure ,
β : Thermal coefficient of volumetric expansion



Program System

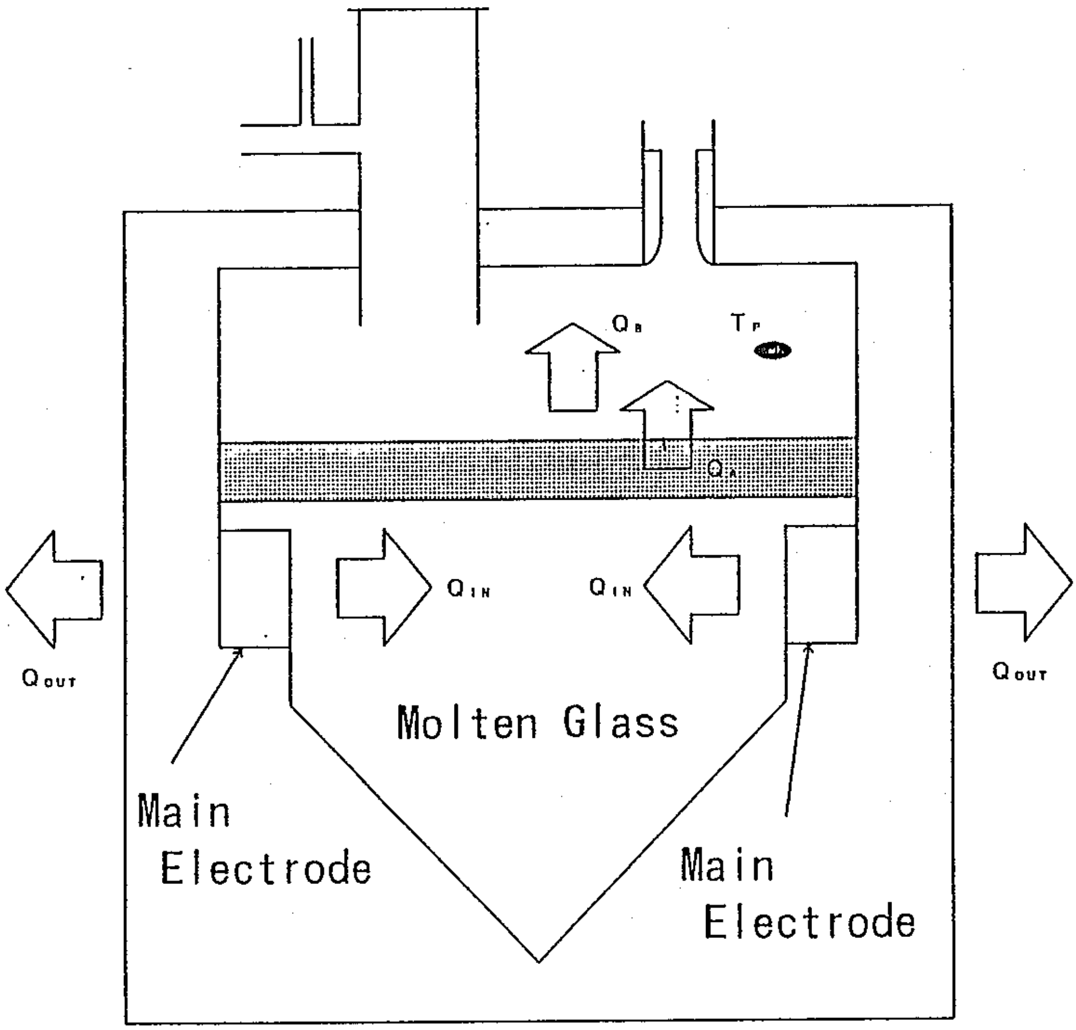


Flowchart for Electric Potential Program



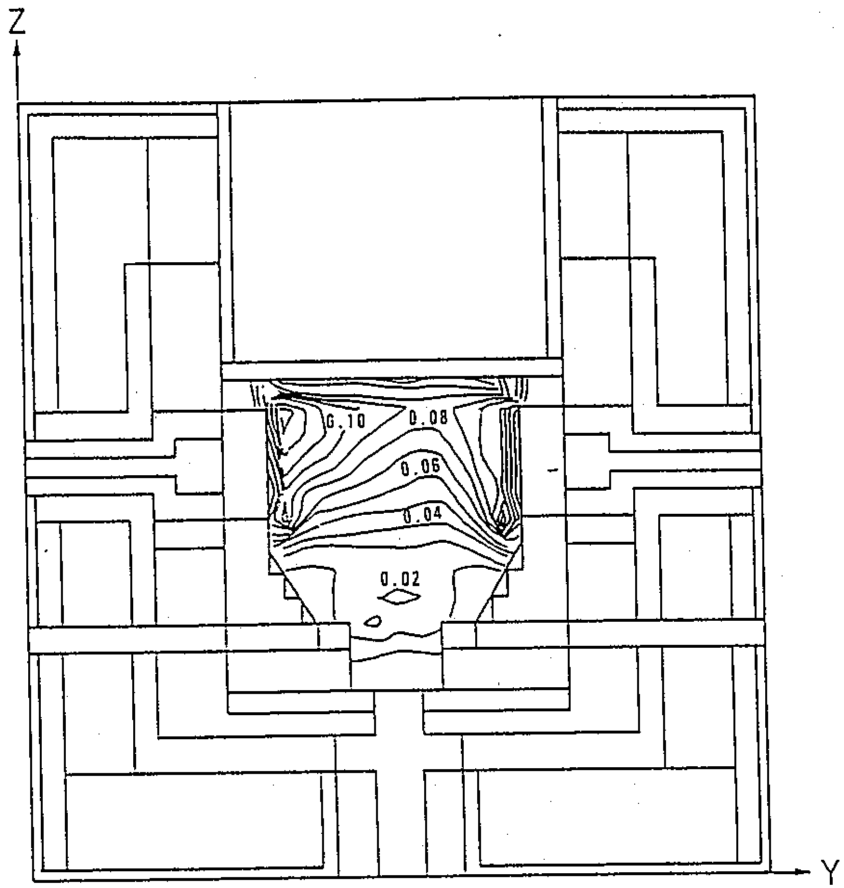
①	Heat-Transfer Rate	⑥	Specific heat
②	Heat loss	⑦	Density of Glass
③	Constant Valtage	⑧	Electrical Resistivity
④	Constant Current	⑨	Coefficient of Thermal Expansion
⑤	Themal Conductivity	⑩	Viscosity

Modelling of Boundery Condition for Energy Equation

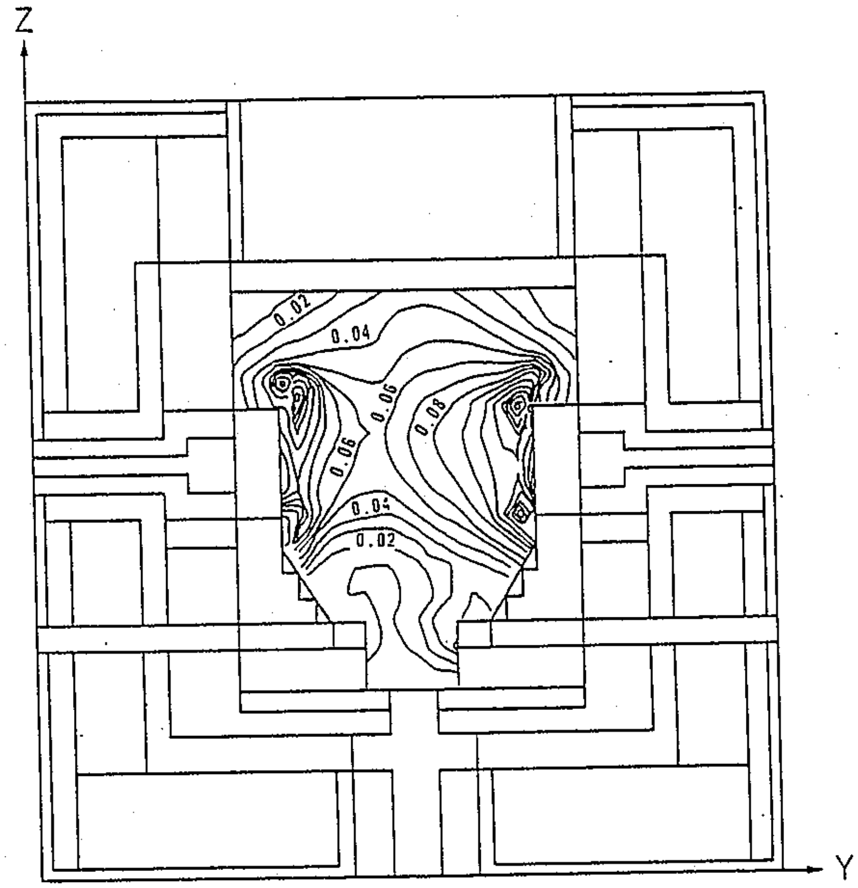


Q_A : Heat loss for Melting
Q_B : Heat loss for Molten Sufase
Q_{IN} : Power Input
Q_{OUT} : Heat loss from The Wall
T_P : Plenum Temperature

Heat Transfer Phenamina in Melter



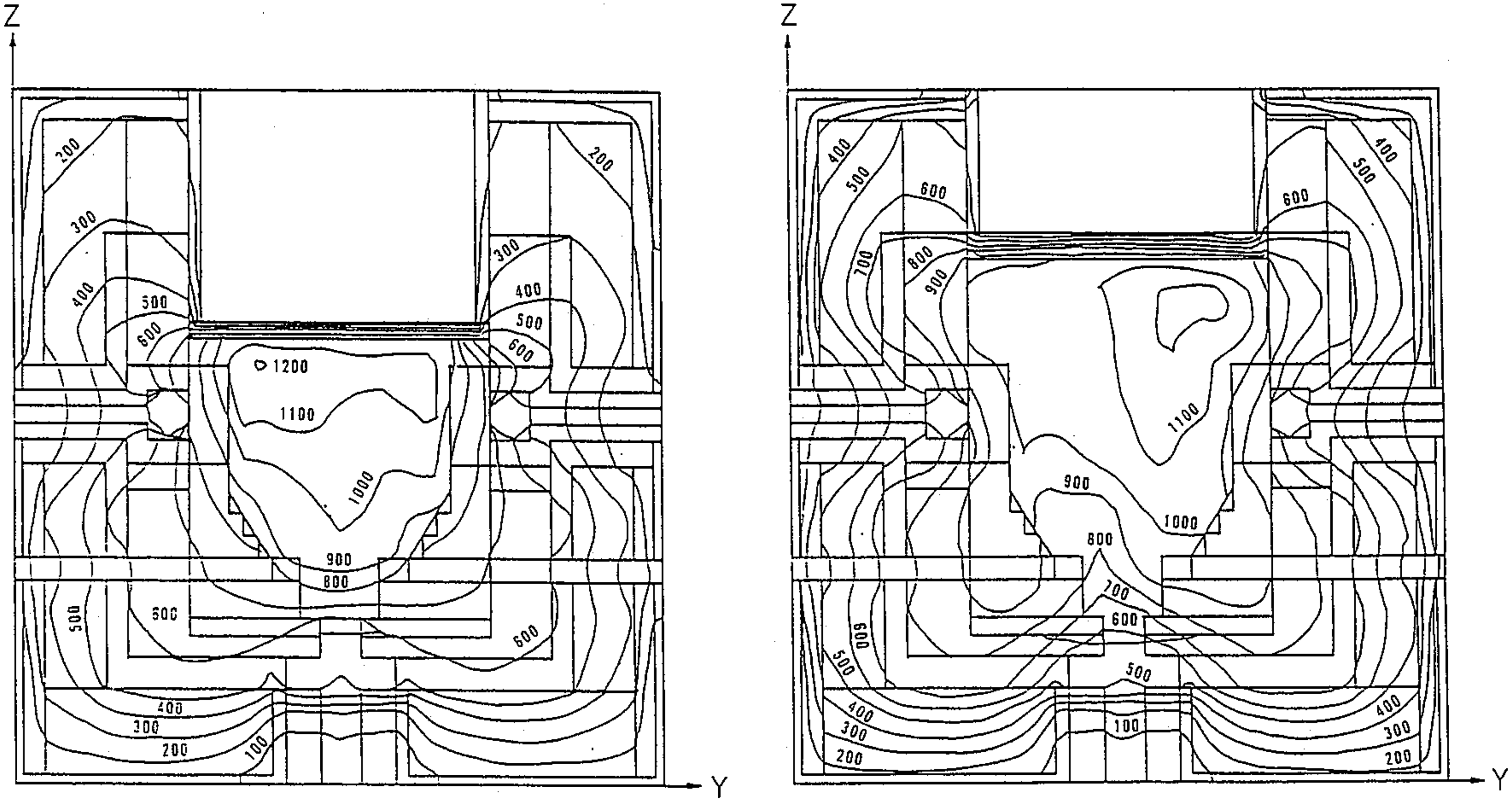
Feed Operation



Idling Operation

Power intensity

(Advanced B Melter, Unit$\times 10^6$ w/m^3)

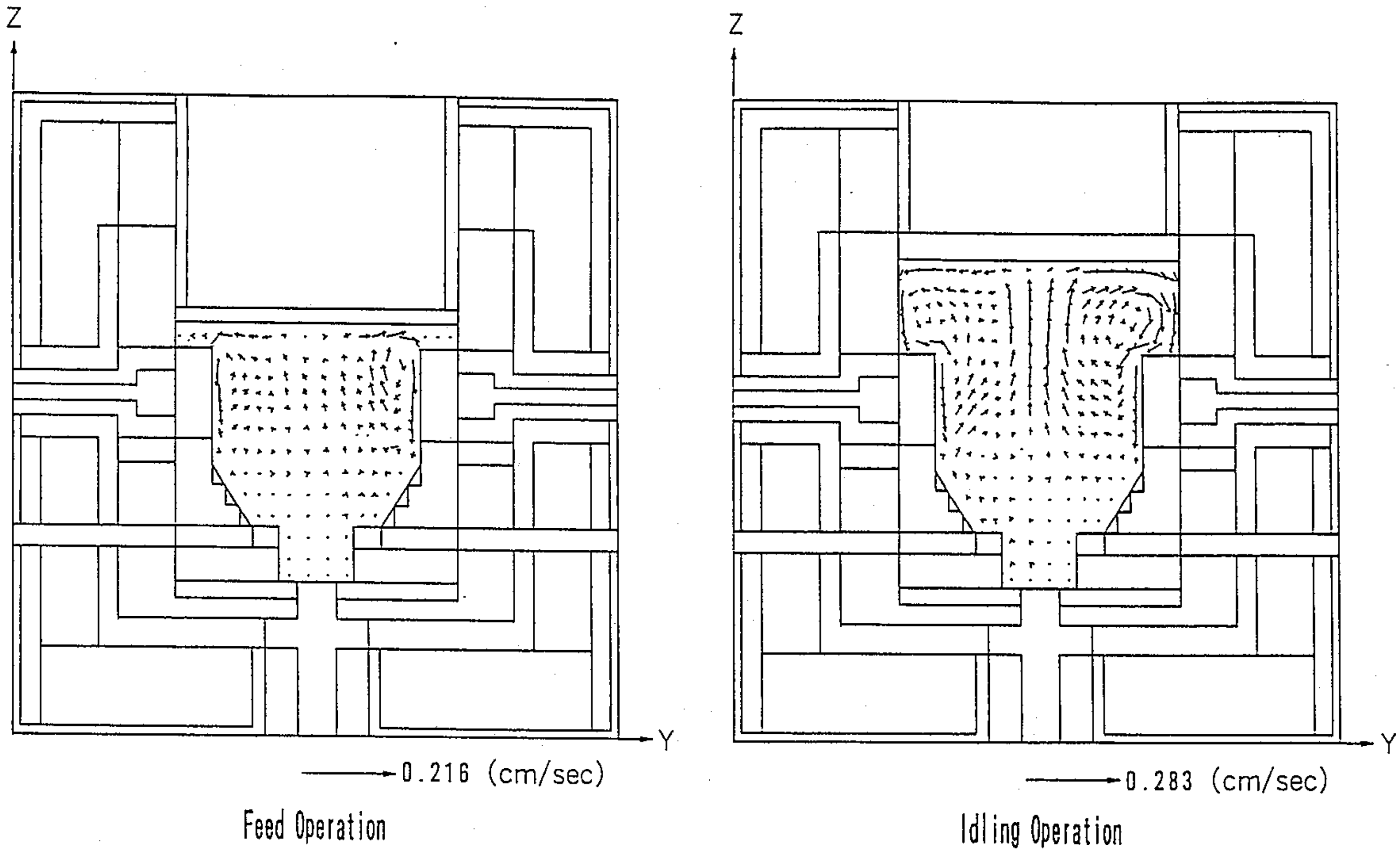


Feed Operation

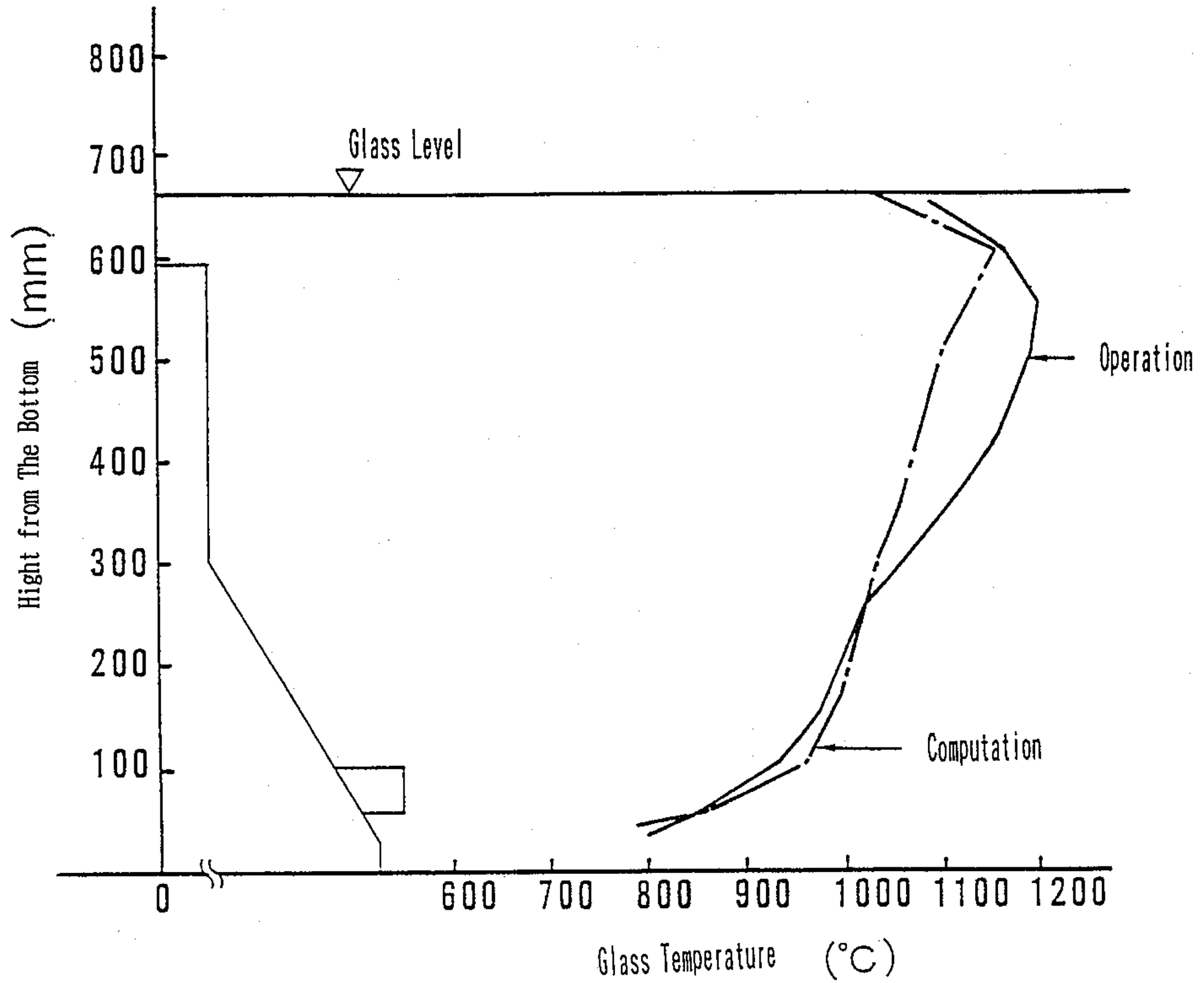
Idling Operation

Temperature Distribution in Melter

(Advanced B Melter, Unit ℃)

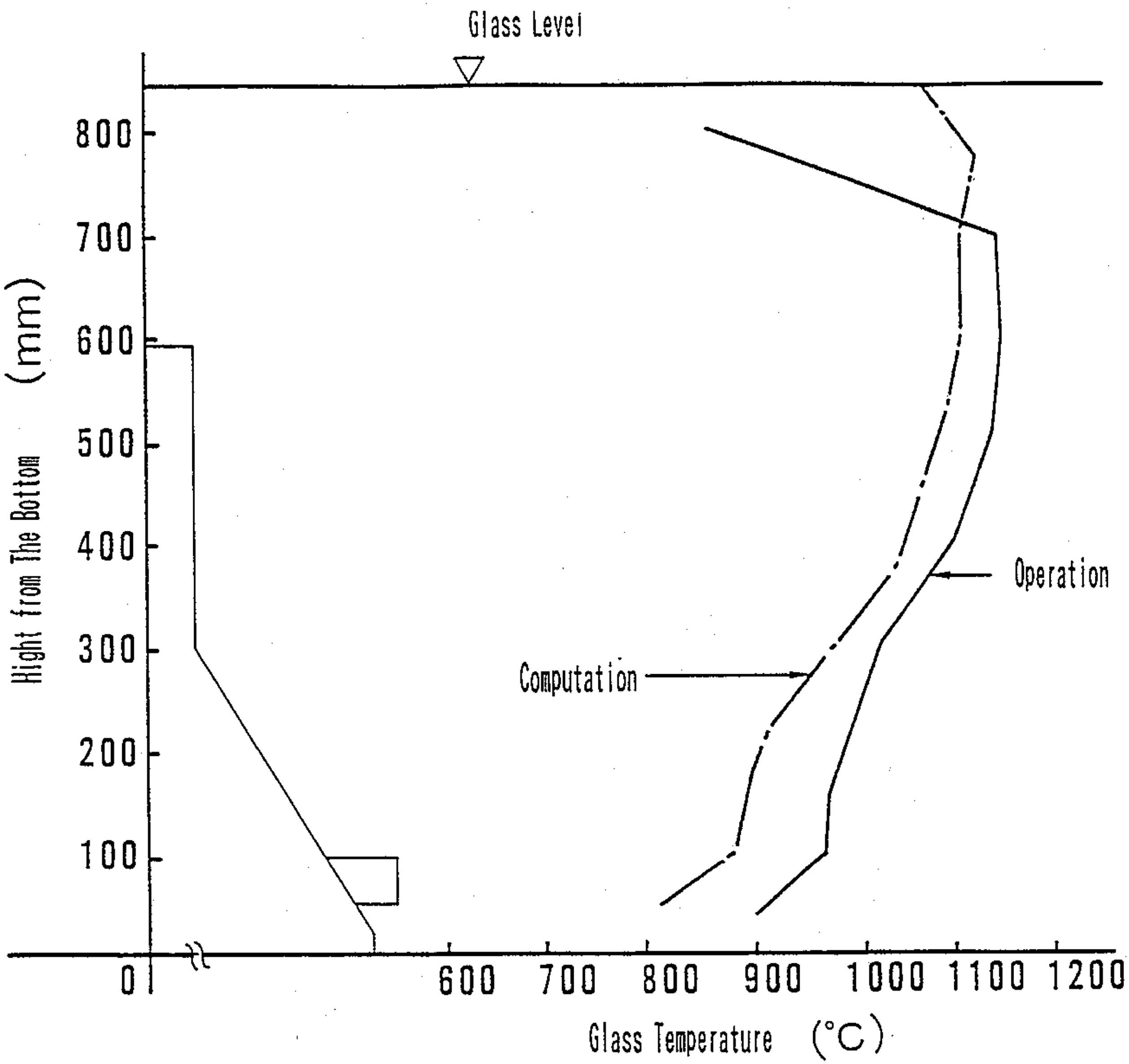


Velocity Vector Profile of Molten Glass
(Advanced B Melter)



Conparison of Glass Temperature Profile

between Calculation and Operation (Feed Operation)



Conparison of Glass Temperature Profile

between Calculation and Operation (Idling Operation)

Modification of analysis code

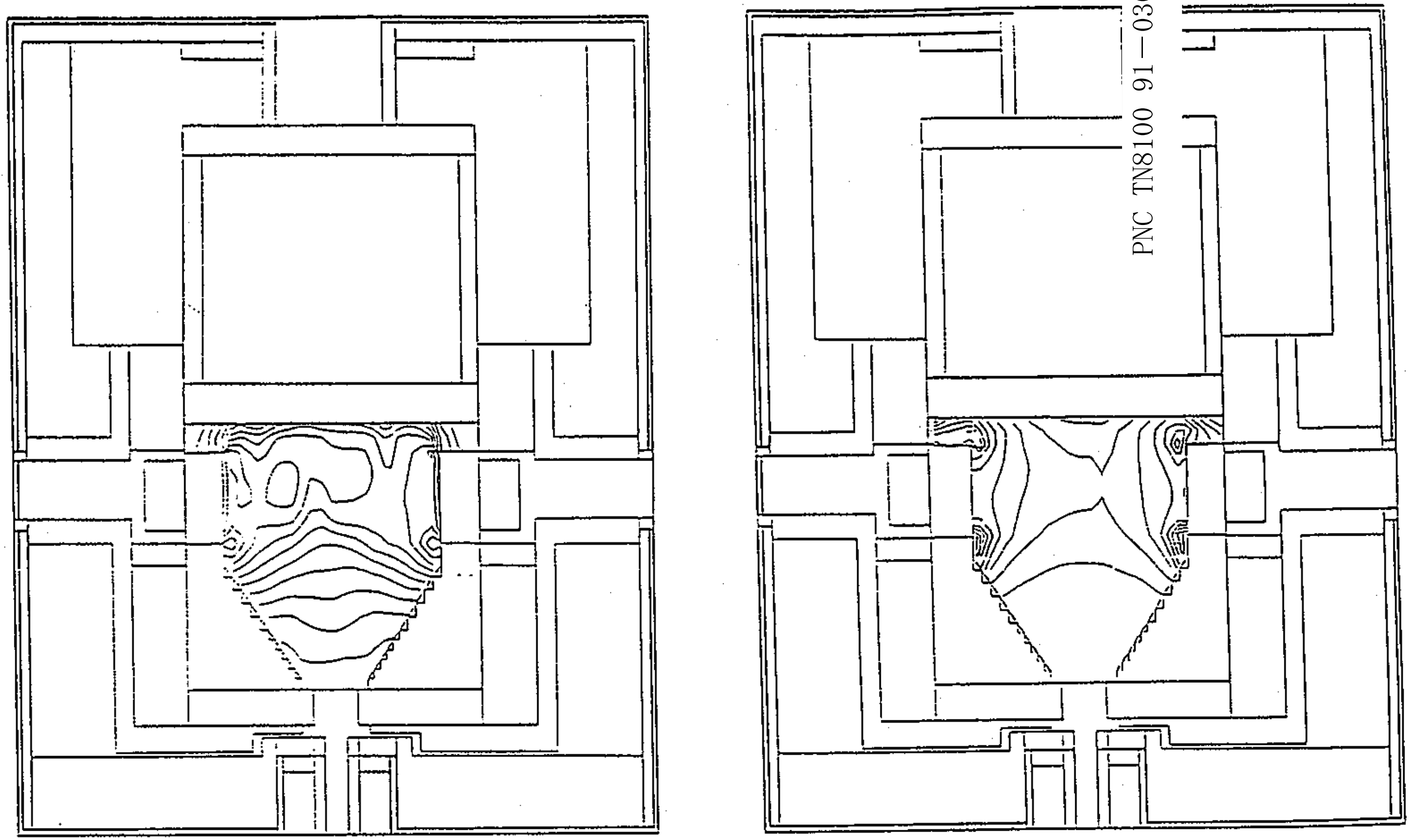
Electric resistivity was assummed to independent of temperature in electric potential equation as expessed below,

$$\nabla^2 \phi = 0$$

In modification, the temperature dependence of electric resistivity was considered. Therefore electric potential equation is

$$\nabla (R^{-1} \nabla \phi) = 0 \quad , \qquad R = \exp\left(\frac{A}{T-C} + B \right)$$

where A, B, C is constant number, T is temperature of molten glss and R is electric resistivity.

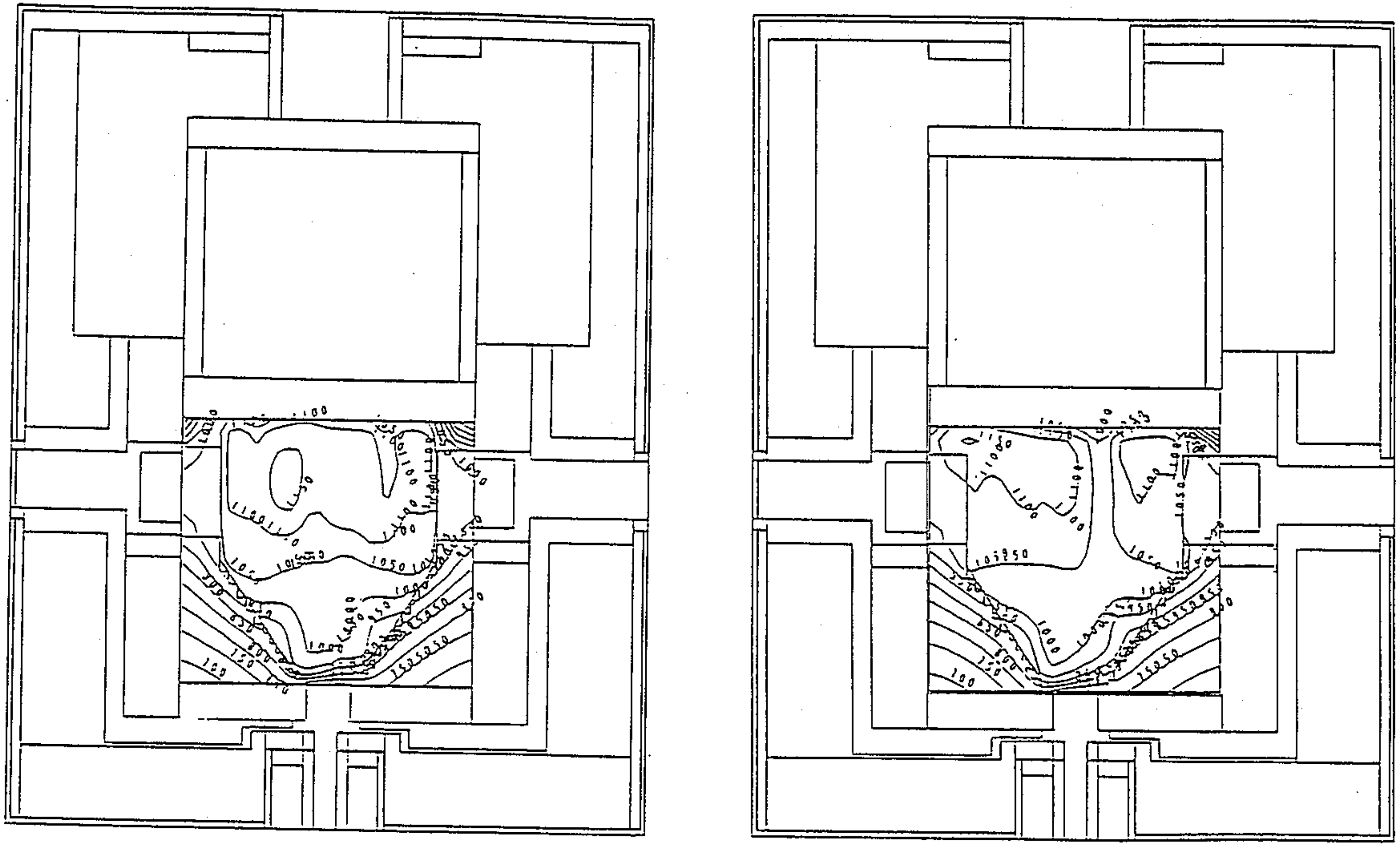


Before Modified MAX. 2590A/m² MIN. 266A/m² Increment 258A/m²

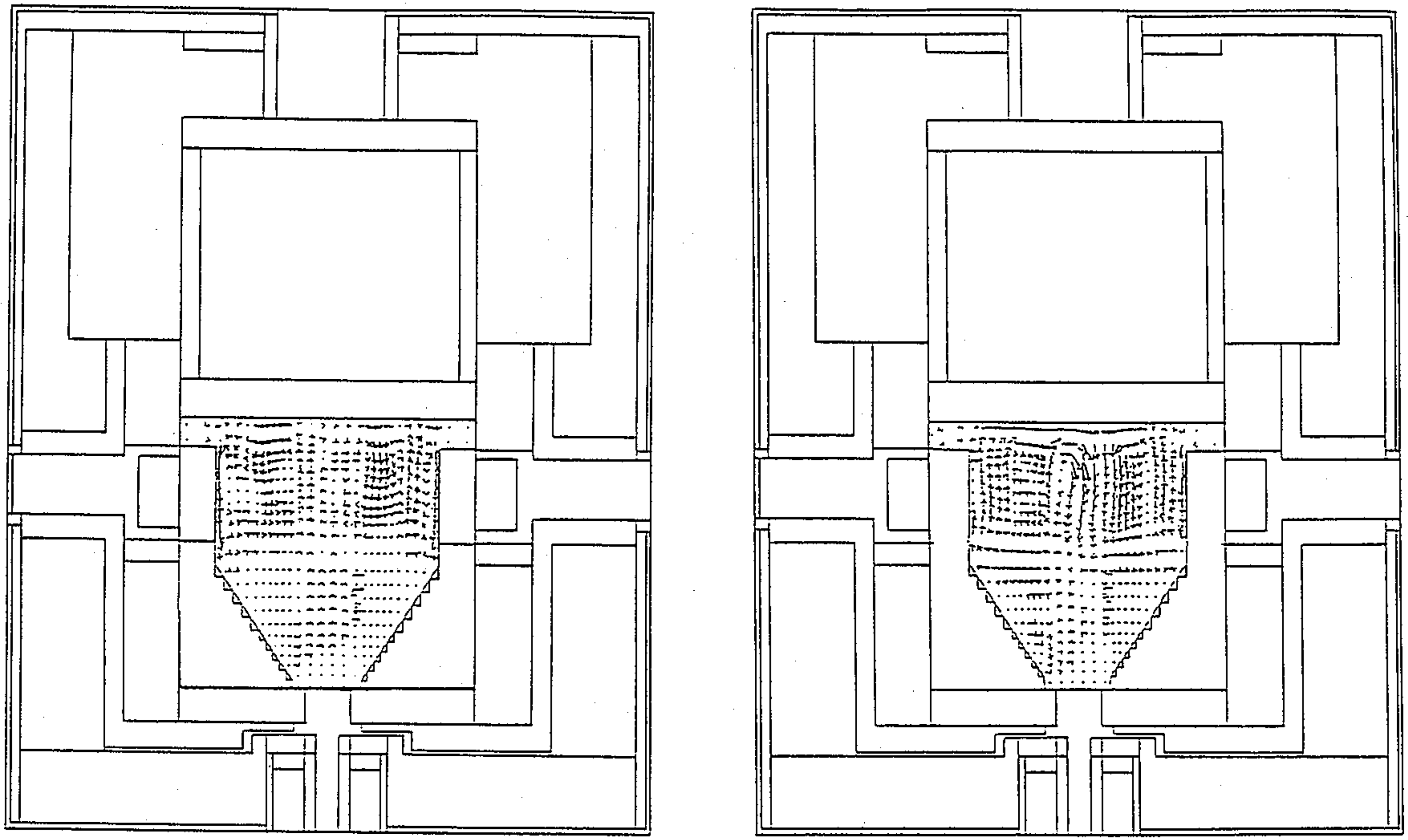
Modified MAX. 1601A/m² MIN. 273A/m² Increment 258A/m²

Effect of Modification(Mock-up 3 Melter)

(Current Density Y-Z section)



Effect of Modification(Mock-up 3 Melter)
(Temperature Distribution Y-Z section)



Effect of Modification(Mock-up 3 Melter)

(Velocity Vector Profile Y-Z section)

Conclusion

① The agreement on the temperature distribution in molten glass obtained between calculation and operation.

② Flow profile of molten glass was satisfartiry from the physical point of view.

③ The effect of temperature dependence of electric resistivity in electric potential equation was cleary observed in electric, themal and flow profile in the melter.

Plan of future

Simulate :

the influence of noble metal on operation.

the influence bubbling in the melting cavity.

Modify the boundary condition to make the compertational results agree with the operational.

2-6 Advanced Electrode and Refractory Material

PNC TN8100 91-030

ADVANCED ELECTRODE AND REFRACTORY MATERIALS

The Tenth Annual KfK-PNC Meeting on cooperation in High-Level Waste Management

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Glass contact materials of melers to vitrify HLLW

Country	Organization	Melter	Refractory material	Electrode (Vessel) material
Germany	KfK	K-6′	ER-2161	Inconel 690
	PAMELA	K-5		
USA	PNL	RLFCM	Monofrax K3	
	SRP	SGM		
	WVDP	SFCM		
Japan	PNC	TVF Melter		
	JNFS	—		
France	AVM	—		Inconel 601
	AVH	—		
UK	WVP	—		Inconel 690

Objectives of development of advanced electrode and refractory materials

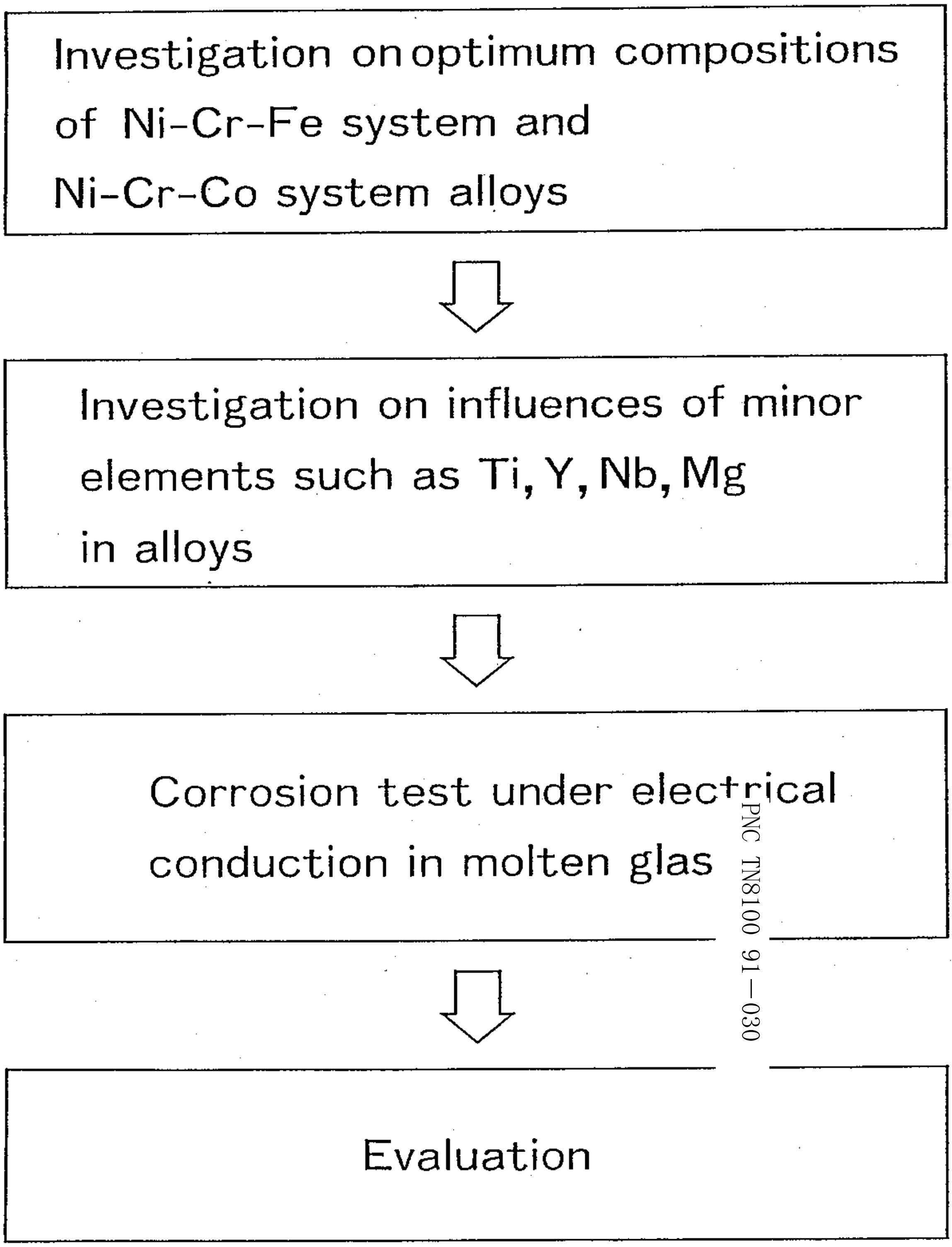
1. Extension of the service life of melter

2. Rise of margins for melter operation

Target performance of advanced electrode material

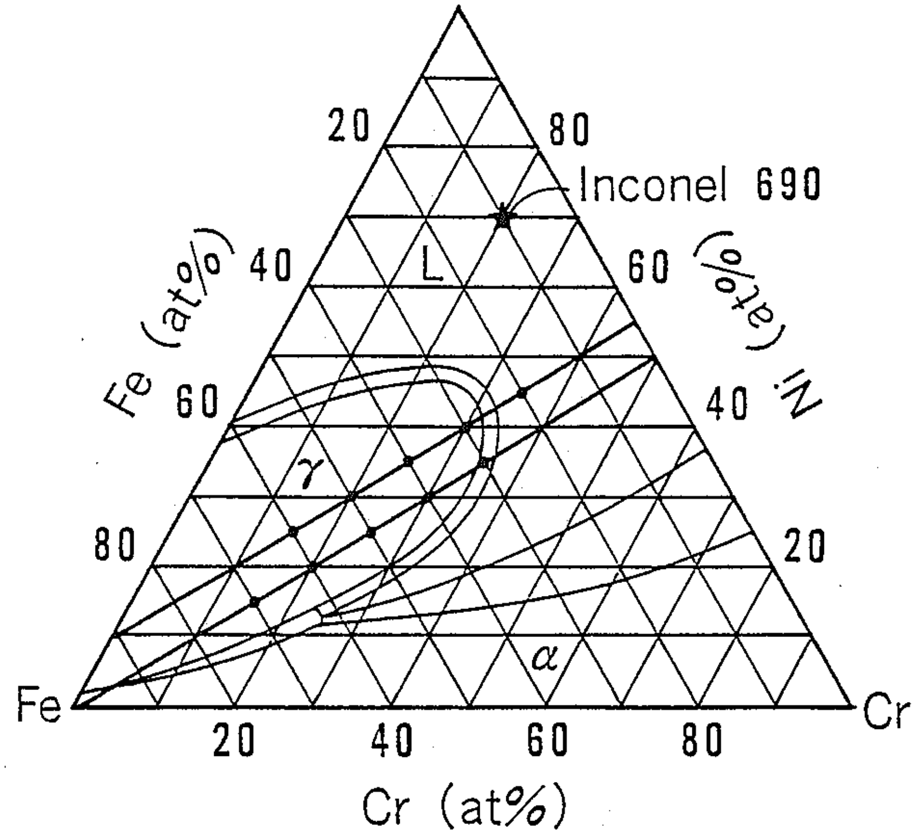
1. Higher melting point

2. Higher corrosion resistance in molten glass

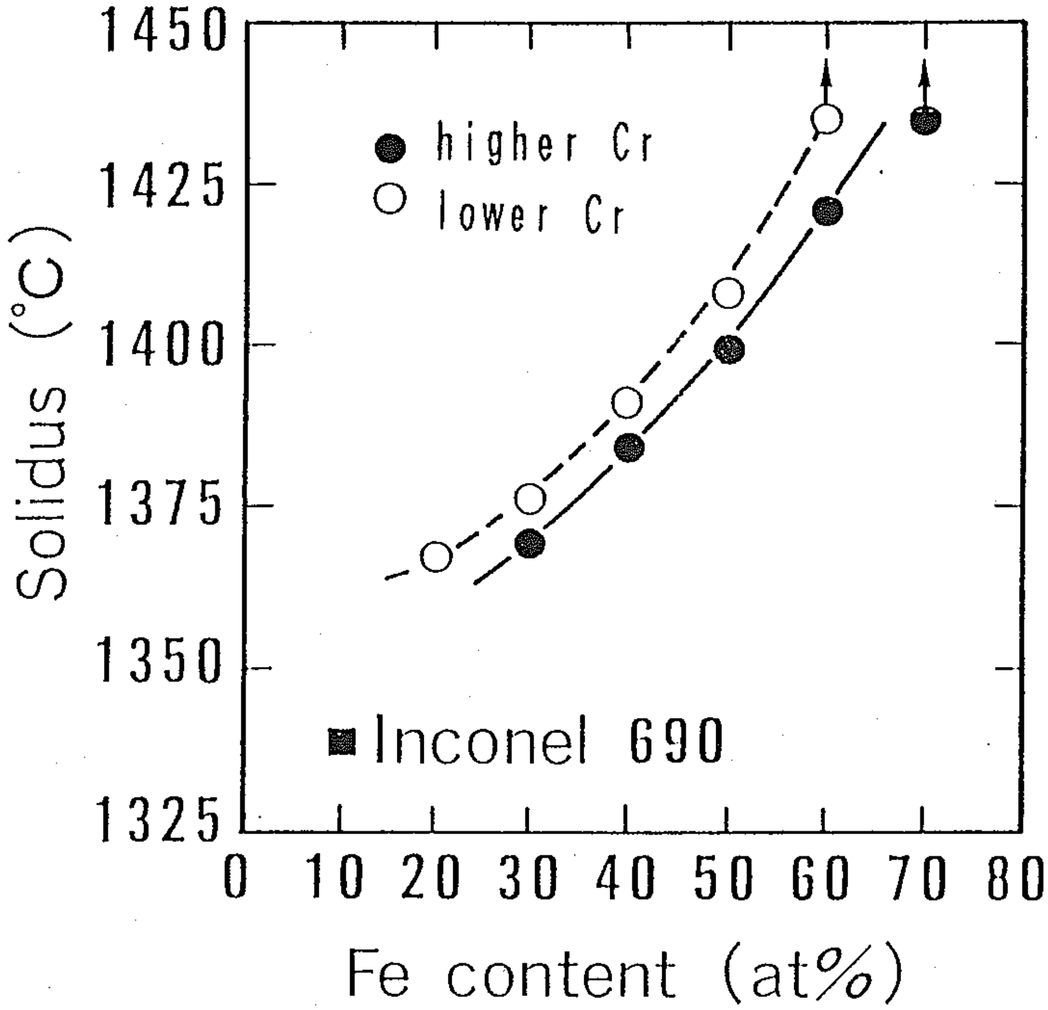


Development steps of advanced electrode material

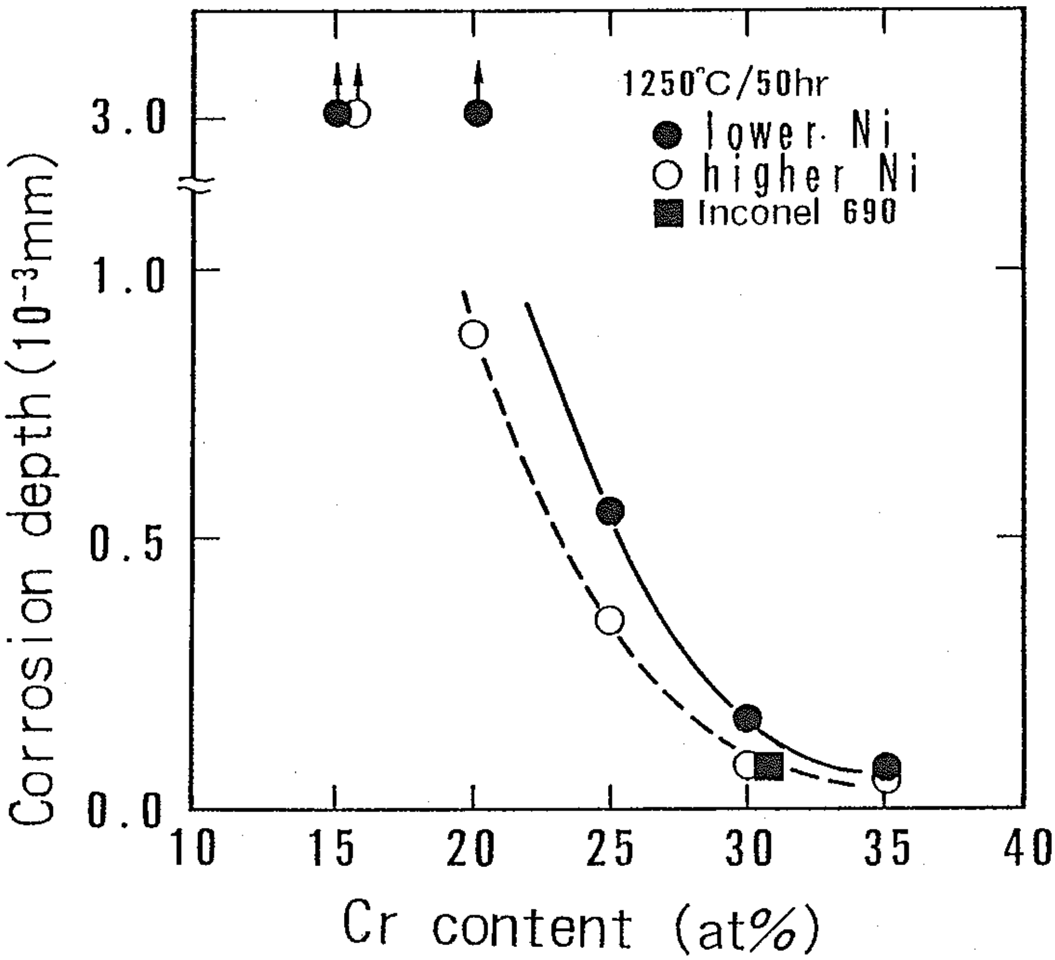
alloy	Chemical Composition (at%)		
	Ni	Cr	Fe
F 1	Bal.	15	70
F 2		15	60
F 3		20	60
F 4		20	50
F 5		25	50
F 6		25	40
F 7		30	40
F 8		30	30
F 9		35	30
F 10		35	20



Experimental Ni-Cr-Fe alloys



Effect of Fe content on solidus

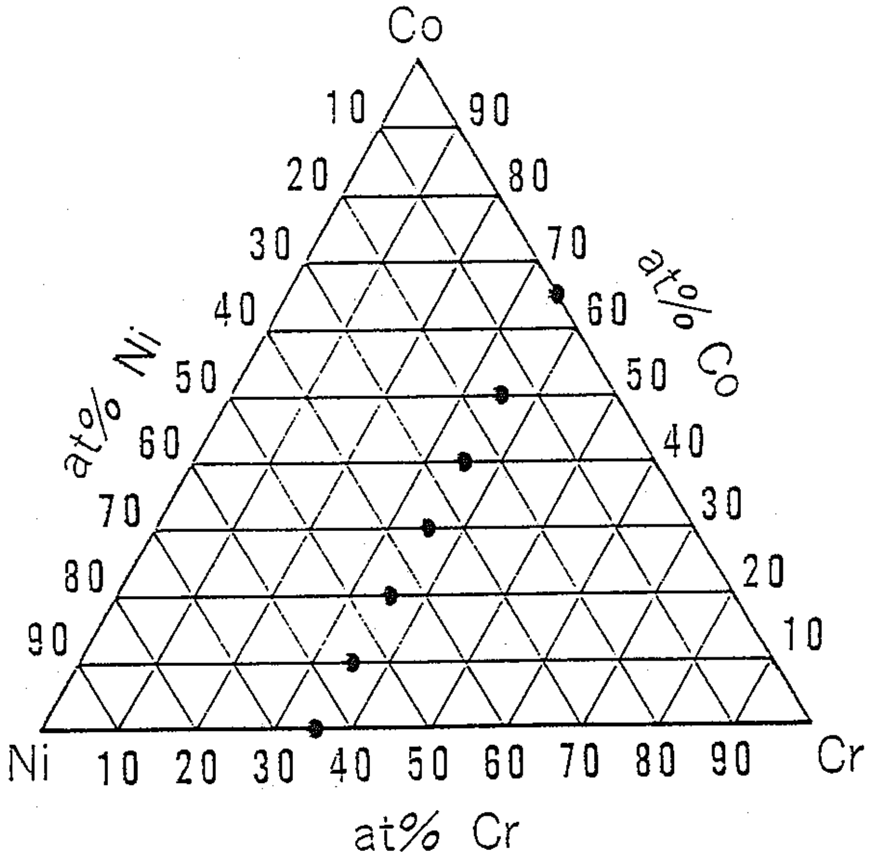


Effect of Cr content on corrosion depth

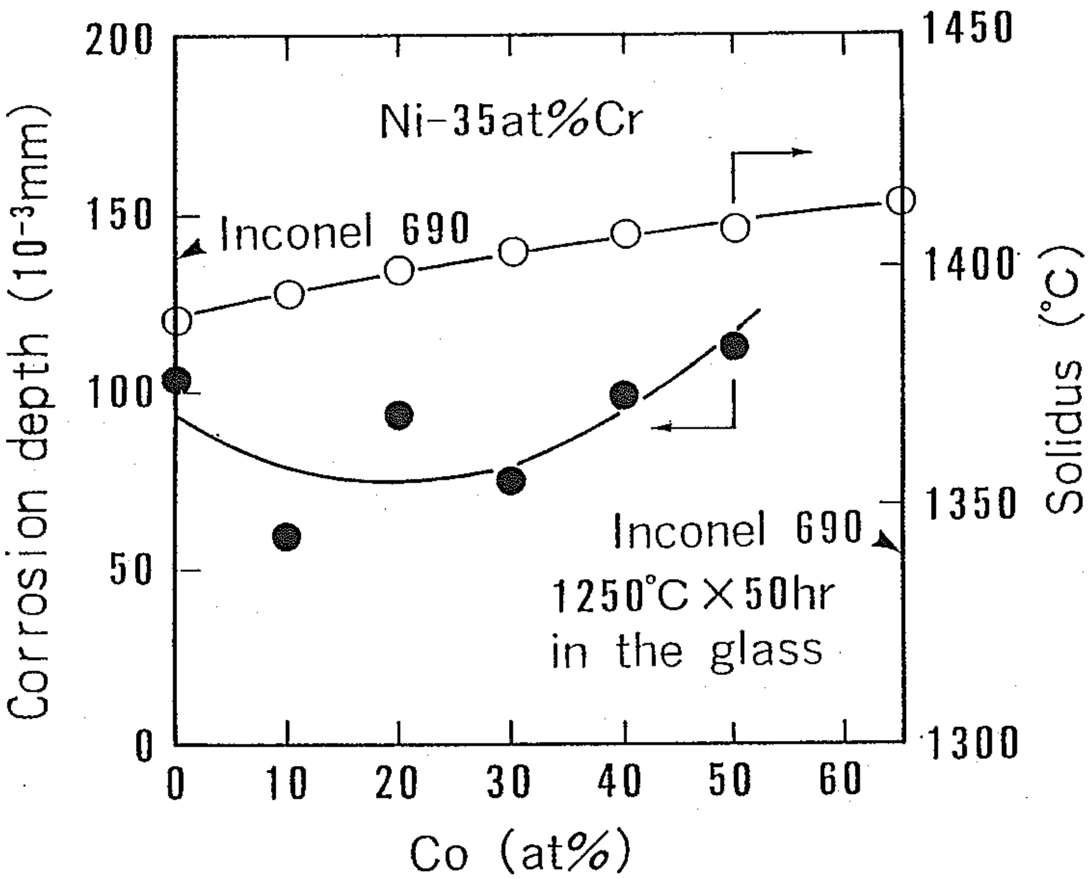
No.	Alloy	addition	Attack Depth (μm)	Solidus (°C)
1	F 8 Fe-40Ni-30Cr	no add.	219	1384
2		+0.3Ti	187	1377
3		+0.3Nb	152	1372
4		+0.3Mg	245	1383
5		+0.1Y	91	1385
6		+0.6Ti	140	1366
7		+1.0Ti	84	1360
8		+1.5Ti	120	1338
9		+1.0Nb	307	1357
10		+1.5Nb	282	1345
11	F10 Fe-45Ni-35Cr	no add.	108	1380

Corrosion depth in molten glass and soidus for experimental alloys

Alloy	Chemical composition (at%)		
	Ni	Cr	Co
C 1	Bal.	35	—
C 2			10
C 3			20
C 4			30
C 5			40
C 6			50
C 7	—		Bal.



Experimental Ni-Cr-Co alloys



Effect of cobalt content on corrosion depth and solidus

No.	Alloy	addition Ti	addition Y	Attack Depth (μm)	Solidus (°C)
1	C 4 Ni-35Cr-30Co	no add.		74	1408
2		+0.3	−.	54	1400
3		+1.0	−.	32	1377
4		.−	+0.1	45	1376
5		+0.3	+0.1	53	1384
6		+1.0	+0.1	100	1359

Corrosion depth in molten glass and solidus for experimental alloys

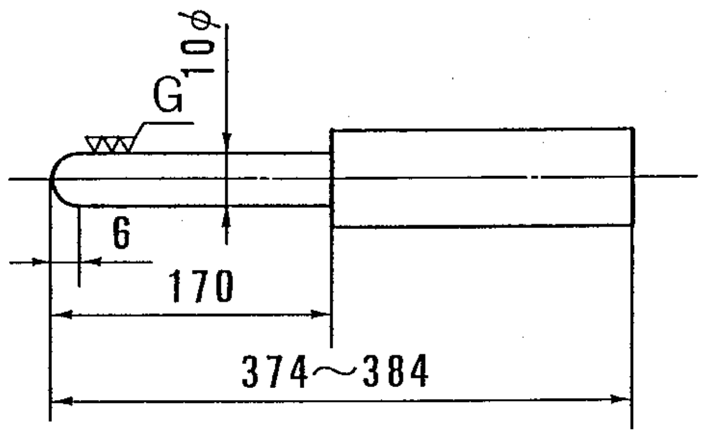
Experimental alloys in corrosion test under electrical conduction in molten glass

- Inconel 690

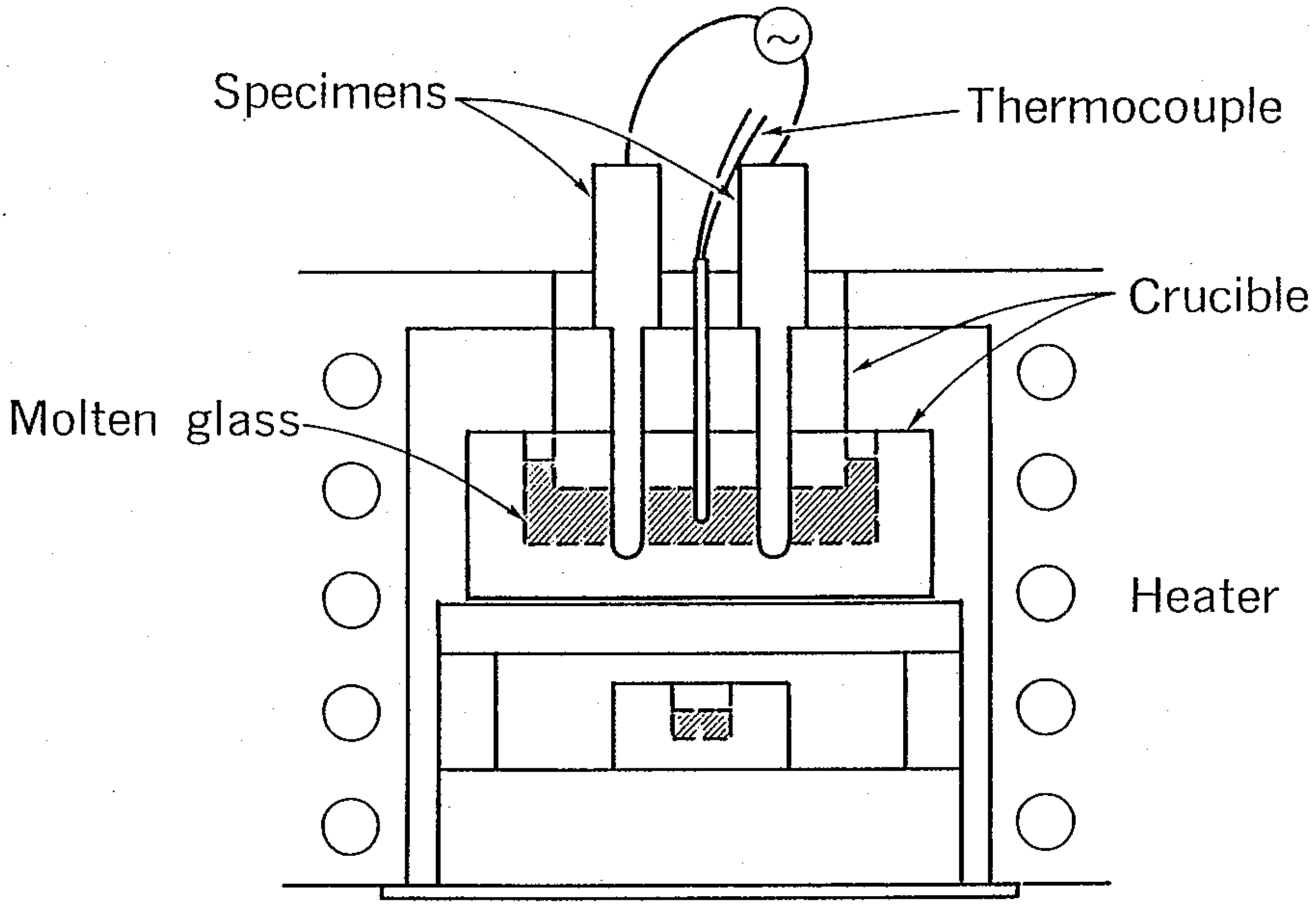
- Ni-Cr-Fe system
  (1) Fe-45%Ni-35%Cr-1.0%Ti
  (2) Fe-45%Ni-35%Cr-0.1Y

- Ni-Cr-Co system
  (1) Ni-35%Cr-30%Co
  (2) Ni-35%Cr-30%Co-0.3%Ti
  (3) Ni-35%Cr-30%Co-1.0%Ti
  (4) Ni-35%Cr-30%Co-0.1%Y

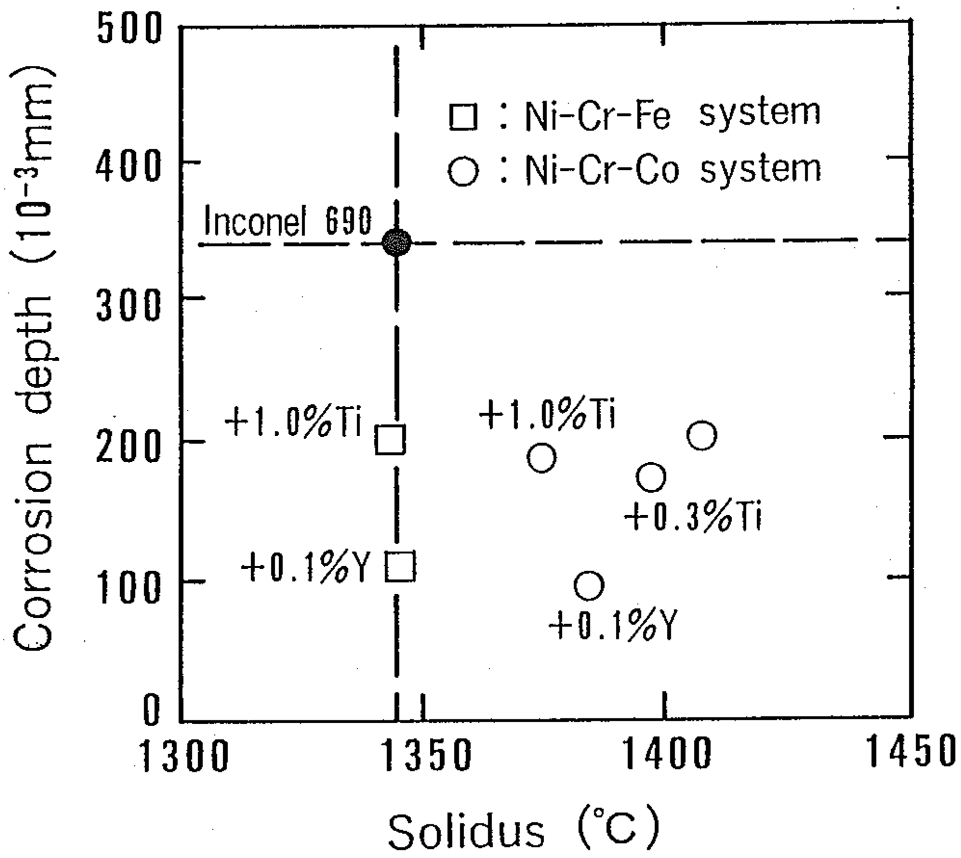
(at %)



Configuration of test specimen



Corrosion test appparatus



Corrosion resistance and solidus of experimental alloys

Corrosion test : in molten glass at 1250℃, 0.8A/cm² for 20 days

Advanced refractory material

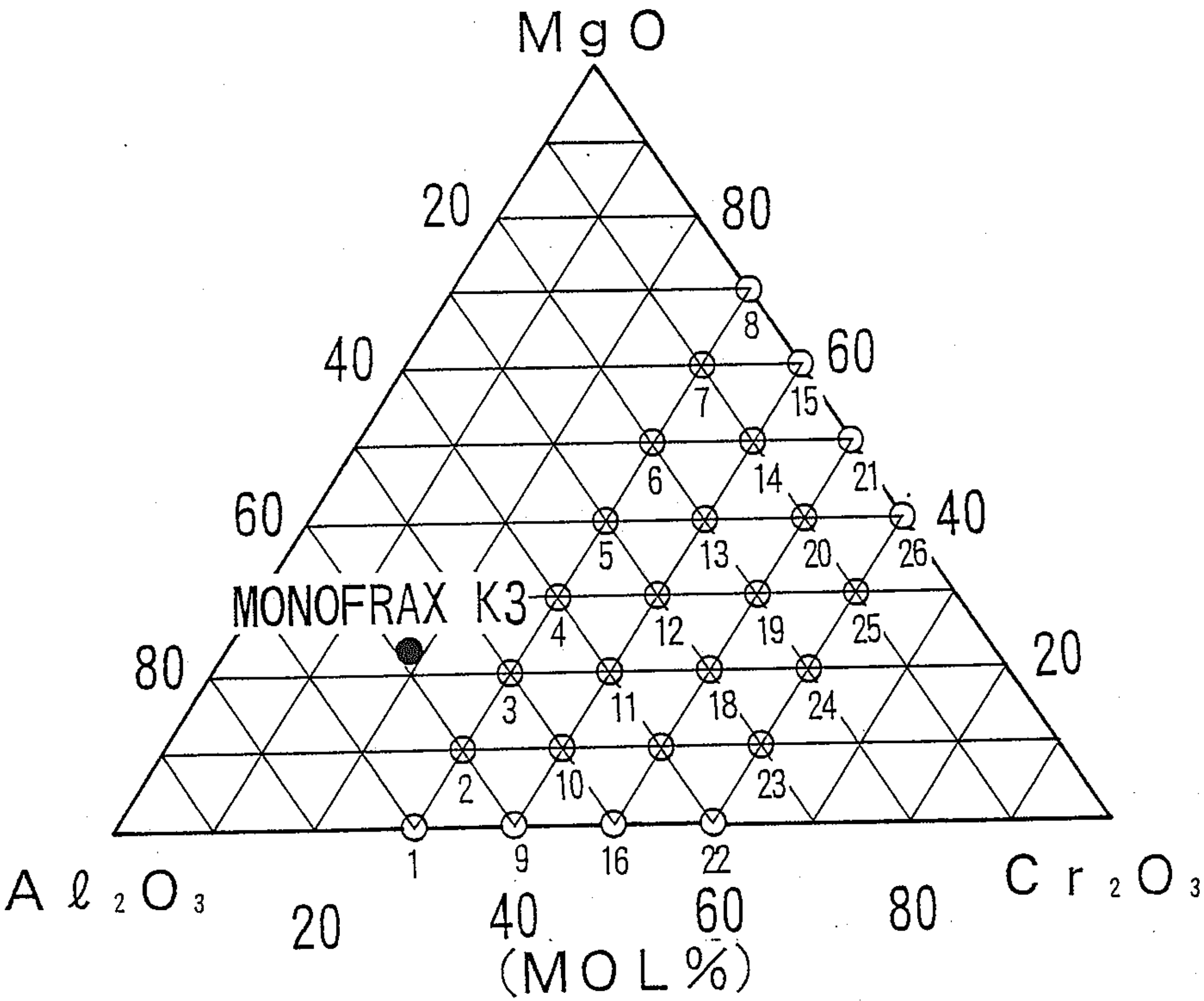
Target performance of advanced refractory material

1. Higher thermal shock resistance

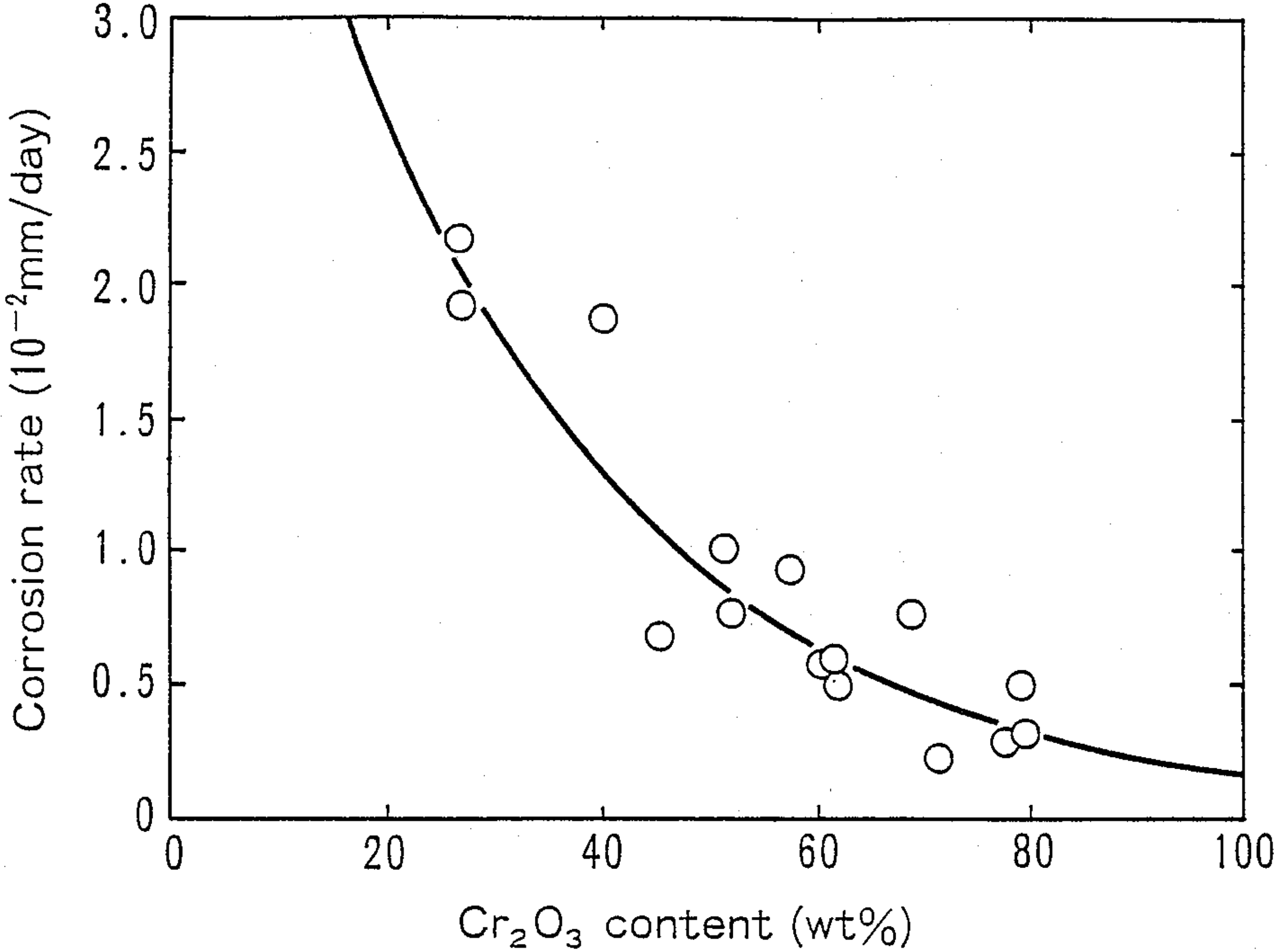
2. Higher corrosion resistance in molten glass

Corrosion test result for commercial fused cast refractories

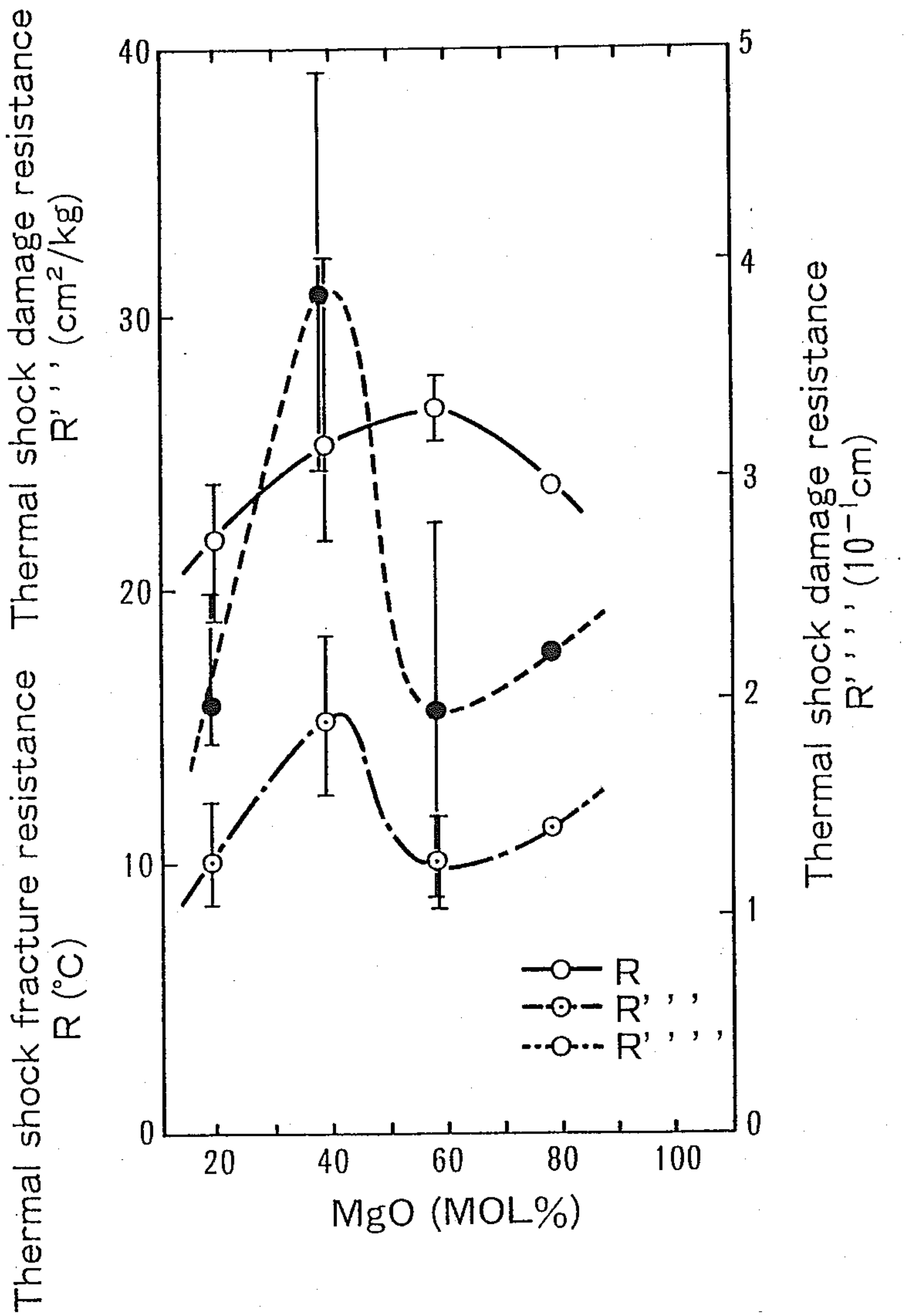
Name of refractory	Major constituents	Main phases	Corrosion rate (10^{-2} mm/d)	
			Static condition 1250°C, 720hrs	Dynamic condition 1250°C, 336hrs 50rpm rotation
MONOFRAX E	Cr_2O_3, Al_2O_3 MgO, Fe_2O_3	Spinel R_2O_3 Solid solution	0.33	N.D.
MONOFRAX K3	Al_2O_3, Cr_2O_3 MgO, Fe_2O_3	Spinel R_2O_3 Solid solution	1.92	2.14
MONOFRAX Z	ZrO_2	Zirconia	5.67	8.38
MONOFRAX CS5	Al_2O_3, ZrO_2 SiO	Corundam Zirconia, Glass	9.84	20.36
ER-2161	Al_2O_3, Cr_2O_3 ZrO_2, SiO_2	R_2O_3 Solid solution Zirconia, Glass	2.33	7.14



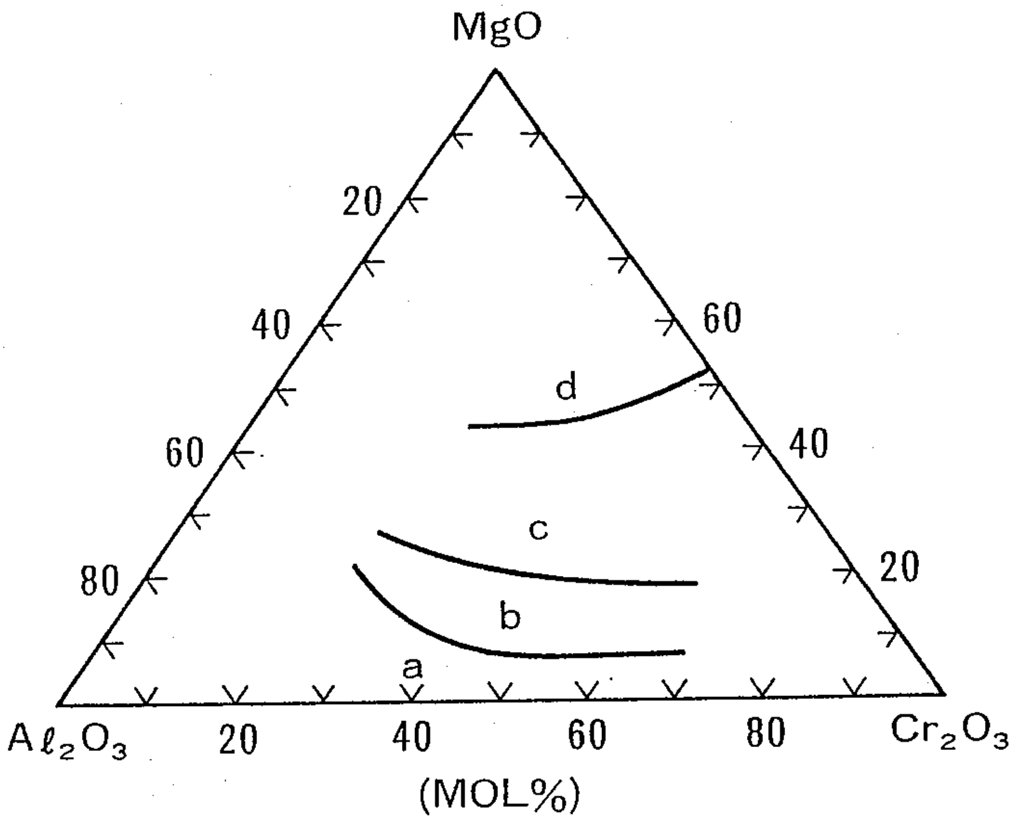
Composition of refractories tested



Relation between Cr_2O_3 content and corrosion rate



Relations between thermal shock resistance and MgO content



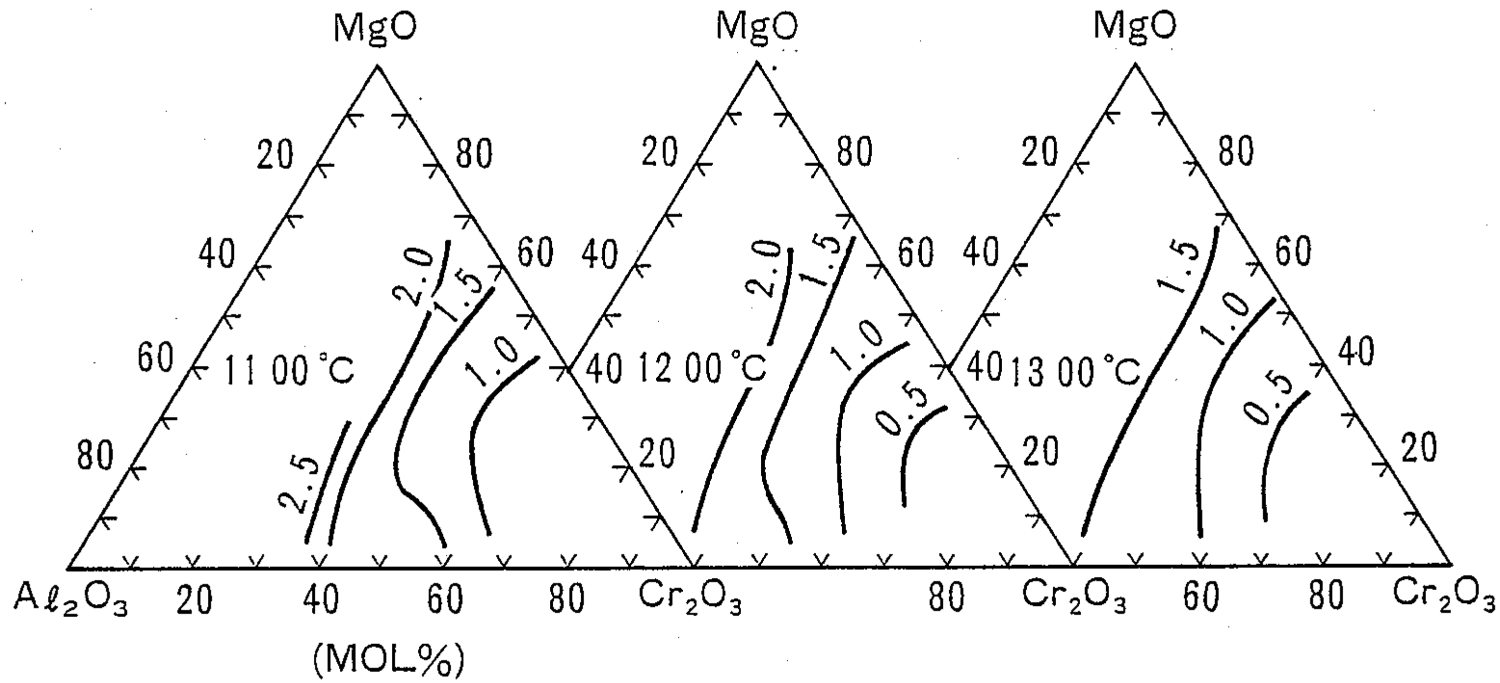
a :Penetration crack

b :No crack

c :Surface crack

d :Penetration and surface crack

Product quality of experimental refractories



Electrical resistance of experimental refractories (1.0² log Ω m)

Chemical composition of advanced chromia fused cast refractory and MONOFRAX K3

	New Composition	MONOFRAX K3®
Cr_2O_3	60.4	27.3
Al_2O_3	31.9	58.3
Fe_2O_3	------	6.0
MgO	3.8	6.0
MnO	2.3	------
CaO	0.3	0.5
SiO_2	1.3	1.5
TiO_2	------	0.1
(wt.%)		
Al_2-Cr_2O_3 s.s.*1	60	65
Spinel*2	39	34
Glass phase, etc.	1	1
(vol.%)		

*1 solid solution　　*2 $(Mg,Fe,Mn)(Al,Cr)_2O_4$

Properties of advanced refractory and MONOFRAX K3

	New Composition	MONOFRAX K3®
Bulk density ($10^3 kg/m^3$)	4.15	3.80
Apparent density ($10^3 kg/m^3$)	4.54	3.97
Apparent porisity (%)	8.5	4.3
Modulus of rupture S ($10^4 Pa$)	48	77
Young's modulus E ($10^6 Pa$)	1940	2650
Poisson's ratio ν	0.21	0.21
Fracture energy γ (J/m^2)	24.8	13.2
Thermal expansion coefficient α (10^{-6} ℃$^{-1}$)	7.73	8.04
Thermal shock fracture resistance parameter*1 R (℃)	24.8	28.0
Thermal shock damage resistance parameter*2 R'''' (m)	27.5	7.9
Corrosion factor		
static condition*3	0.301	1.000
dynamic condition*4	0.231	1.000
Electrical resistivity*5		
at 1000℃ ($10^2 \Omega m$)	136	478
1200℃	55	180

*1 $R=S(1-\nu)/E\alpha$ *2 $R''''=E\gamma/S^2(1-\gamma)$ *3 1250℃, 720hrs.
*4 1250℃, 336hrs., 50r.p.m.
*5 Electrical resistivity of PNC glass is $10\times10^2 \Omega m$ at 1000℃ and $4\times10^2 \Omega m$ at 1200℃

Conclusions

Advanced Ni-Cr-Fe system alloy (compared with Inconel 690)

1. Melting point is the same
2. Corrosion resistance is three times higher

Advanced Ni-Cr-Co system alloy (compared with Inconel 690)

1. Melting point is higher by 30°C～60°C
2. Corrosion resistance is more than three times higher

Advanced refractory (compared with MONOFRAX K3)

1. Corrosion resistance is three to four times higher
2. Thermal shock damage resistance is two to three times higher

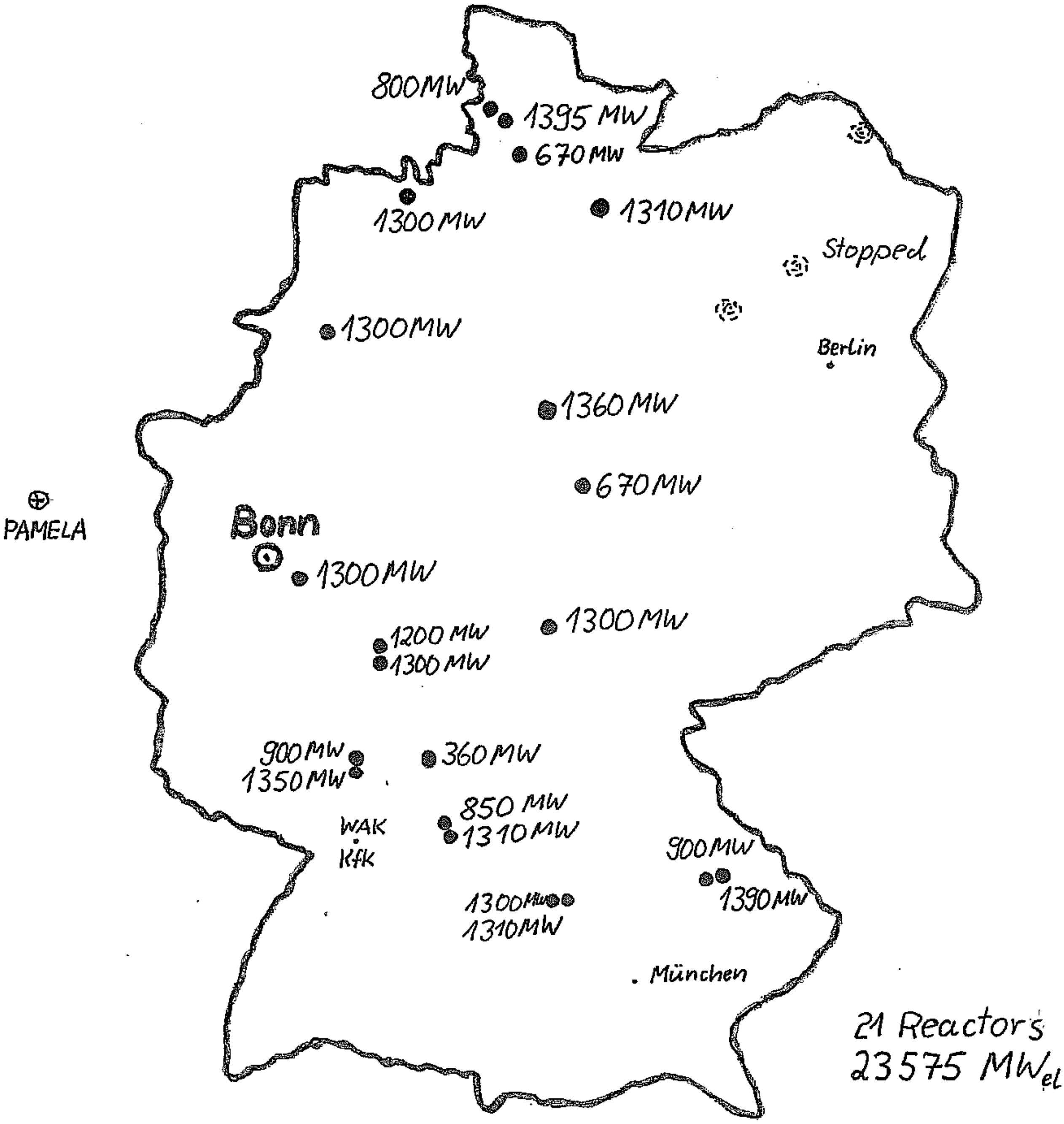
3. K f K 側の発表資料

3-1 State of The Art of HLLW Conditioning Program in Germany

STATE OF THE ART OF THE HLLW CONDITIONING PROGRAM IN GERMANY

Prepared for the tenth annual PNC-KfK meeting on HLLW management

Nov.19, 1990



Nuclear Power Station in Operation in GERMANY

Developments, Studies, and Activities

1. KfK - INE

- Development of noble metals compatible vitrification technique for the very high active WAK-HAWC (500-1000 Ci/l)
- Supplement of technical-scale work by lab-scale melter tests
- Optimization of remote melter components and of the off-gas line
- Glass characterization for final disposal in Germany
  - for WAK-HAWC glass
  - for glasses coming from abroad (France, Great Britain)
- Canister Corrosion experiments
- Near field investigations of glass products in the final disposal repository

2. WAK

- Efforts are made to get the licence for transportation of WAK-HLLW from Karlsruhe to Mol/Belgium
- As an option studies are made for vitrification of the WAK-HLLW at the Karlsruhe reprocessing site

3. PAMELA

- Vitrification status of the HLLW stored at the former Eurochemic site
- Plans for melter decomissioning (preliminary)

KfK-INE

■ Noble metals compatible melter development

- Development is for WAK-HLLW, 100m³ expected, high specific activity 500-1000 Ci/l; in total approximately 0.5 to of the noble metals Ru, Rh and Pd are present in the WAK-waste

- Main condition for present melter development: melter must be suitable in respect to both outside dimension and remote installation in the PAMELA melter cell. Also melter weight is limited to 18.5 to.

- K-6' melter serves as the prototype; 1st test carried out in May/June 1990. 10m³ of fully simulated HLLW were vitrified (but without Rh). Feed throughput 25 l/h, higher feed rates up to 30 l/h seem to be possible.

- For active use in PAMELA in case the WAK-HAWC would be vitrified by this plant: A new melter designated K-6 will be designed by INE in 1991/92 (basis is INE's operation experience with the K-6'-melter)

- Further K-6' melter tests are scheduled for 1991 and 1992

- Primary principles of the K-6' design:
  - collect settling noble metal particles in a collecting chamber (60 to 75° wall inclination)
  - remove the settled particle as completely as possible by routine glass pouring via bottom drain

- First test results look promising. They will be presented in the technical sessions during this meeting

- Mathematical melter modelling work are continued (focus: noble metal effects, worst case modelling)

KfK-INE

▪ Lab-scale melter tests

- Objective

  Basic research on noble metals behaviour in HLLW-glass melts

  Phase formation, particle shapes, compositions etc.,

- Several test runs have been performed or are planned, respectively

  - with Pd and Rh only (test already carried out)
  - with Ru only (planned)

- Result of the test with Pd and Rh

  A high viscous Pd-Rh enriched glass layer was formed near the bottom of the lab-scale melter, although no RuO_2 was present.

- Current lab-scale melter: 45° inclination
  Alternative lab-scale melter: 60° inclination
  (1st test performed)

- Additional plans for lab-scale tests

  - glass frit beads replaced by glass frit powder
  - use of reducing agents

KfK-INE

■ Optimization of the off-gas line and of remote melter components

- Most probably the WAK-HAWC must be vitrified at the WAK-site
- In this case the German licencing procedure for the process must be applied
- The off-gas line is expected to become the major licencing point. Very low release limits and high safety standards for the off-gas line are required
- Operational safety of the whole vitrification plant must be considered too in the light of German regulations
- Remote melter components will become subject for change and optimization too (according to the melter cell configuration and cell dimension at the WAK-site)

KfK-INE

■ Glass characterization
(for glasses expected from abroad for final disposal in Germany)

- Physical properties of the expected glasses are provided or already available from Cogema and BNFL, resp.

- Detailed glass corrosion studies under defined final disposal conditions are made in Germany and are continued

- Results of the investigation of chemical stability of simulated "Cogema glass" become available in the near future (HMI-work 1987-90)

- Actual high active Cogema-glass R7 T7 will be investigated in a Mg-rich salt solution, INE programm from 1991-94

  Times (days): 1300, 1000, 180, 130, 45
  Temperatures (°C) : 110, 150, 190

- Similar investigations will be started with two different simulated "BFNL-glasses" in three different salt solutions. Variation of time and temperature. Programm will start 1991 and last until late 1993.

KfK-INE

■ Glass characterization

(for WAK-HAWC glass, to be produced in PAMELA or in a new facility at the WAK-site)

- Two glass frit compositions are currently candidates for the final frit
- Melting parameters of the glass product are acceptable for both frits (el. resistivity, viscosity), 15 wt% waste oxide loading
- Glass development is continued, knowledge of exact composition of the actual WAK-HLLW will be made available by WAK for this purpose
- Complete physical characterization of the selected glass will be started (crystallization, etc.)
- Chemical stability will be investigated similar to "Cogema glasses" and "BNFL glasses"

KfK-INE

■ Canister Corrosion Experiments

- Objective: long-term corrosion resistent package (several hundred years) for vitrified waste as a barrier during the high-temperature time period (> 100°C) of a final disposal repository (salt)

- Candidate materials selected

  Unalloyed steel (reference)
  Ti 99.8-Pd
  Hastelloy C4

- Detailed lab-scale and in situ (Asse) experiments are performed

Influence of Time	(40 days, 1 year)
Temperature	(90, 150, 170, 200°C)
Solutions	- 33% $MgCl_2$ - 25.9% NaCl - $MgCl_2$ rich with 27% $MgCl_2$ (Q-solution)
Gamma-field	10 Gy/h

- Project is continued, individual results are available

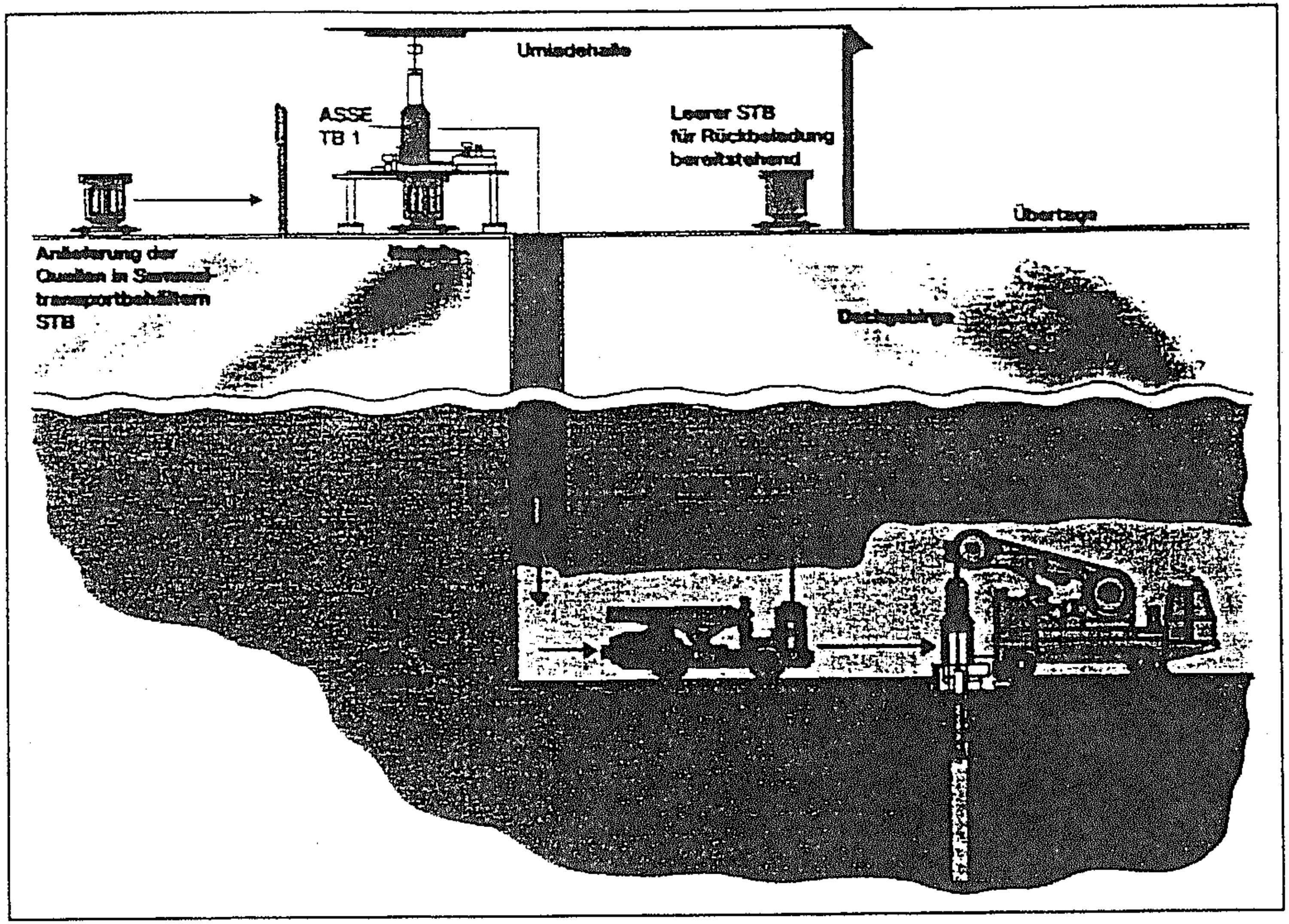


Abb. 2
Prinzipielle Darstellung des HAW-Handhabungssystems.

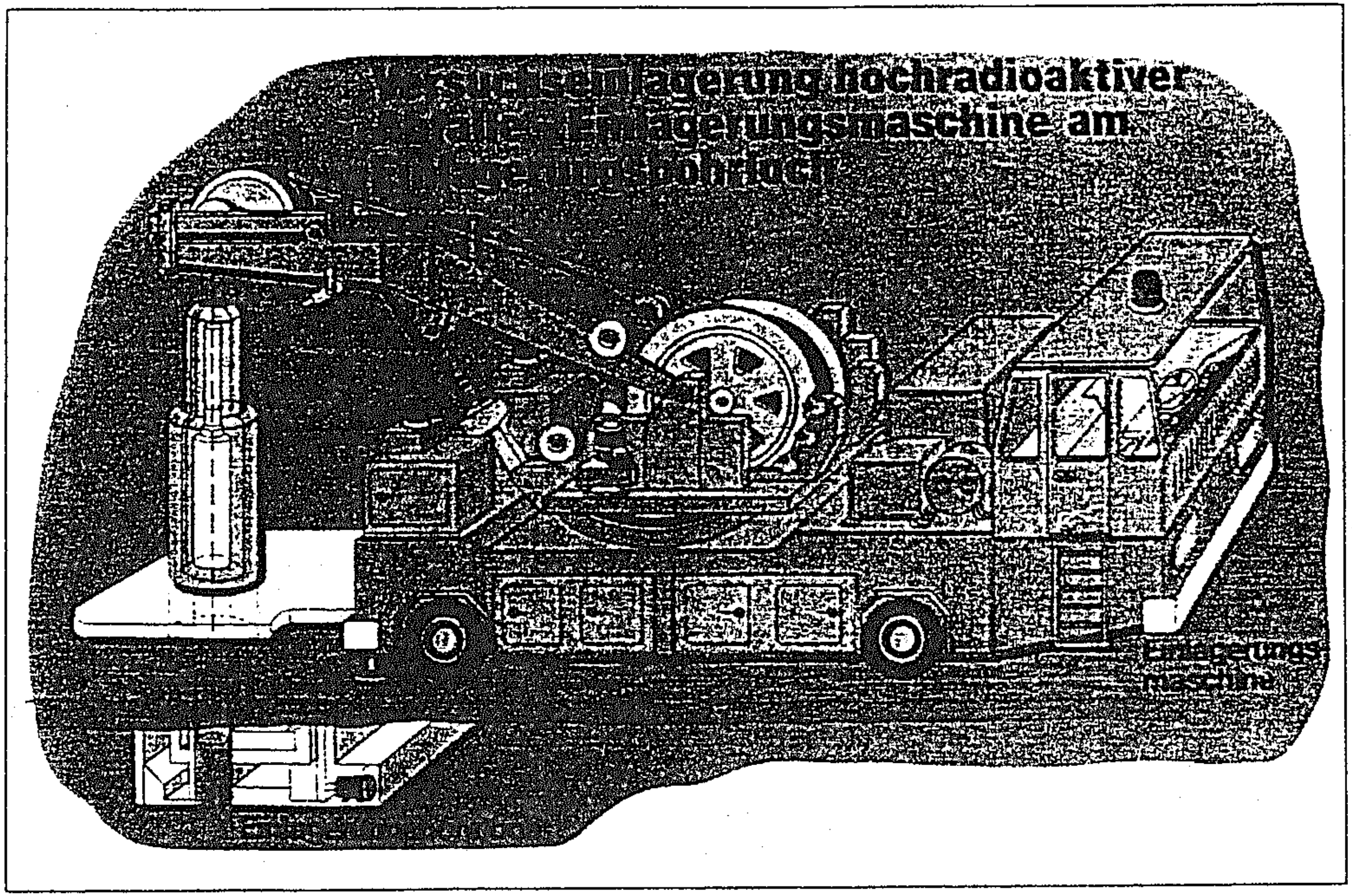


Abb. 10
Graphische Darstellung der Einlagerungsmaschine mit Abschirmglocke. Die Einlagerungsmaschine dient zum Einbringen und spateren Rückholen der Versuchsquellen in bzw. aus den Einlagerungsbohrlöchern.

KfK-INE

■ Near field investigations of HLLW-glass and spent fuel in a final repository

- Mathematical modelling of the near field
- Thermomechanical calculations
- Temperature field calculation
- Test disposal of 40 glass cans produced by BPNL in Richland, WA in 1984/85; can transportation licence from US to Germany very difficult to get

WAK

■ Licencing procedure for transportation of liquid HLW from Karlsruhe to Mol

- Licencing authority is the "Baden-Württembergische Sozialministerium", BWS, Stuttgart
- To get a licence for transportation of liquid HLW is very questionable
- In case the licence will be given, it is expected to take 4-8 years (Problem, what to do with the PAMELA plant and staff in the meantime, PAMELA will finish HEWC-vitrification in 1991)
- Transport planned via railway, batches may be ≤ 1m³ HLLW
- Approximately 100 m³ HLLW are to transport and vitrify
- Extremely high safety requirements and thus costs for the WAK-filling station are expected (earthquake, plane crash, etc.)
- Substantial costs for the refilling station at PAMELA are also a difficult issue
- For the time being, WAK has made, on behalf of KfK (as the owner of WAK), an application to BWS to get a licence
- If the WAK-HAWC would be vitrified in PAMELA, the disposal repository for the resulting 50 tons of glass must be finally decided (130 glass blocks)
- Background: Near PAMELA plant, Belgium has a final disposal site under design for glasses coming from abroad

WAK

■ Studies for vitrification of the WAK-HAWC at the site

- Unused hot cells (LAVA-complex) would be available and probably suitable for installation of a LFCM vitrification process
- Melter size appr. 60-80% of that of PAMELA melters
- Modifications of the cell structure may be necessary
- Vitrification process could be installed in the modified cells but a few disadvantageous compromises had to be made
- Alternatively, a new process building may also be considered in the near future
- Except the high costs for process installations and eventually a new cell-building, some substantial advantages exist to do the vitrification job for WAK-HAWC on site
- The lowest cost solution, however, would be PAMELA, according to the present knowledge

PAMELA

■ Status of vitrified HLLW (LEWC, HEWC)

- A few days ago, the 2000th high active glass block has been poured
- Melter K-5 operates with approximately 30-33 l/h
- Total amount of vitrified HLLW: 720 m^3
- Rest of HEWC (130m^3) currently vitrified
- End of operation expected for summer 1991
- Tank rinsing period (5 tanks) and vitrification of rinsing solutions are scheduled for the 2nd half of 1991
- It is hoped, that the K-5 melter can outlast the rest of the PAMELA vitrification period
- The K-5' melter, currently having the function of a spare part, would then be unnecessary
- Festival for 2000th glass block

  Tuesday, Nov. 20, 7 p.pm. at Mol

PAMELA

■ Melter decomissioning

- Two radioactive applied melters have to undergo decomissioning

K-4	operated	1985-88
K-5	operation since	1988, expected until 1991 (summer)

- Time schedule for decomissioning not yet finally fixed

- Low cost decomissioning is required. Nevertheless, inspection of melter components will be included, but there will be no comprehensive sampling and analysis work as planned previously

- In the focus of inspection

  Failed power electrodes of K-4 melter
  Failed bottom drain area of K-4 melter
  Bottom of K-5 melter
  Condition of power electrodes K-5 melter

3-2 Operation Results with The K-6' Melter

Operation results with the K-6'melter

- Status of the HAWC-WAK vitrification project
- The new vitrification plant VA-WAK operating the molter K-6'
- Firet operation runs

WAK

* Operation terminated in 1991
* Total HAWC-volume 83 m^3 (stored in 2 tanks)
* Licence for construction and operation of the waste filling station should be given in 1993

PAMELA

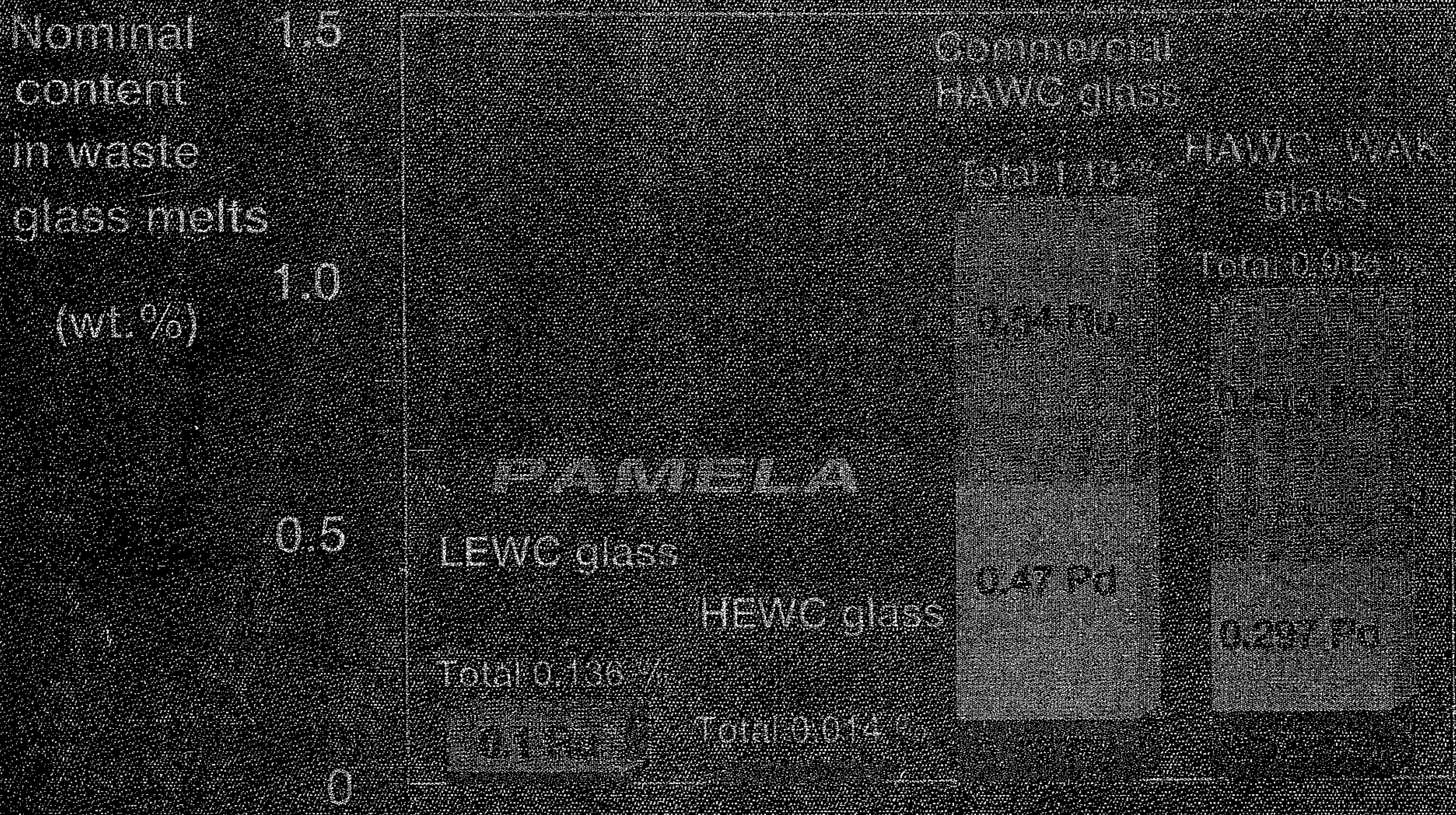
* Since Nov. 1990 about 720 m^3 HLLW (in total) vitrified
  Rest amount of 130 m^3 HEWC will be processed until spring 1991
* Integration of the new melter K-6
* Start vitrification of HAWC-WAK waste in 1993 (original schedule)

INE

* First vitrification of HAWC-WAK simulate without noble metals in the VA-2 plant
* Construction of the new 1:1 mock-up plant VA-WAK until end of 1989 operating the melter K-6'
* First noble metal campaign in 1991

Jap.'90

Status of the HAWC-WAK vitrification project



Comparison of nominal noble metal content in waste glass melts

* Vitrification experience with HLLW simulate

- **Vitrification of HAWC-WAK simulate in the VA-2 plant in December 1987**

  3.8 m^3 Simulate without noble metals
  2.1 to Glass product (Frit SWA 752 FR)
  13.9 % Glass loading
  21-23 l/h Feeding rate
  Processing without major problems

- **Vitrification of HAWC-WAW simulate in the V-W1 plant in 1988 and 1989**

  63 m^3. Simulate incl. noble metals
  25 to Glass product (frit VG 98/12.2LiW4)
  13.5 % Glass loading
  ca. 50 l/h Feeding rate
  Experimental results require a new melter design

* Objects for the VA-WAK operation

- Demonstration of the K-6' melter function
- Demonstration of the target feeding rate of 25 l/h
- **Testing of an improved glass frit**
- **Cleaning efficiency of the off gas line**

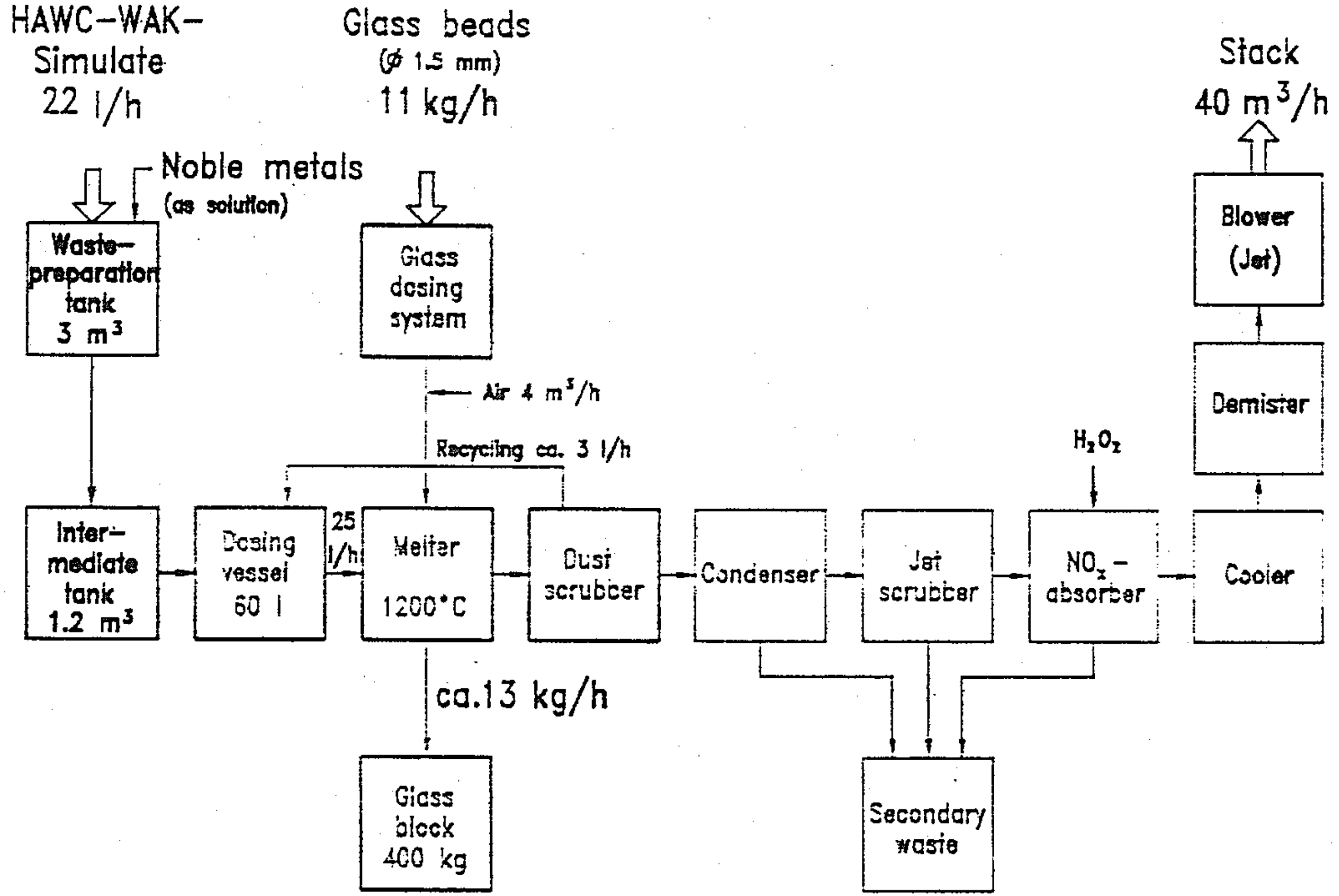
------------------------------ Jap.'90 ------

Objects for the K-6' operation, vitrifying noble metal containing HAWC-WAK simulate

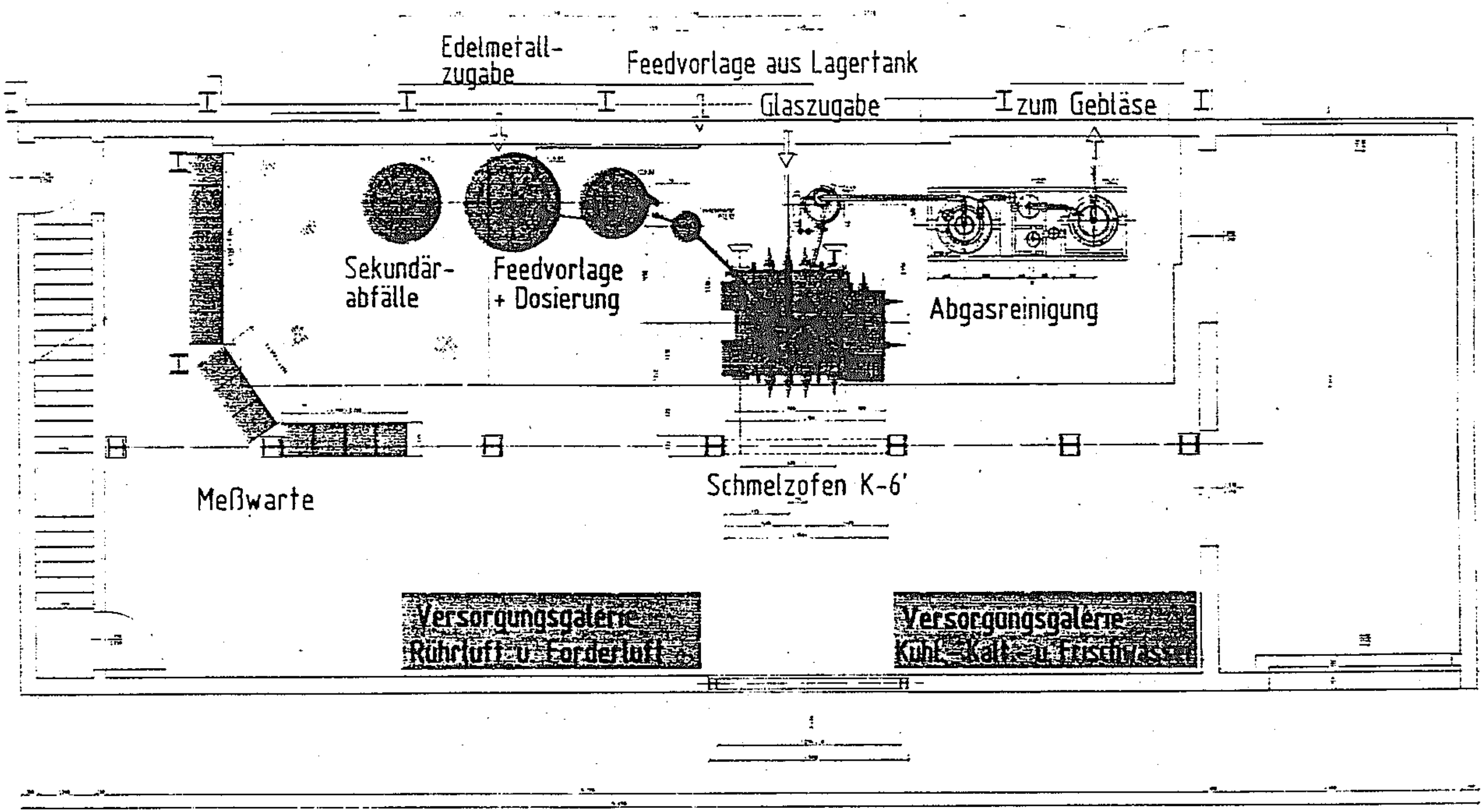
Operation run	D0	D1	D2
Time period	21.–25.5.	25.5.–4.6.	4.6.–24.6.
Operation time	102 h	233 h	472 h
Interruptions	17.5 h	–	–
Availability	83 %	100 %	100 %
Waste simulate	H_2O	HAWC–WAK	HAWC–WAK
Ru content (target)	–	–	2.91 g/l
Pd content (target)	–	–	1.79 g/l
Oxide content	–	75.71 g/l	81.09 g/l
Glass frit	SWA 752FR	SWA 752FR	SWA 752FR
Glass loading (target)	–	13.13 %	13.96 %
Glass loading (exp.)	–	12.9 %	14.0 %
Type of canister	COGEMA	Cogema	Cogema
Σ Waste simulate	2.3 m^3(H_2O)	4.9 m^3	10.3 m^3
Σ Ru	–	–	30 kg
Σ Pd	–	–	18 kg
Σ Glass frit	0.4 to	2.5 to	5.1 to
Σ Glass product	0.3 to	2.7 to	6.1 to
average feeding rate	29 l/h	25 l/h	25 l/h
Σ Glass samples	23	70	512
Σ Liquid samples	–	67	131

Jap.'90

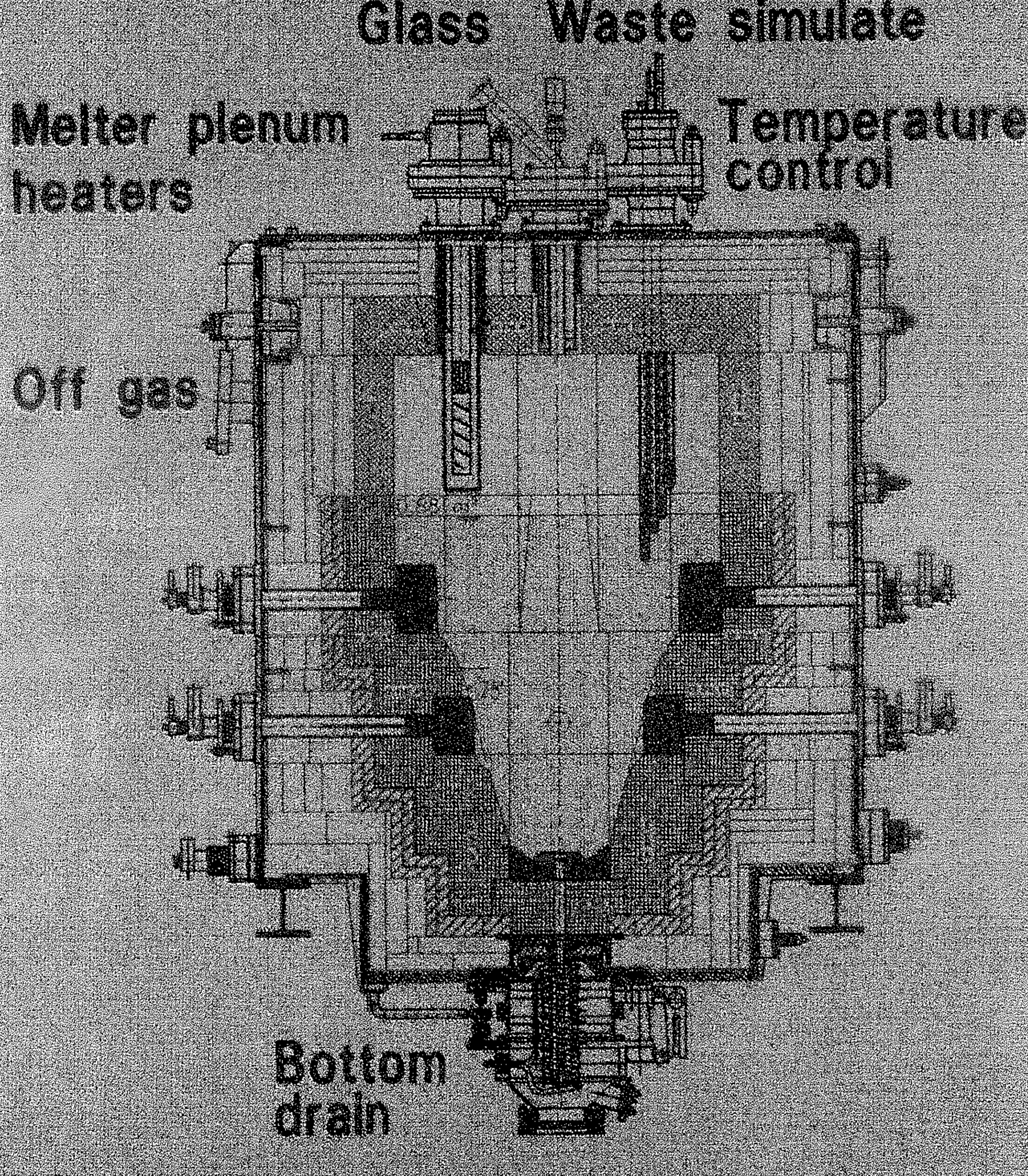
Operation of the VA–WAK plant



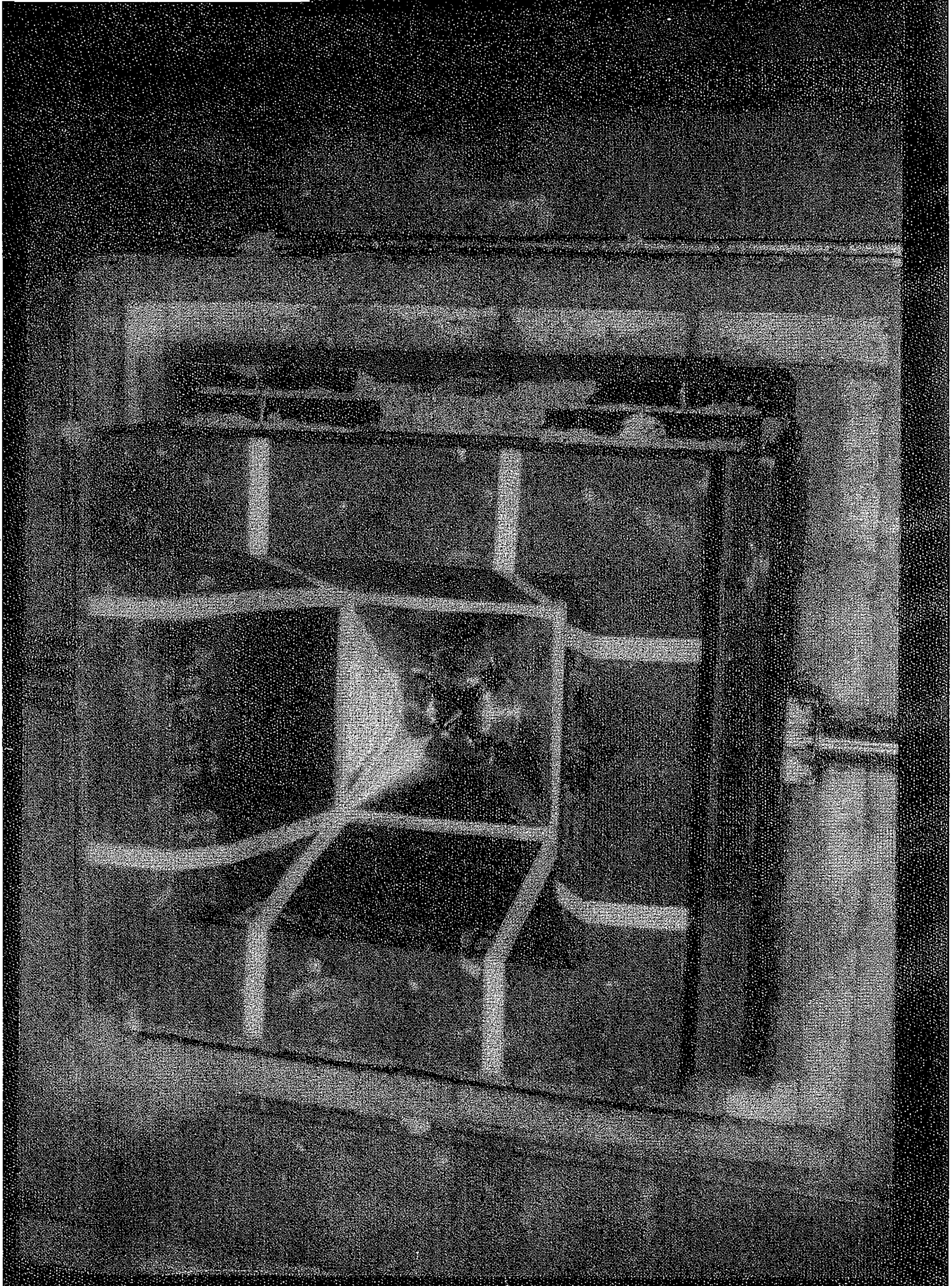
Block diagram of the vitrification plant VA−WAK

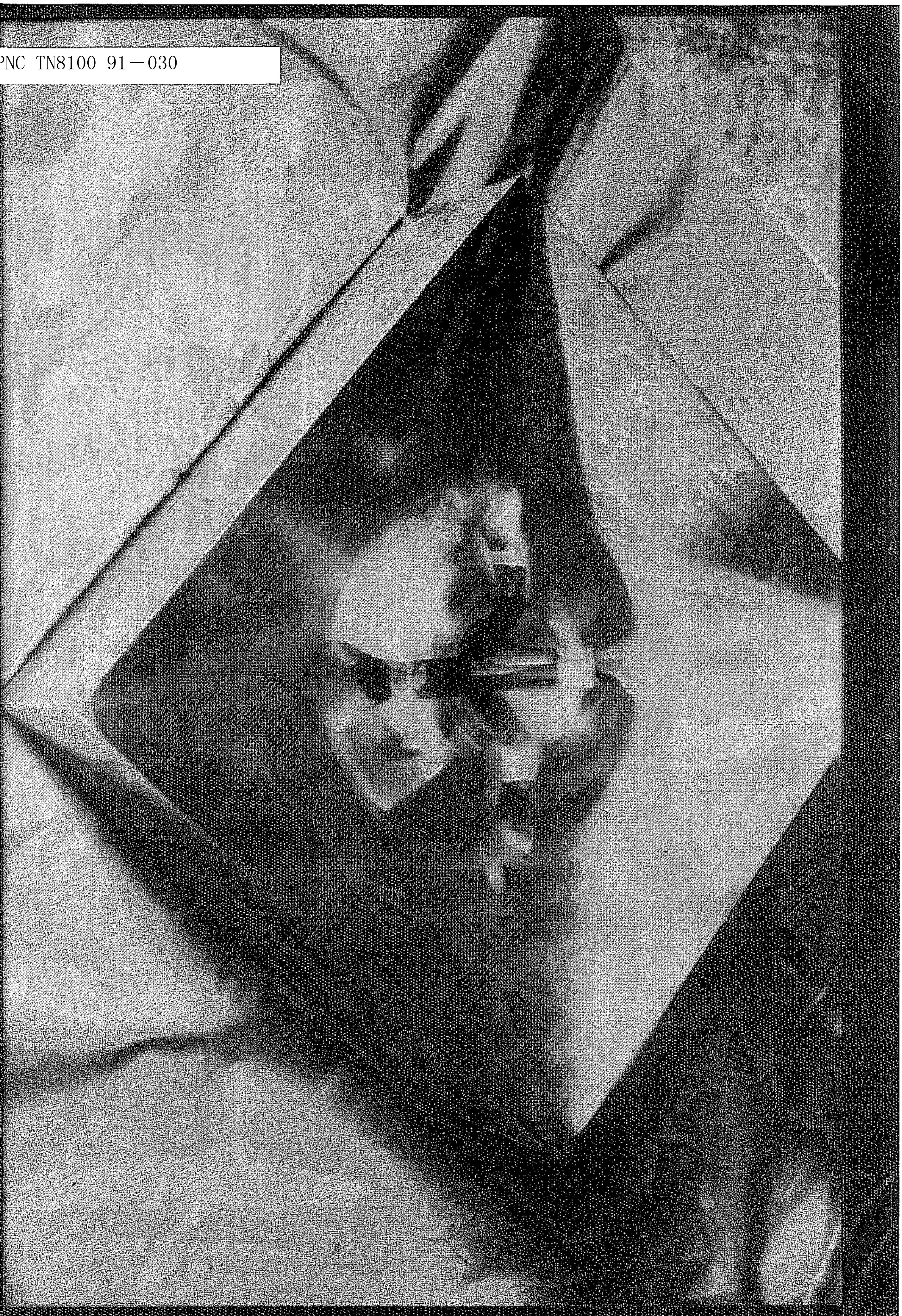


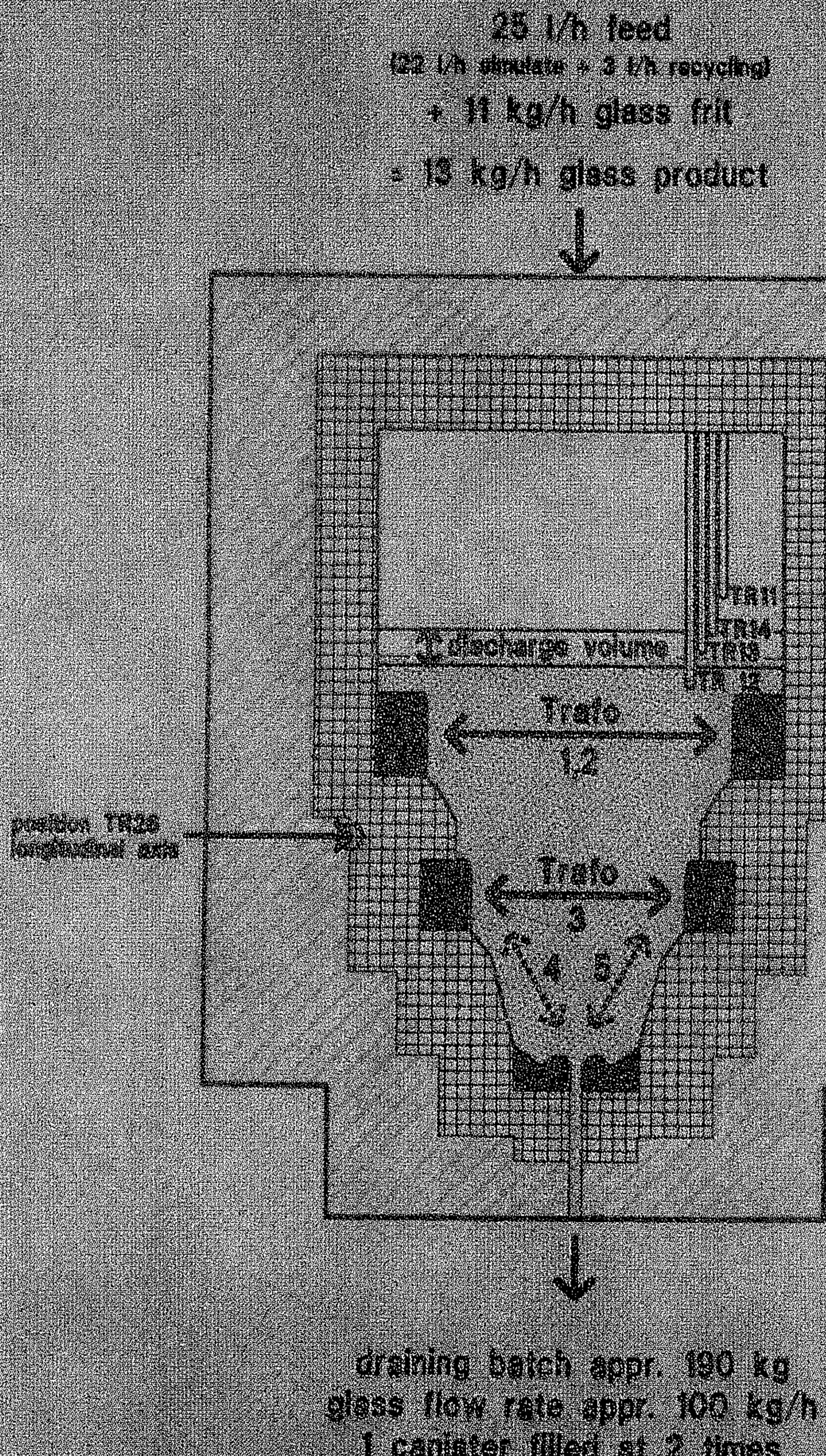
Aufstellungsplan der Verglasungsanlage VA-WAK (Geb. 551)

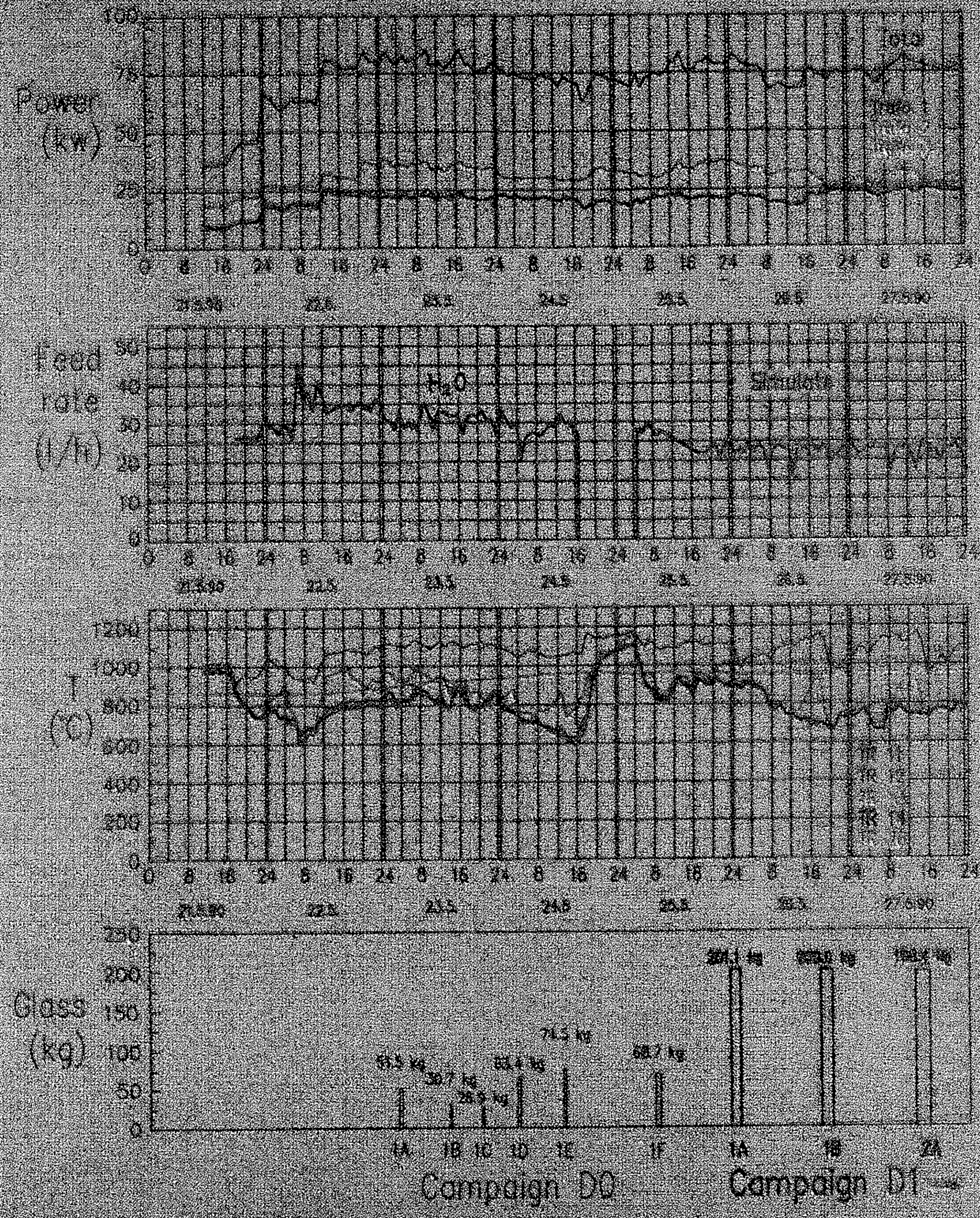


Melter design K-6'

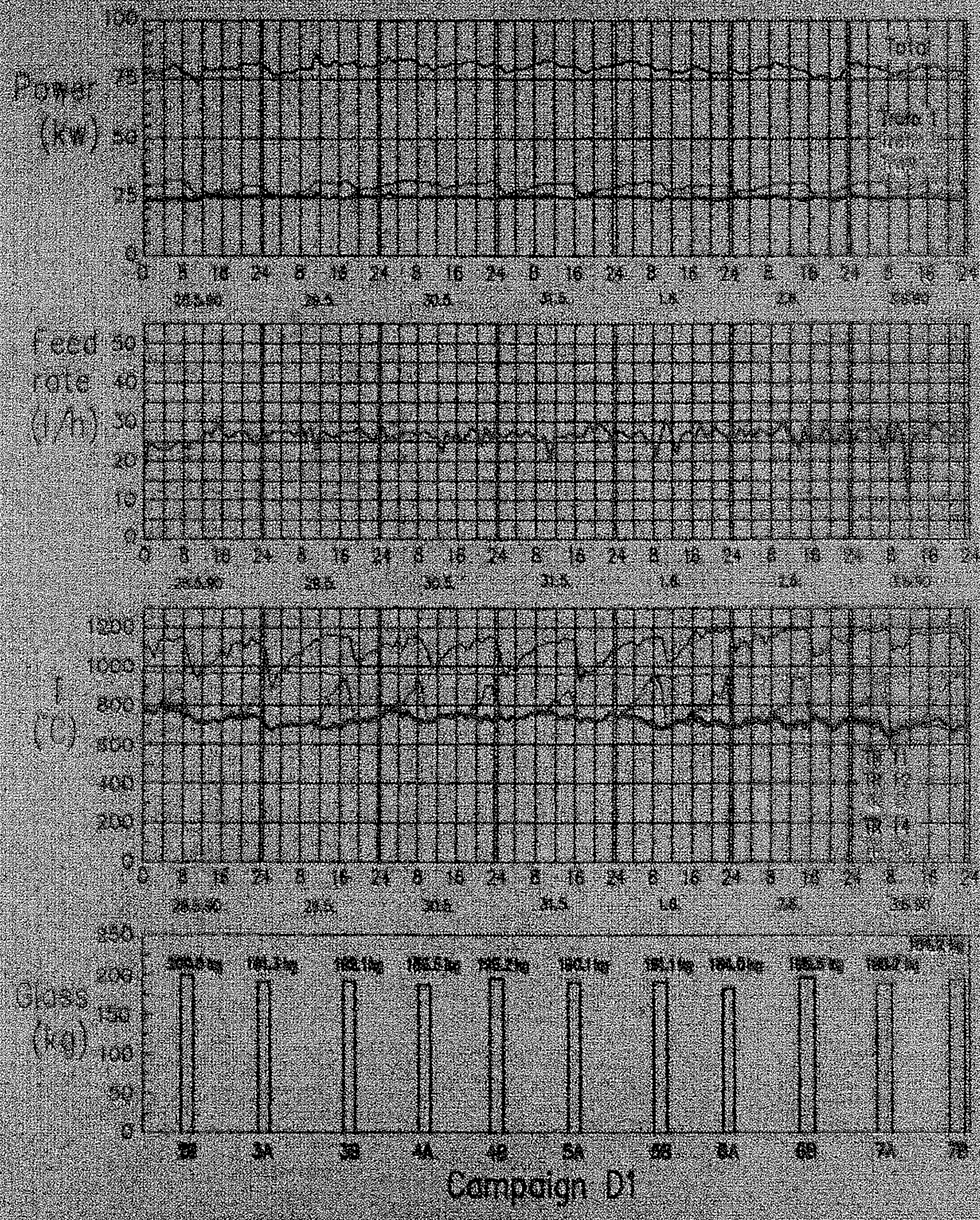




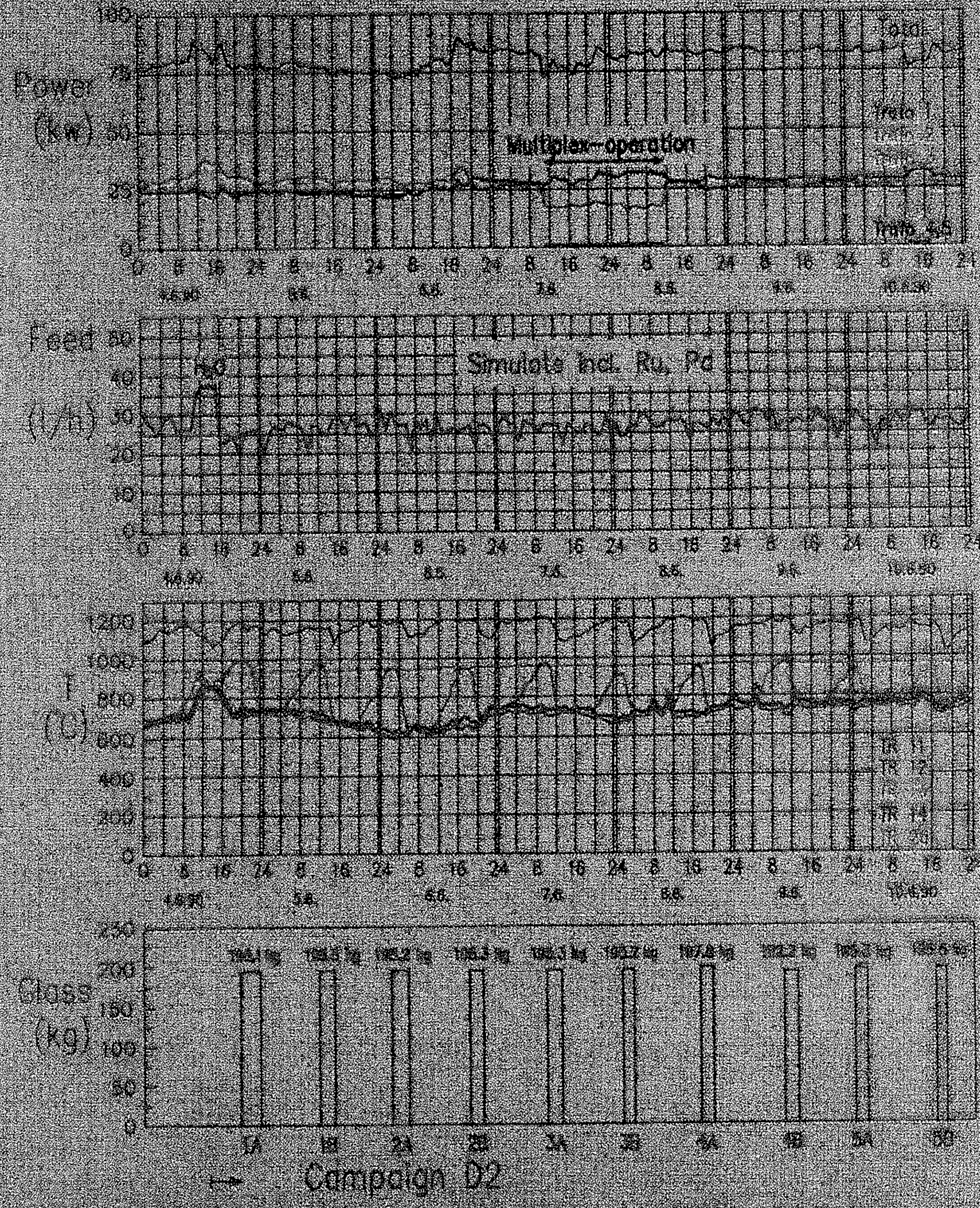

25 l/h feed
(22 l/h simulate + 3 l/h recycling)
+ 11 kg/h glass frit
= 13 kg/h glass product
TR11
TR14
TR 12
discharge volume
Trafo
1,2
position TR26
longitudinal axis
Trafo
3
4
5
draining batch appr. 190 kg
glass flow rate appr. 100 kg/h
1 canister filled at 2 times



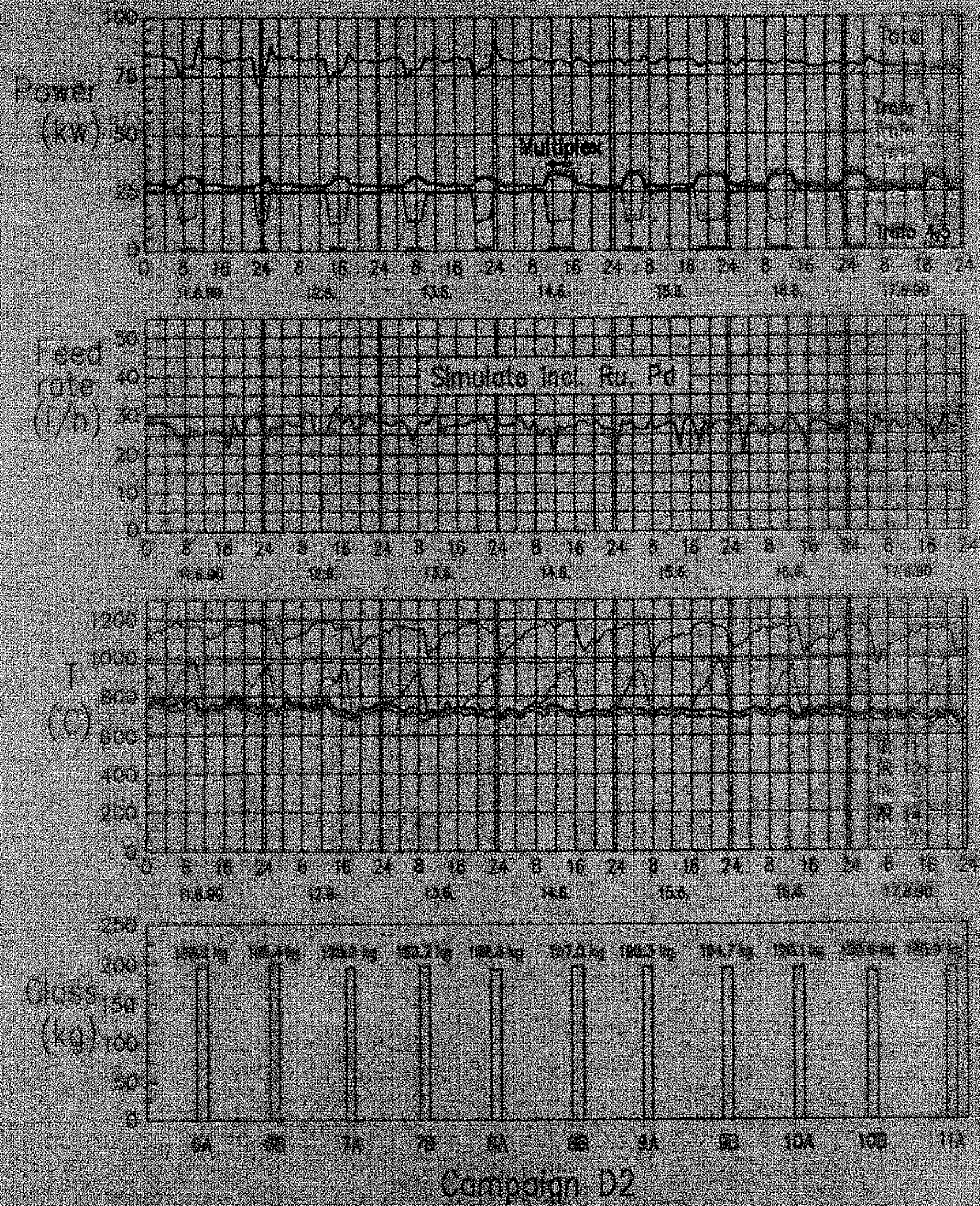
Operation diagram for the first week of the campaigns D0 – D2



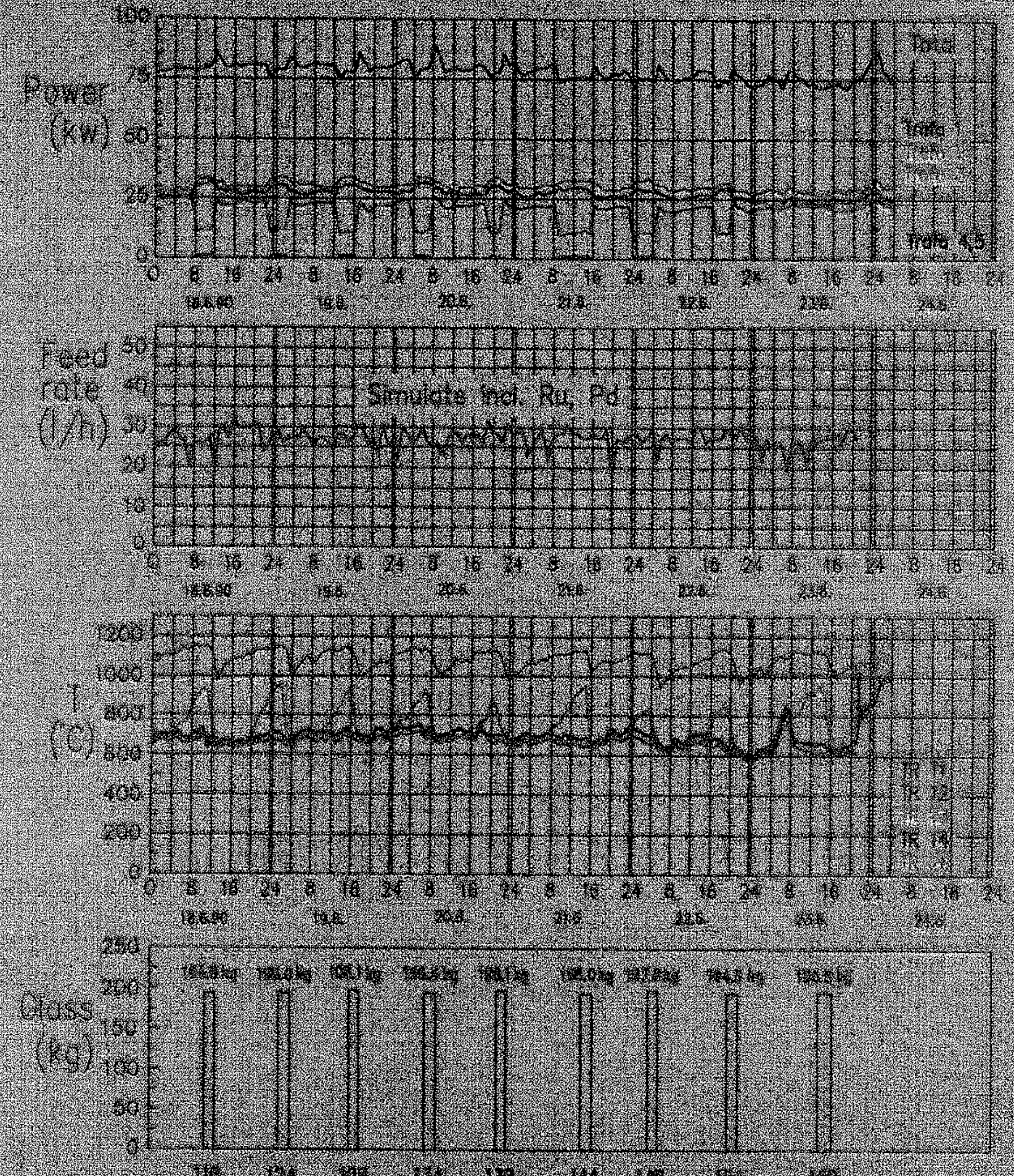
Operation diagram for the second week of the campaigns D0 − D2



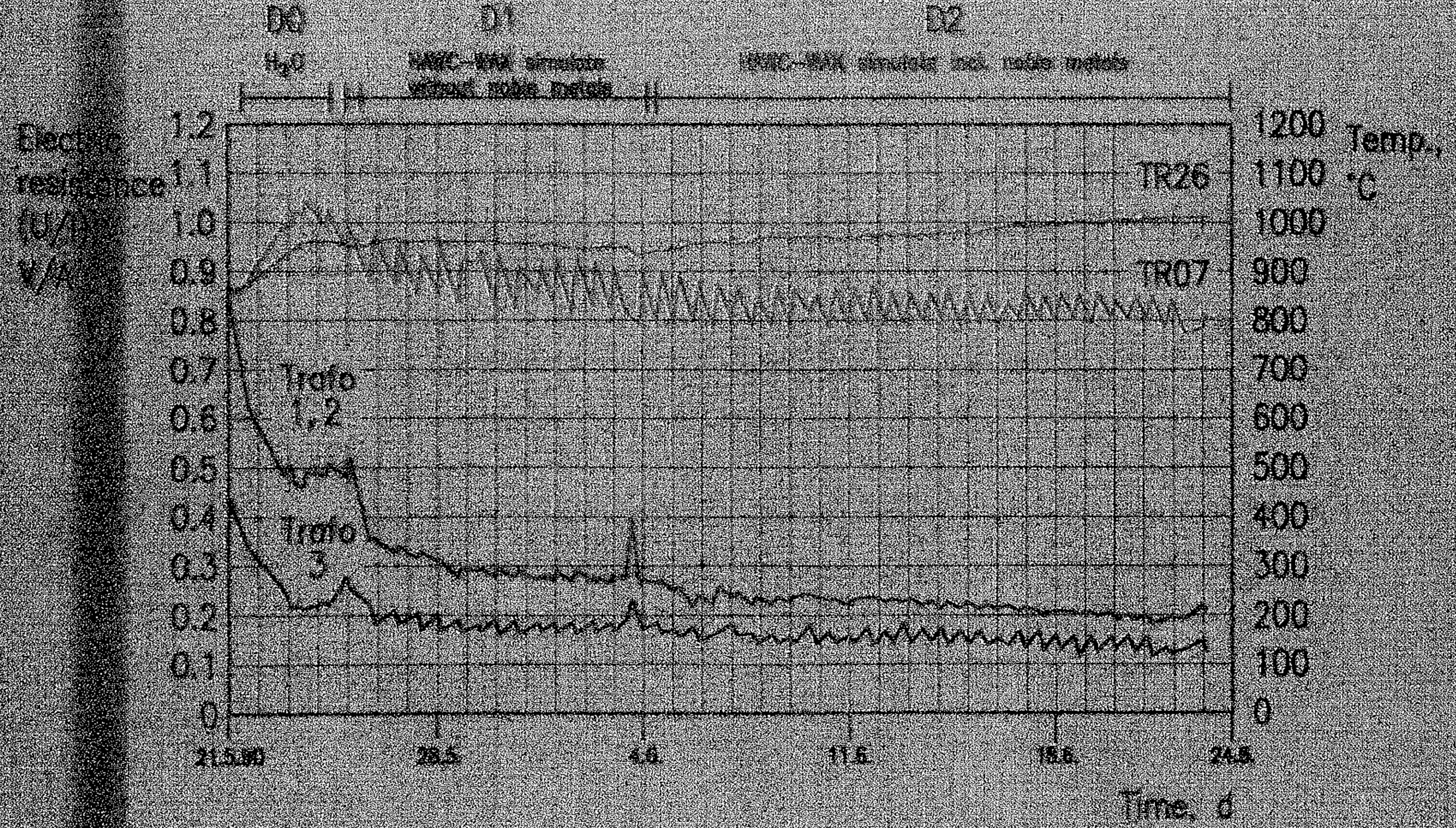
Operation diagram for the third week of the campaigns D0 − D2



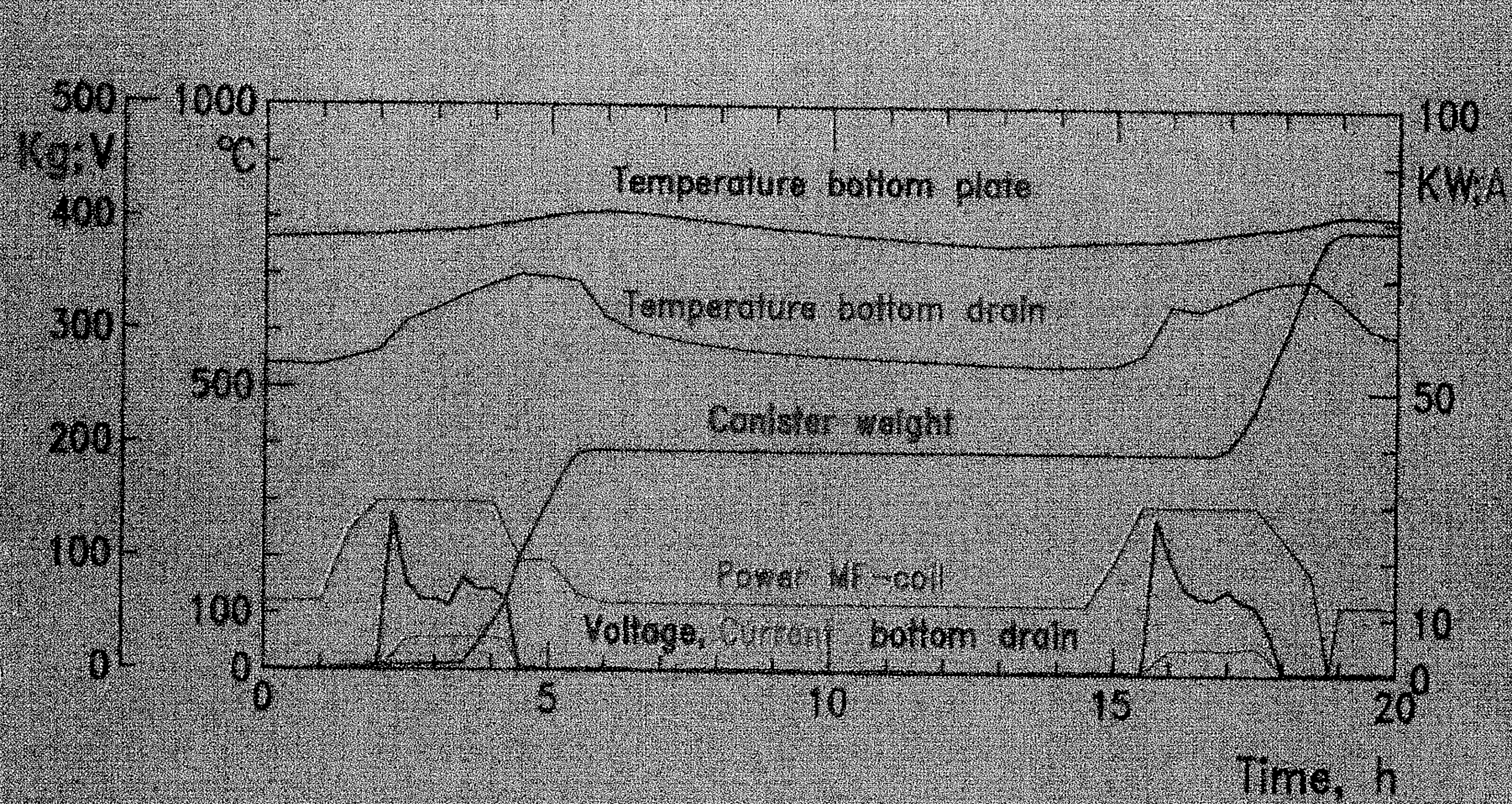
Operation diagram for the fourth week of the campaigns D0 － D2



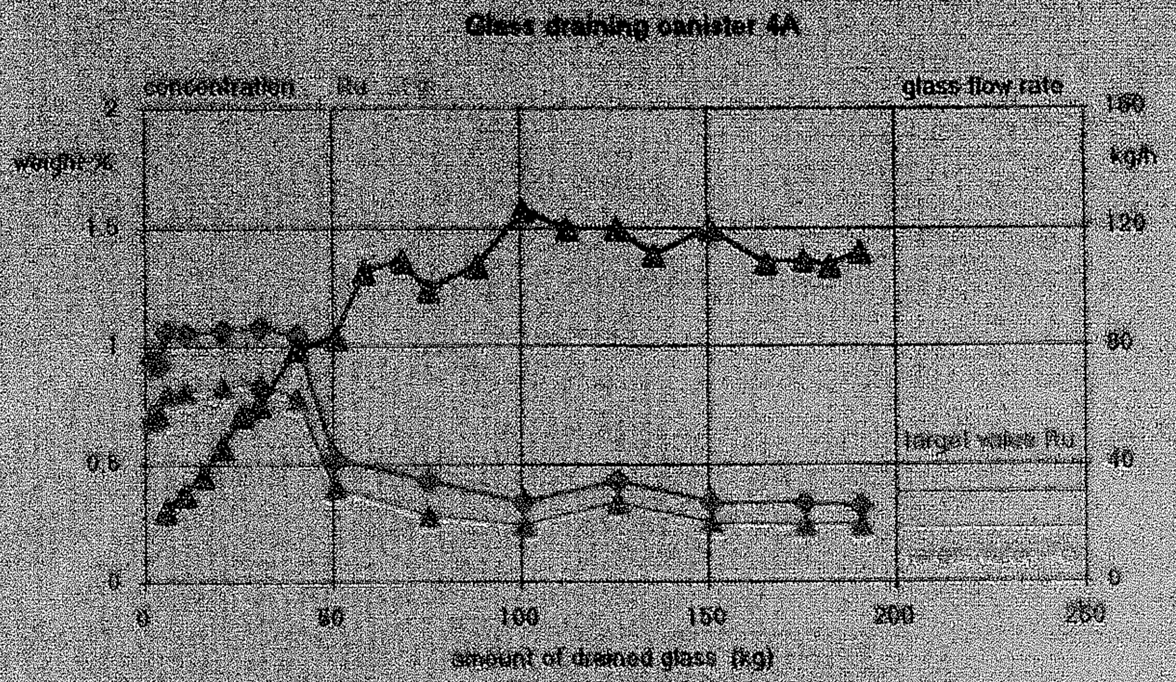
Operation diagram for the fifth week of the campaigns D0 – D2

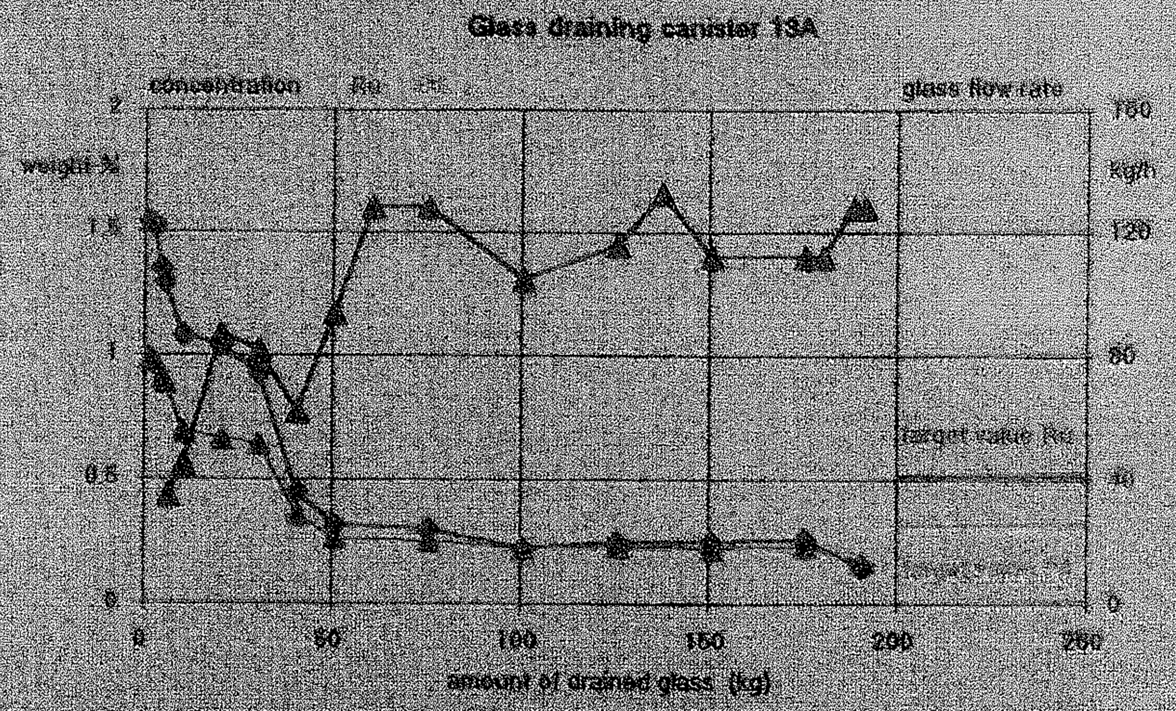


Coarse of the electric resistivity of the glass pool during the operation period



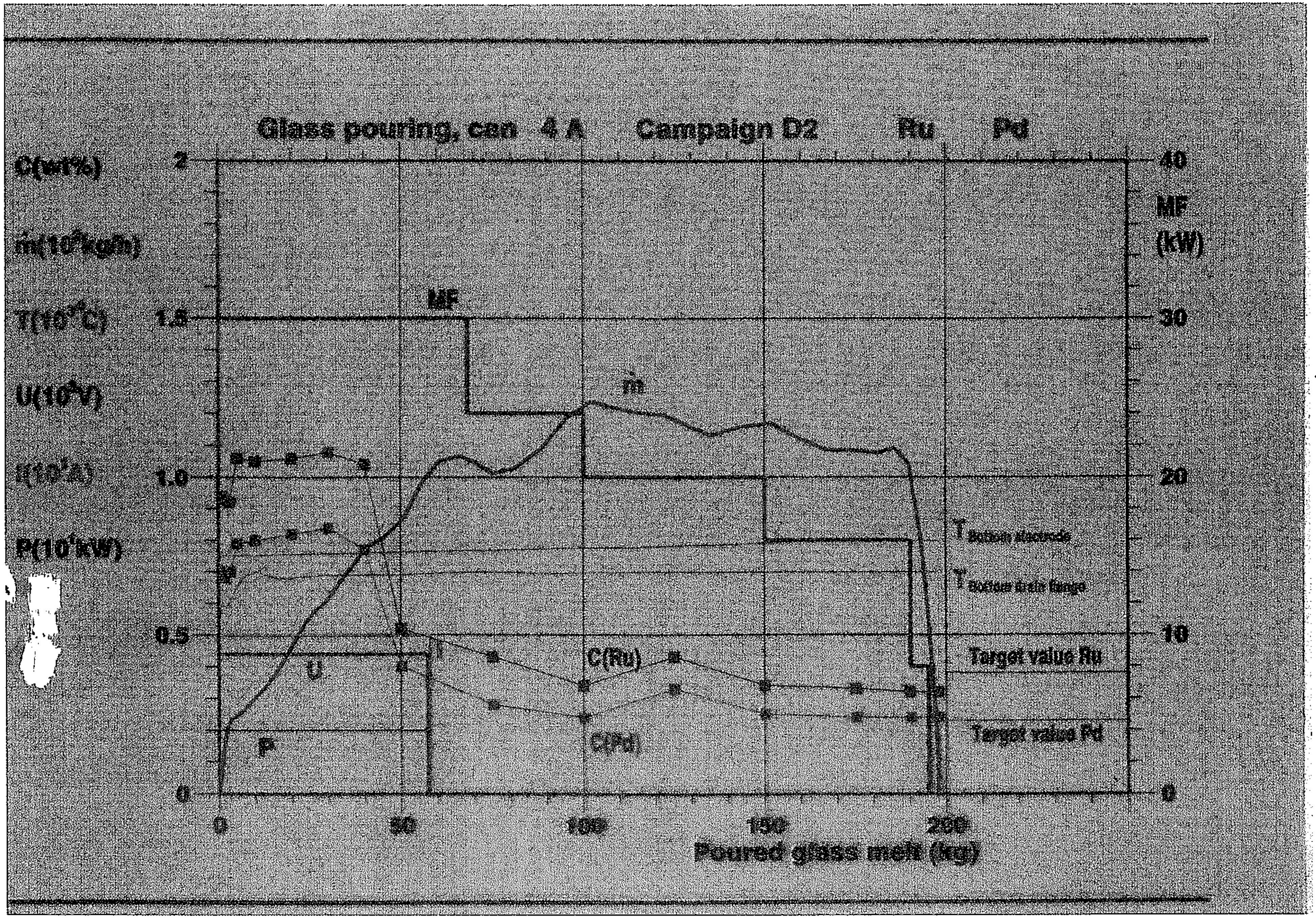
Glass filling data of canister no. D2-14

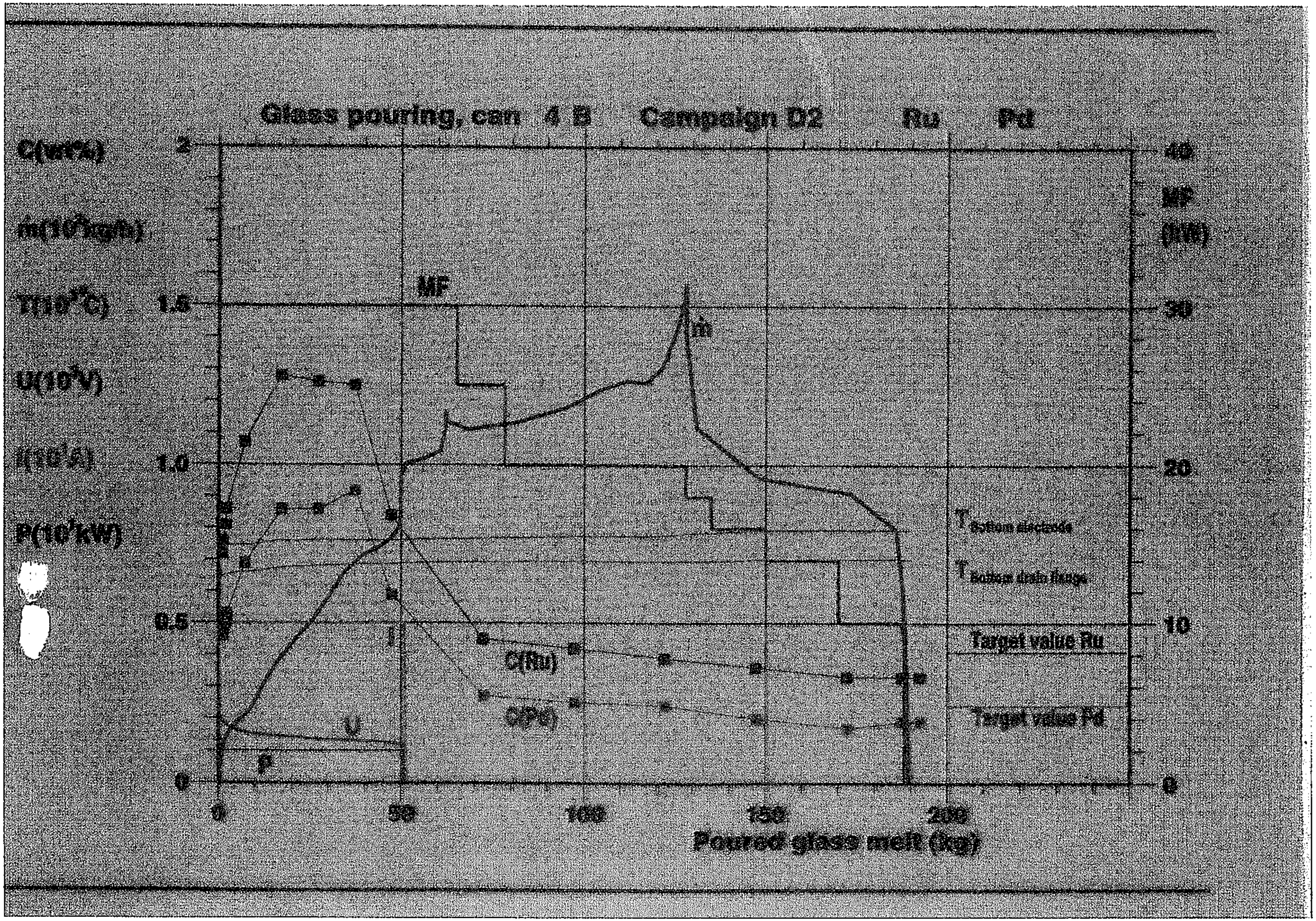




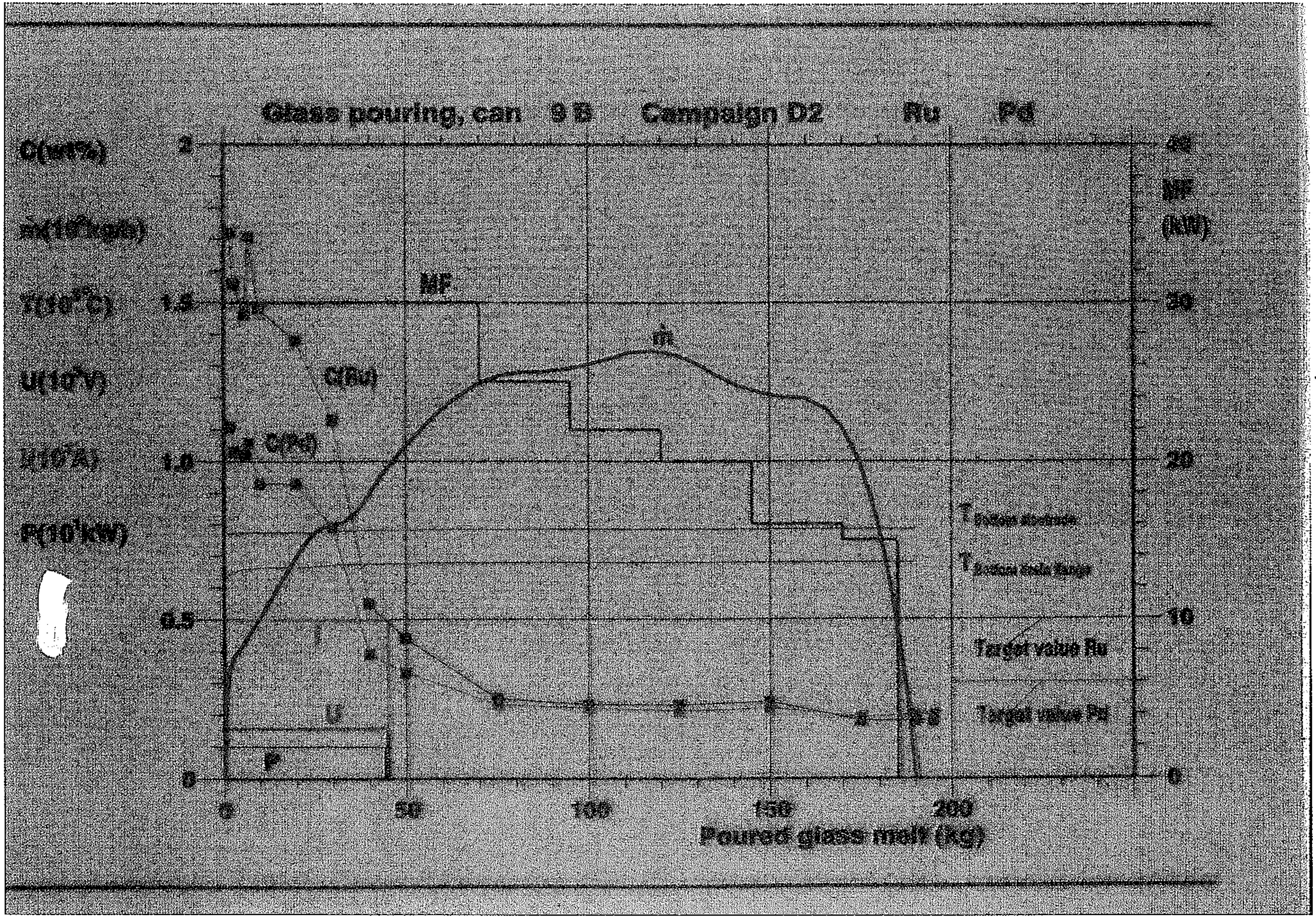
comparison of canister filling no 4A and 13 A

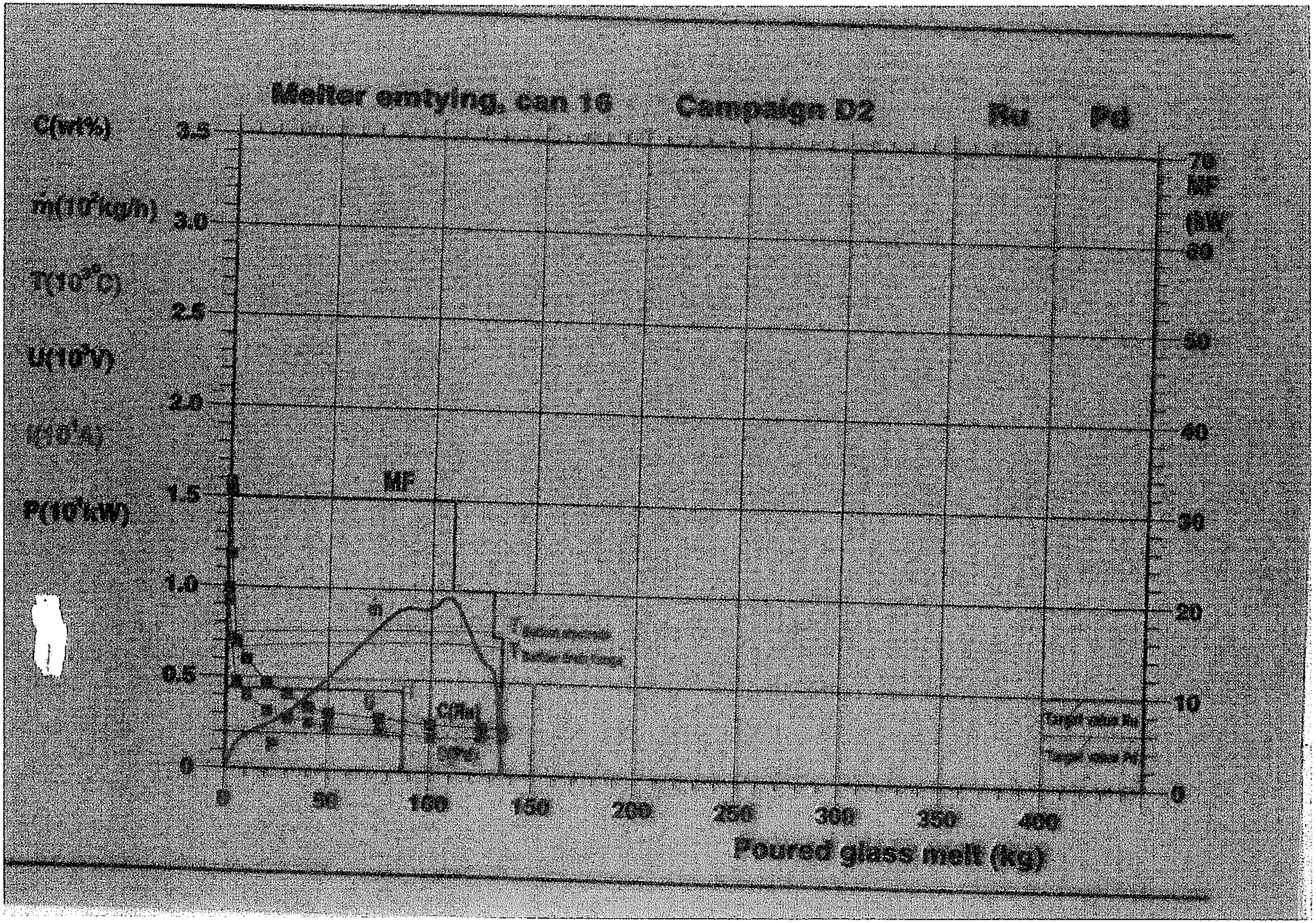
- dependance of glass flow rate

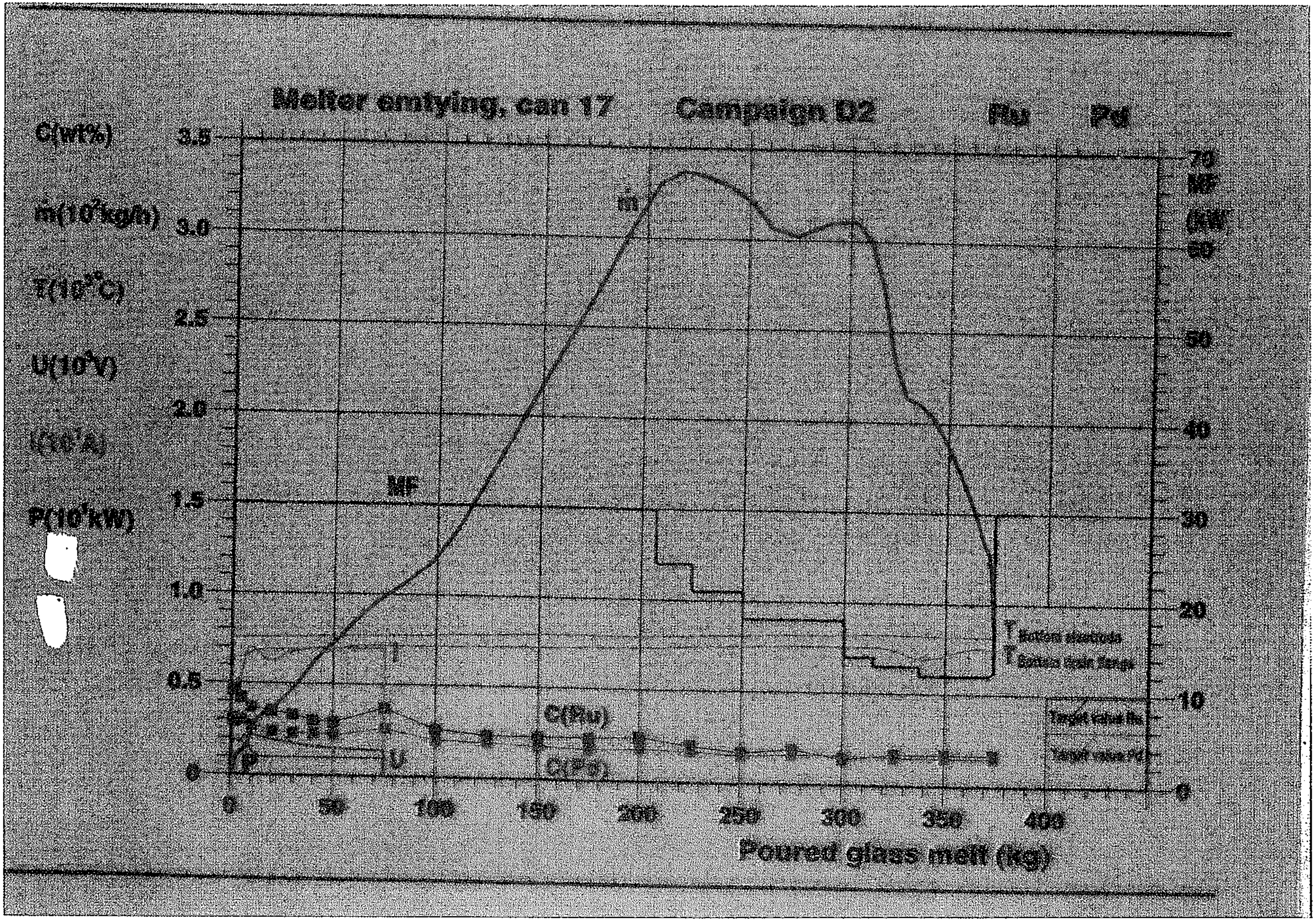

Glass pouring, can 4 A Campaign D2 Ru Pd
C(wt%)
ṁ(10²kg/h)
T(10³°C)
U(10²V)
I(10³A)
P(10²kW)
MF
(kW)
MF
ṁ
U
P
C(Ru)
C(Pd)
Target value Ru
Target value Pd
T Bottom electrode
T Bottom drain flange
Poured glass melt (kg)
0
50
100
150
200
0.5
1.0
1.5
2
10
20
30
40

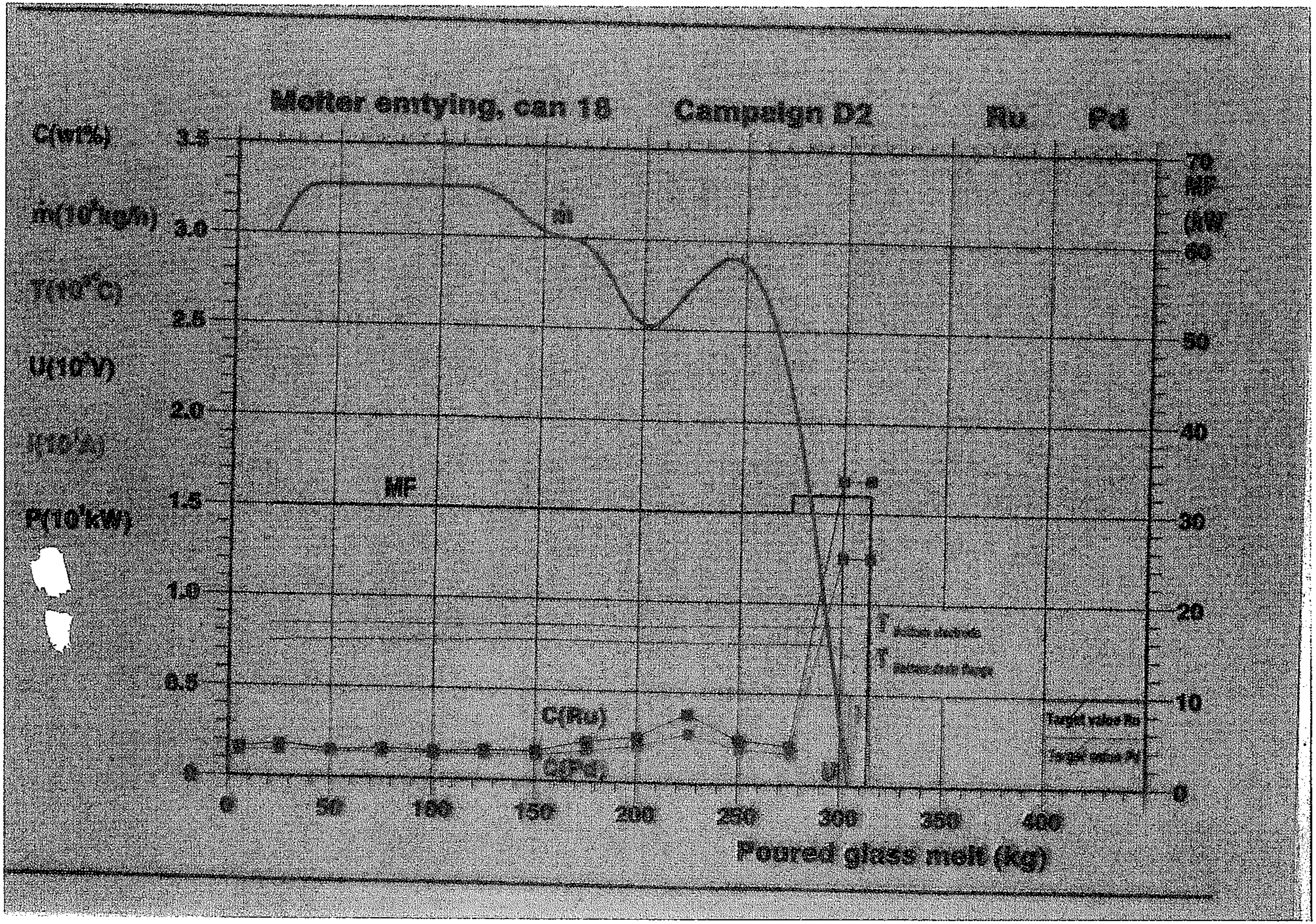


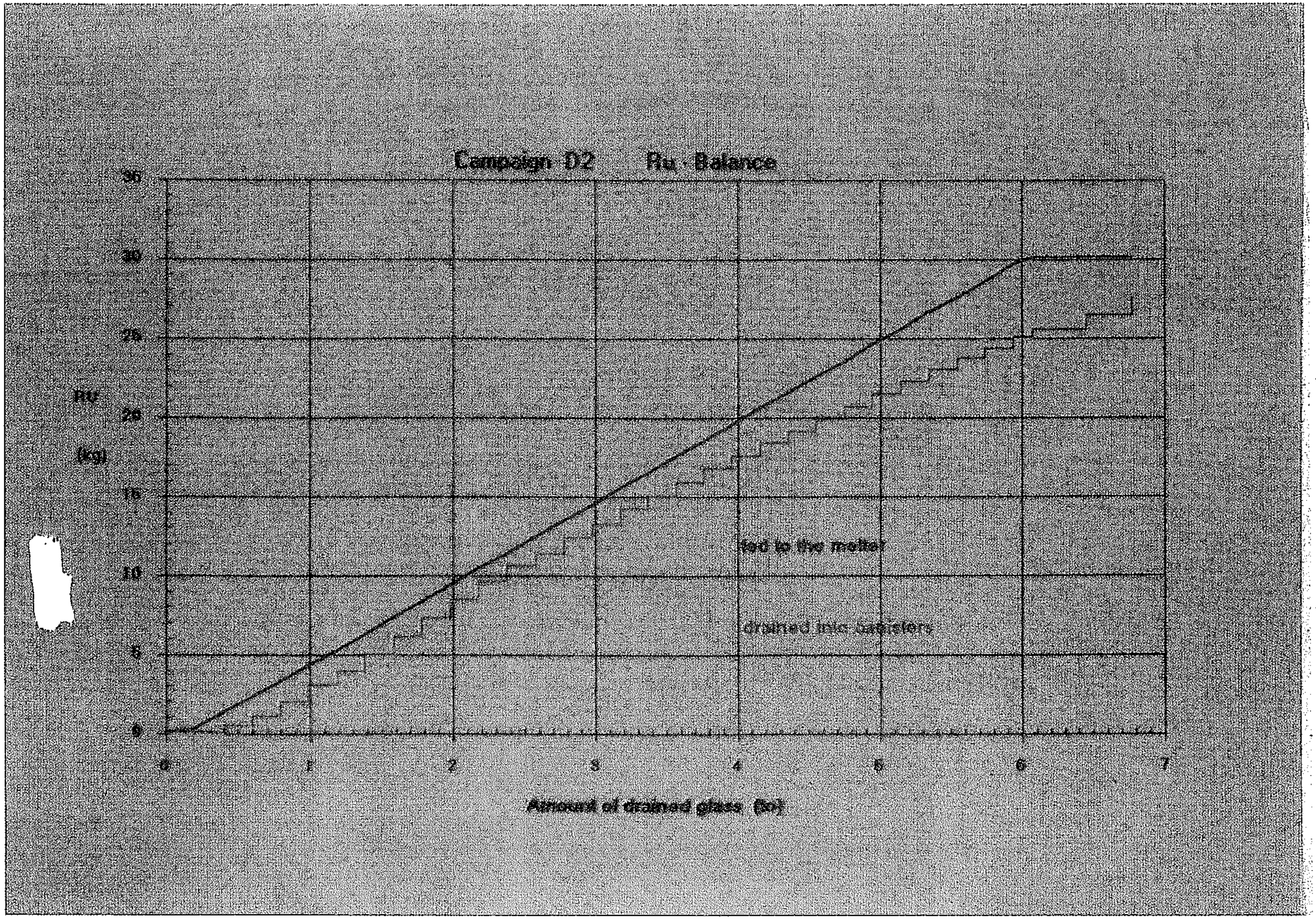
Glass pouring, can 4 B Campaign D2 Ru Pd
C(wt%)
m(10³kg/h)
T(10³°C)
U(10²V)
I(10³A)
P(10¹kW)
MF (kW)
MF
m
U
P
I
C(Ru)
C(Pd)
T Bottom electrode
T Bottom drain flange
Target value Ru
Target value Pd
0
0.5
1.0
1.5
2
0
10
20
30
40
0
50
100
150
200
Poured glass melt (kg)

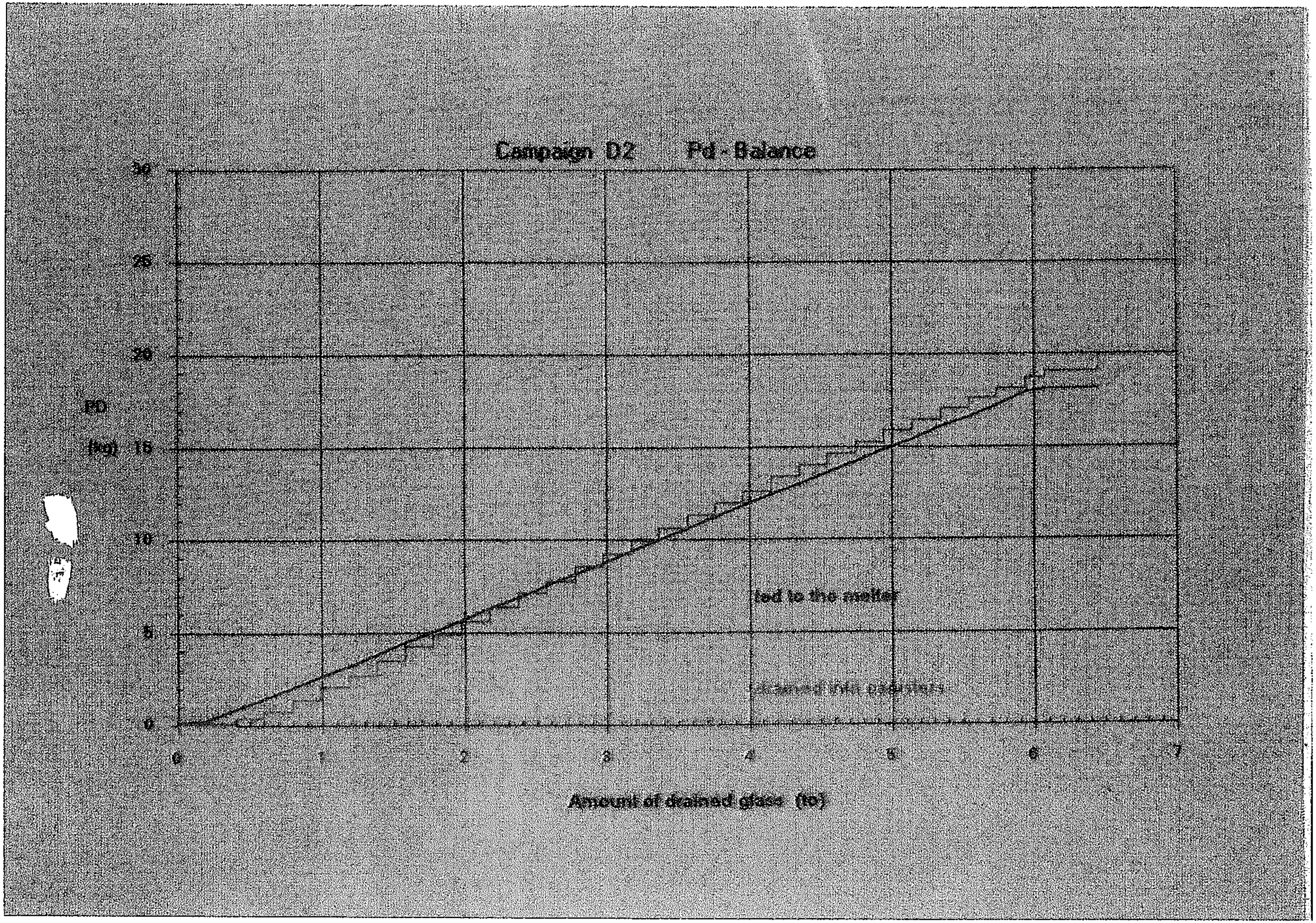

Glass pouring, can 9 B Campaign D2 Ru Pd
C(wt%)
MF
(kW)
MF
C(Ru)
C(Pd)
I
U
P
T bottom electrode
T bottom drain flange
Target value Ru
Target value Pd
0
0.5
1.0
1.5
2
0
10
20
30
40
0
50
100
150
200
Poured glass melt (kg)

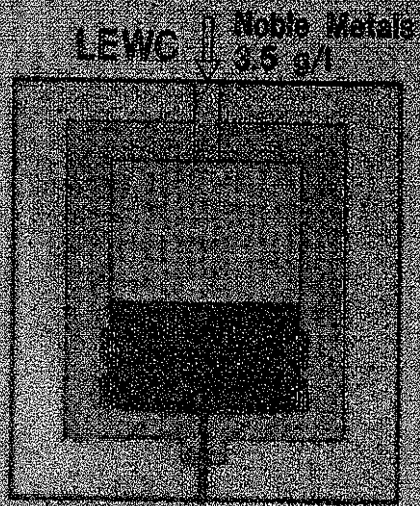

Melter emtying, can 16
Campaign D2
Ru
Pd
C(wt%)
ṁ(10²kg/h)
T(10³°C)
U(10²V)
I(10³A)
P(10²kW)
MF
(kW
Poured glass melt (kg)
0
50
100
150
200
250
300
350
400
0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
0
10
20
30
40
50
60
70
C(Ru)
C(Pd)
Target value Ru
Target value Pd


Melter emtying, can 17
Campaign D2
Ru
Pd
C(wt%)
ṁ(10²kg/h)
T(10⁻³°C)
U(10²V)
I(10³A)
P(10¹kW)
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0
70
MF
(kW
60
50
40
30
20
10
0
ṁ
MF
T
U
P
C(Ru)
C(Pd)
T Bottom electrode
T Bottom drain flange
Target value Ru
Target value Pd
0
50
100
150
200
250
300
350
400
Poured glass melt (kg)


Molter emtying, can 18
Campeign D2
Ru
Pd
C(wt%)
ṁ(10^1 kg/h)
T(10^2 °C)
U(10^2 V)
I(10^1 A)
P(10^1 kW)
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0
70
MF
(kW
60
50
40
30
20
10
0
ṁ
MF
C(Ru)
C(Pd)
U
0
50
100
150
200
250
300
350
400
Poured glass melt (kg)

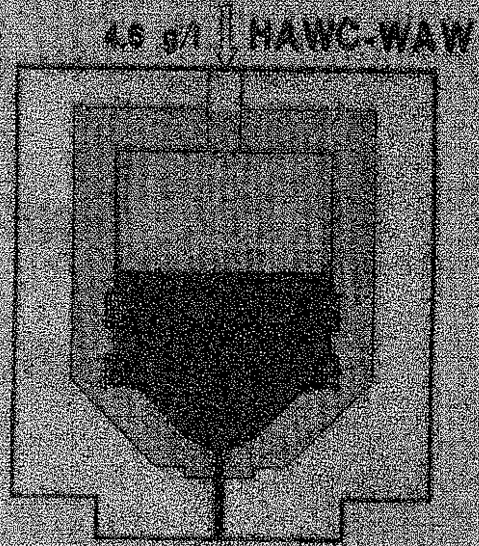

Campaign D2 Ru - Balance
Ru
(kg)
35
30
25
20
15
10
5
0
0
1
2
3
4
5
6
7
fed to the melter
drained into canisters
Amount of drained glass (to)


Campaign D2 Pd - Balance
PD
(kg)
30
25
20
15
10
5
0
0
1
2
3
4
5
6
7
Amount of drained glass (to)



PAMELA Melter K-4

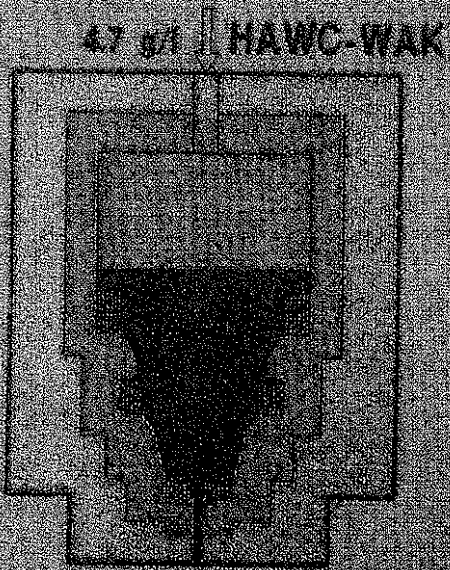
Noble Metal Discharge
Efficiency (Σ Ru, Pd, Rh) : 30-40 %



Industrial-scale Melter K-W2

Discharge Efficiency

Ruthenium	: 66 %
Palladium	: 58 %
Rhodium	: 62 %



Test Melter K-6'

Discharge Efficiency

Ruthenium	: 92 %
Palladium	: 95-98 %

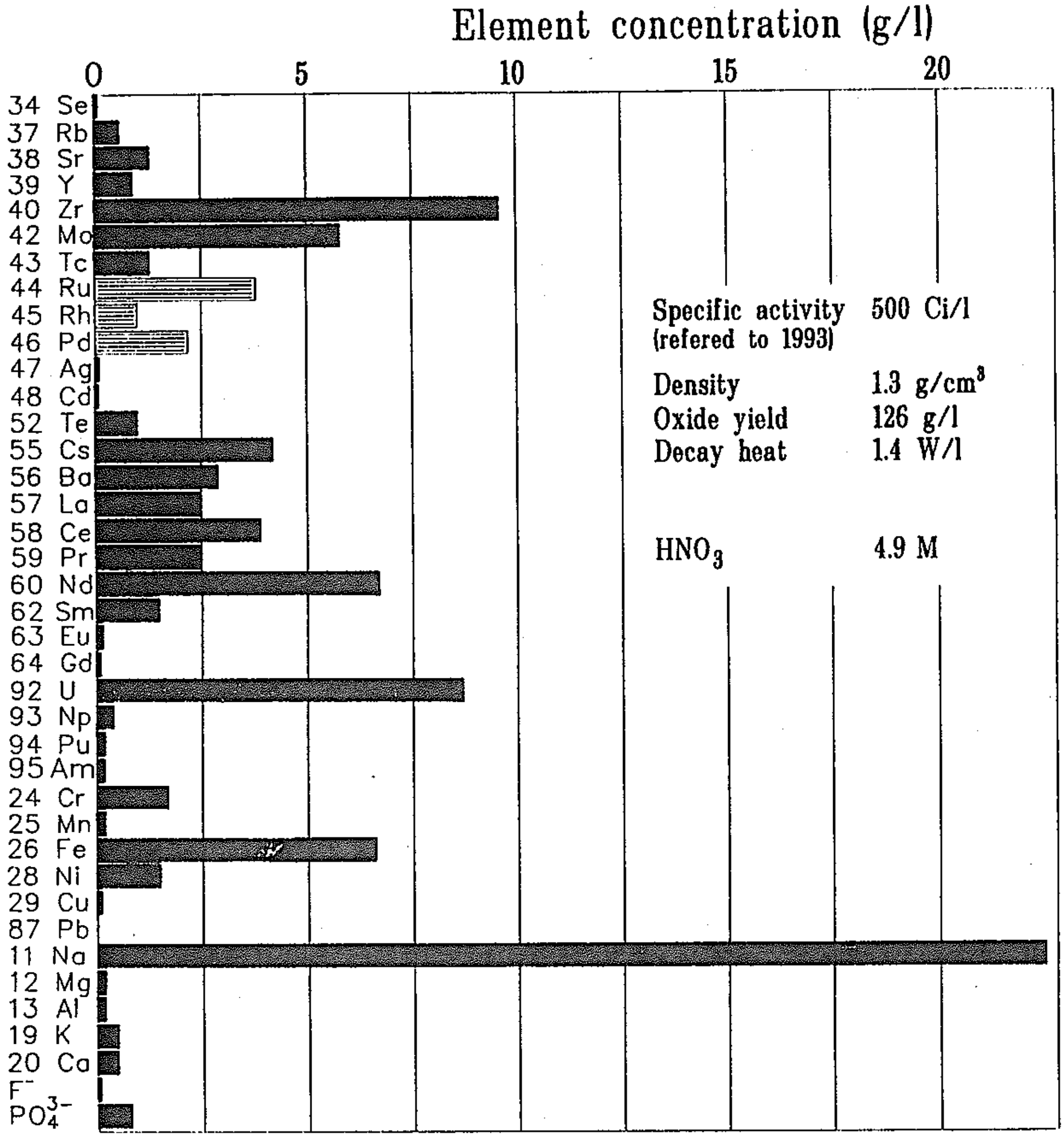
3-3 Performance of The Off-gas Treatment System

Performence of the Off-gas Treatment System

Prepared for the tenth annual PNC-KfK meeting on HLLW management

Technical Overview Session

Nov. 20, 1990



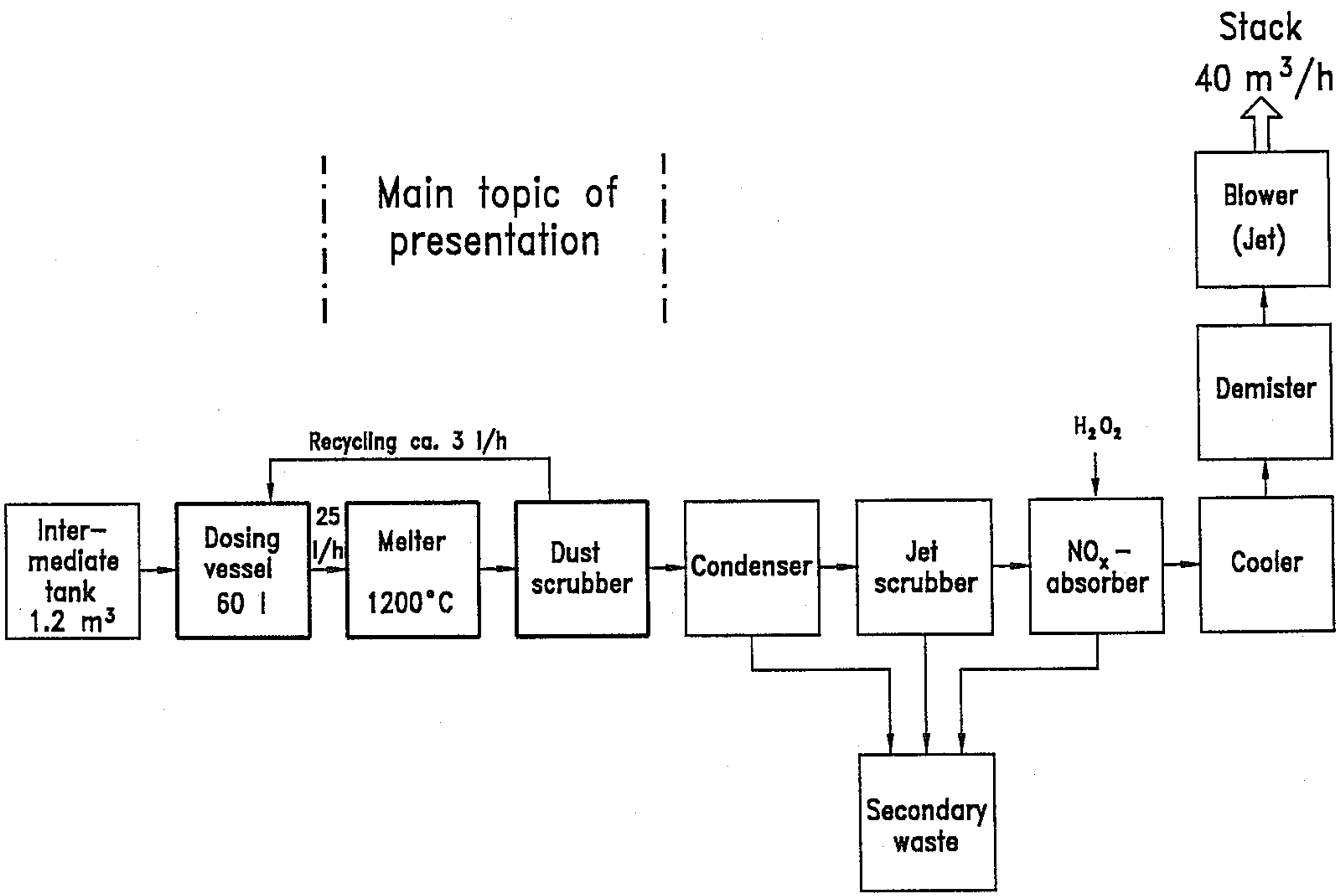
INE

Concentration of the different chemical elements in the WAK−HAWC currently stored in tank 210.02

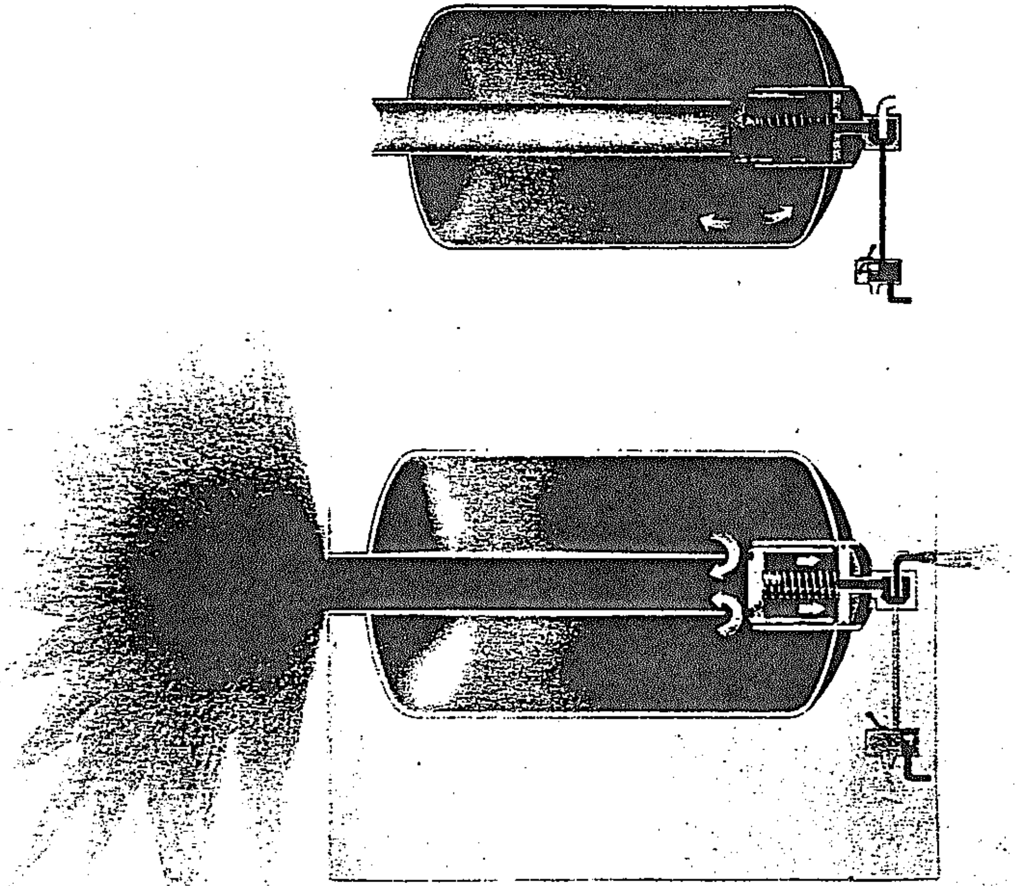
Chem. Zusammensetzung des HAWC-WAK Simulats (Kampagne D1/D2)

Element	g/l
Se	0.08
Rb	--[1]
Sr	0.87
Y	0.68
Zr	6.02
Mo	4.47
Ru	2.91[3]
Pd	1.75[3]
Te	0.78
Cs	2.91
Ba	2.23
La	1.94
Ce	3.01
Pr	1.94
Nd	5.29
Sm	1.17
Eu	--[2]
Gd	0.78
Cr	1.26
Mn	0.17
Fe	5.05
Ni	1.17
Cu	0.78
Zn	0.10
Pb	0.10
Na	15.53
Mg	0.19
Al	0.19
K	0.59
Ca	0.39
PO_4^{3-}	0.58
HNO_3	5.30 Mol/l

1) 0.46 g/l Rb ersetzt durch 0.21 g/l K
2) 0.16 g/l Eu ersetzt durch 0.15 g/l Nd
3) Kampagne D1 ohne Ru, Pd



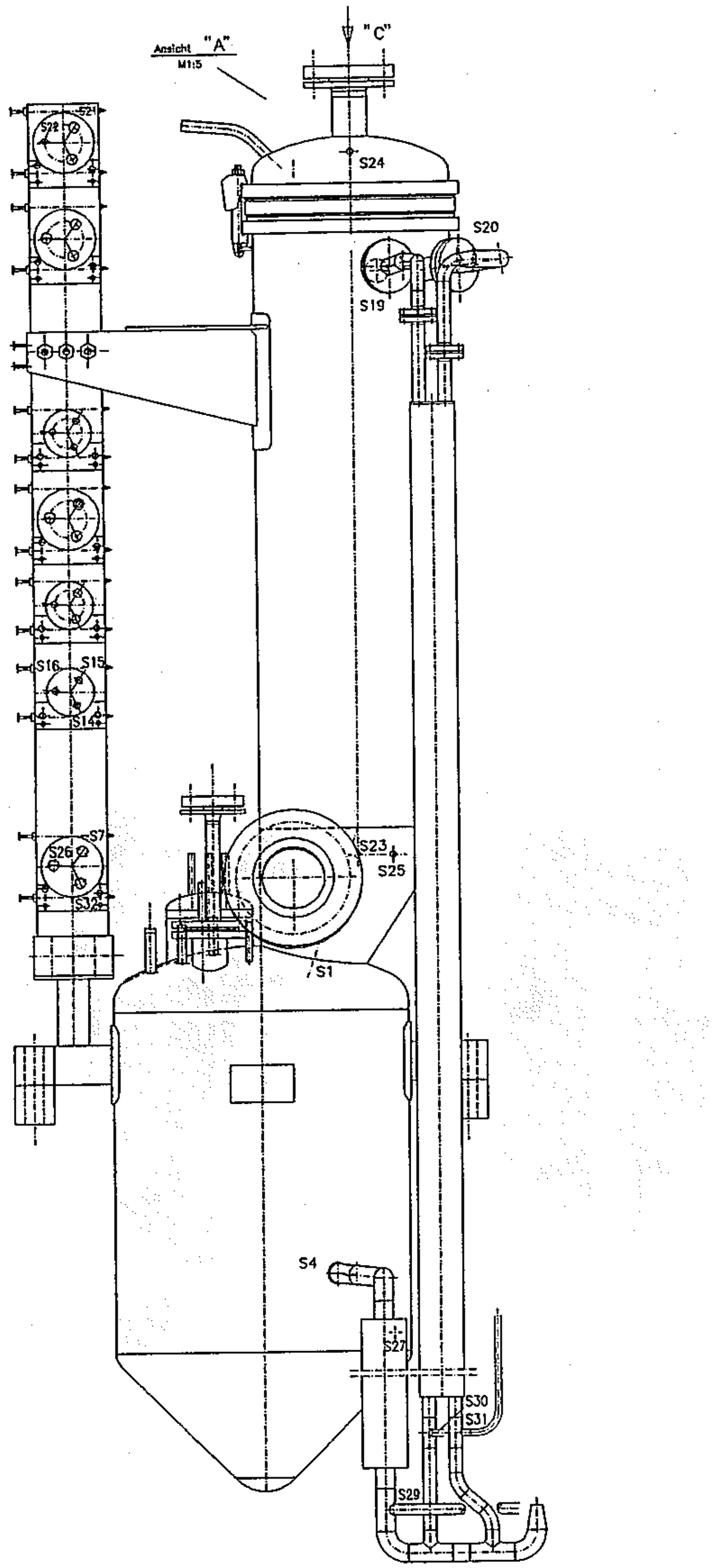
Off-gas system of the VA-WAK facility

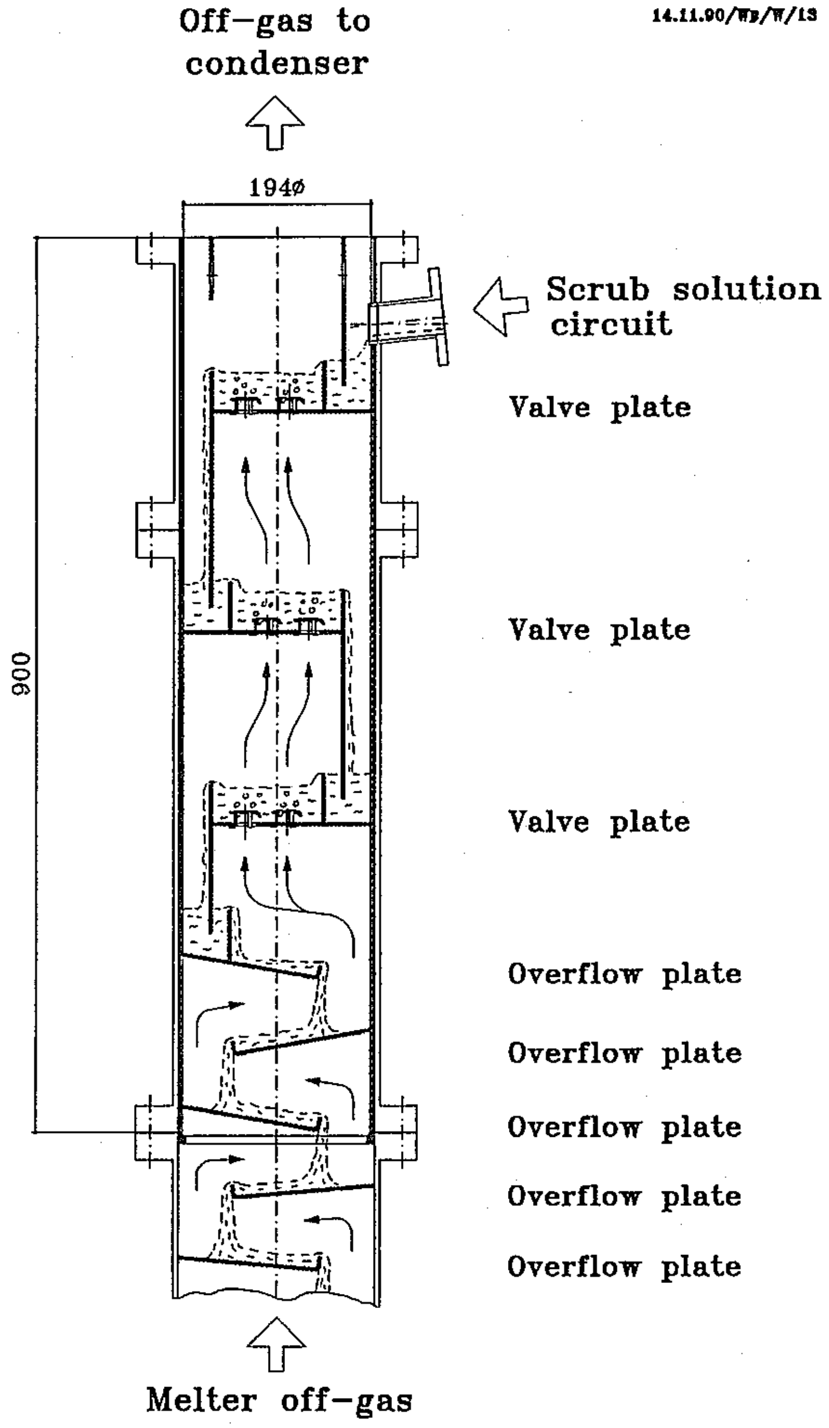


Technical specifications

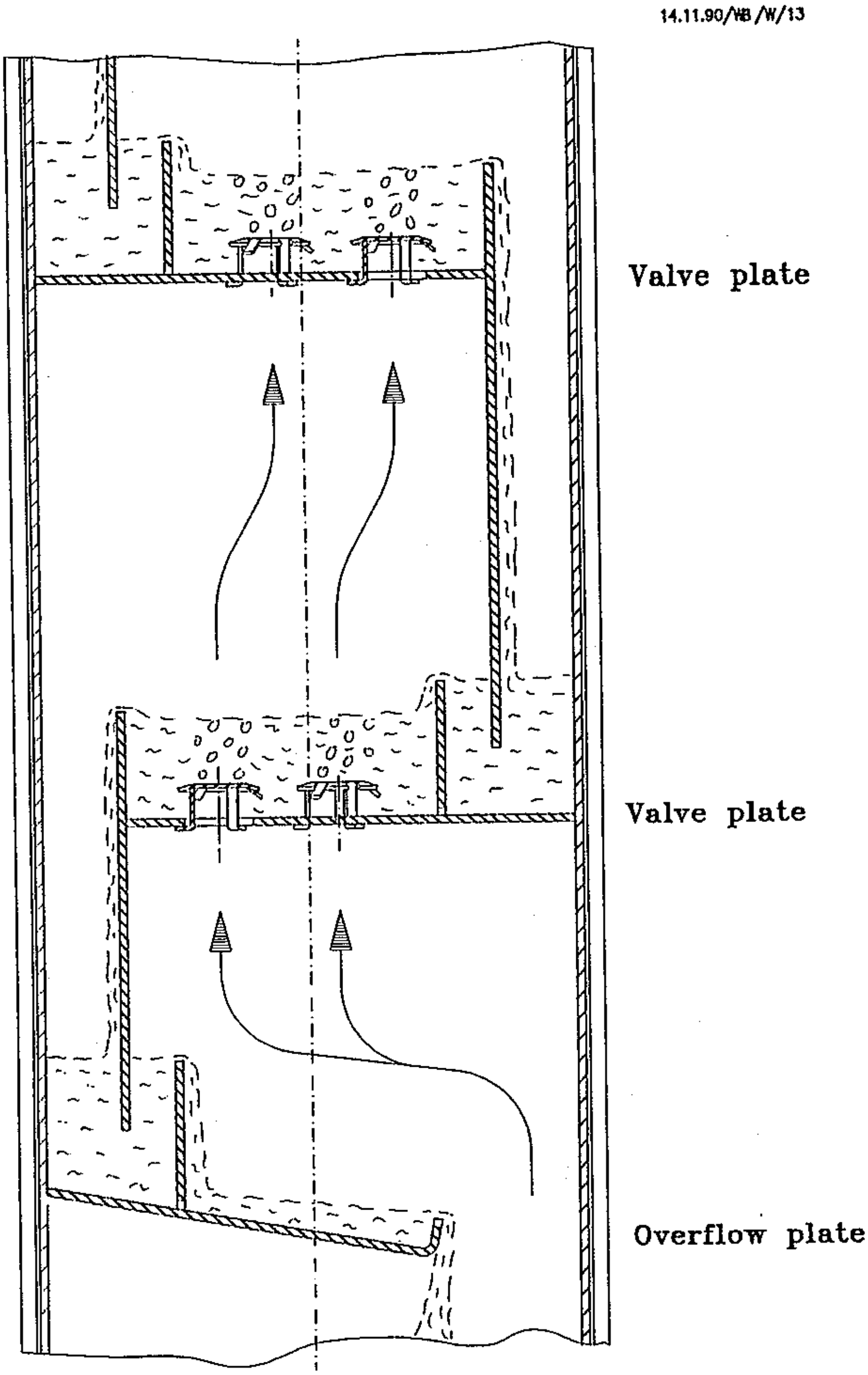
Dimensions	405/206 mm
Weight	14 kg
Volumne capacity	12 l
Air pressure	6—10 bar
Discharge time	< 15 ms

V—W1 Off—Gas treatment: Air cannon

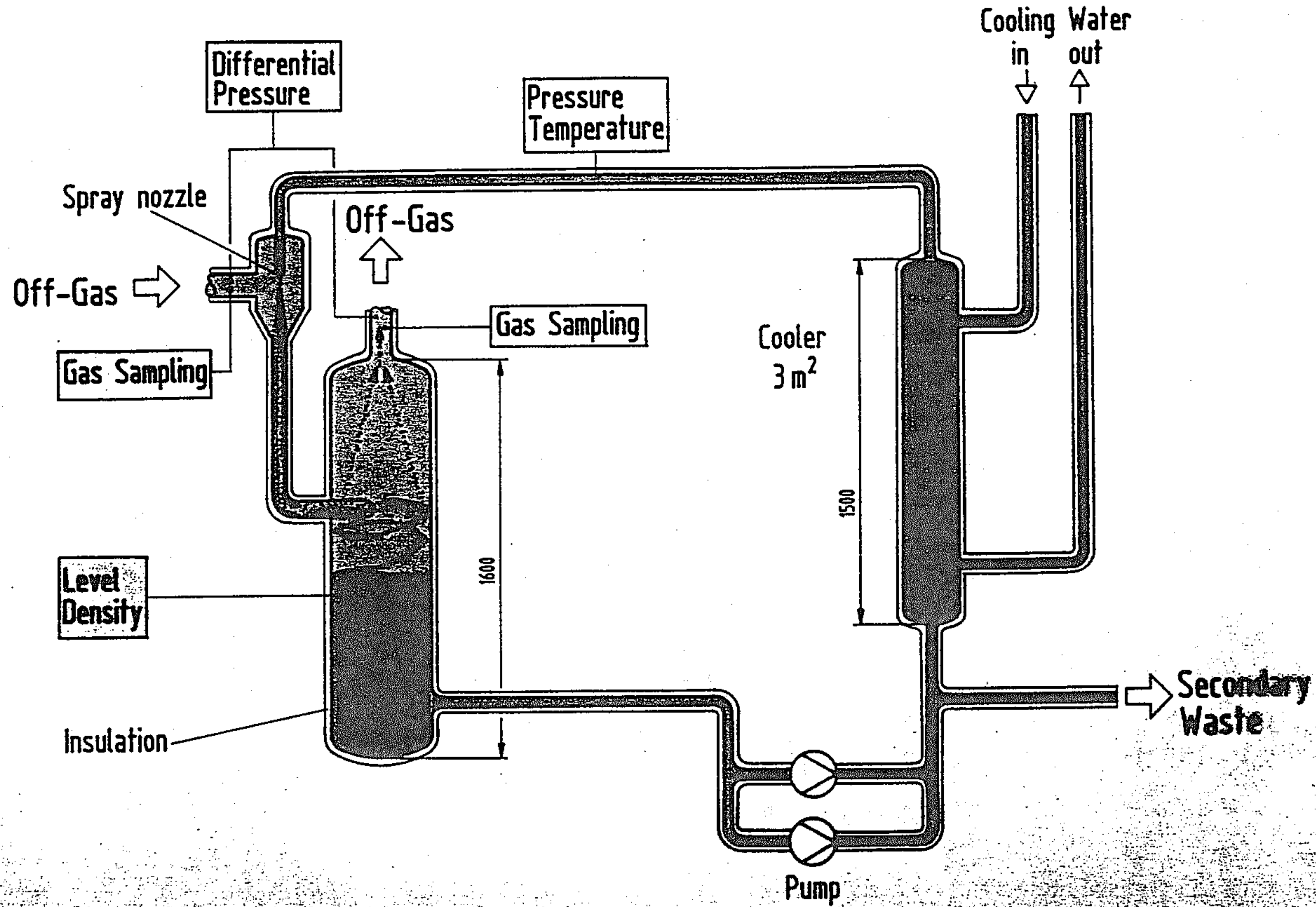

Ansicht "A"
M1:5
"C"
S21
S22
S24
S20
S19
S16
S15
S14
S7
S26
S32
S23
S25
S1
S4
S27
S30
S31
S29



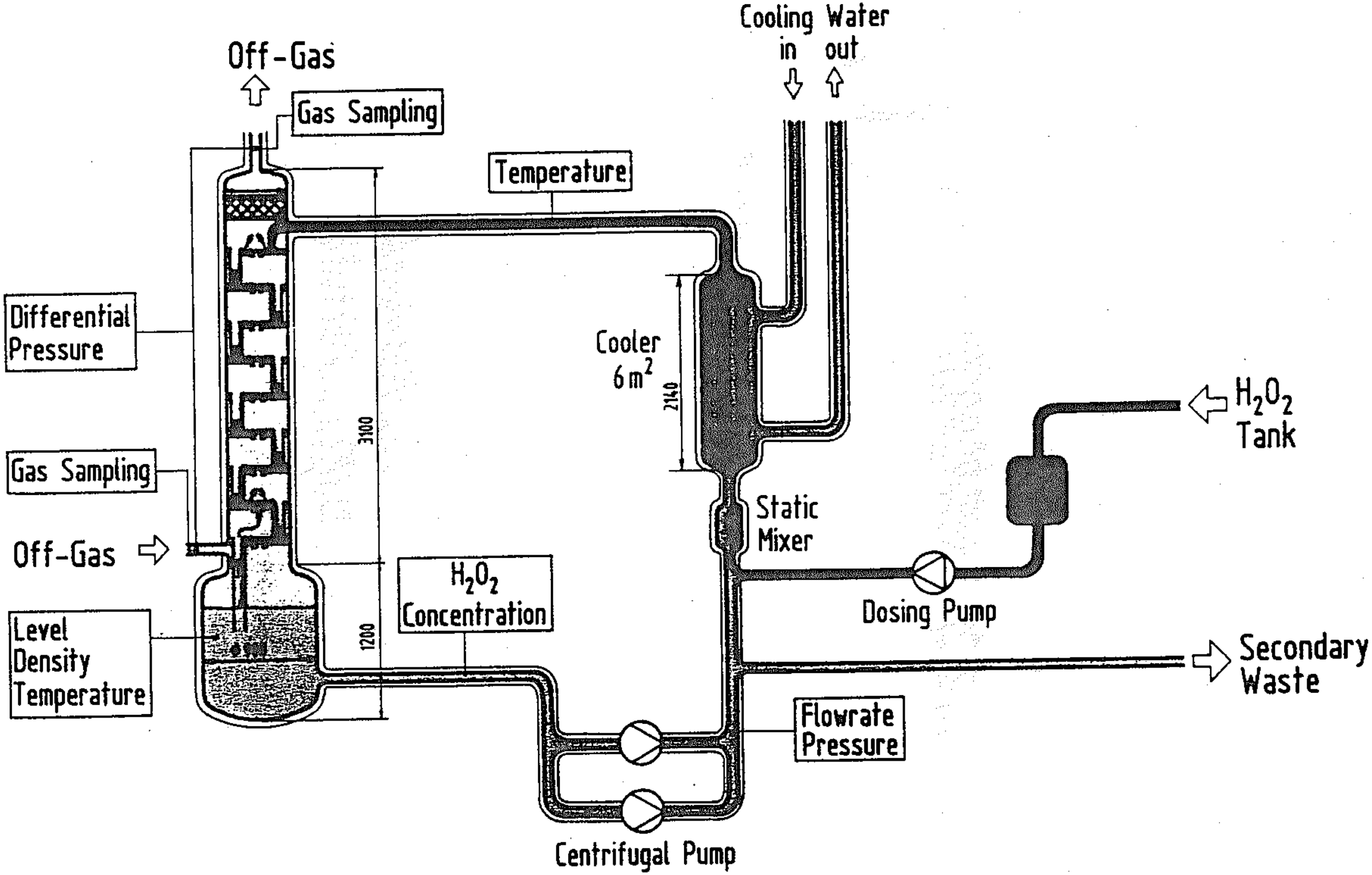
VA−WAK: Optimized dust scrubber column



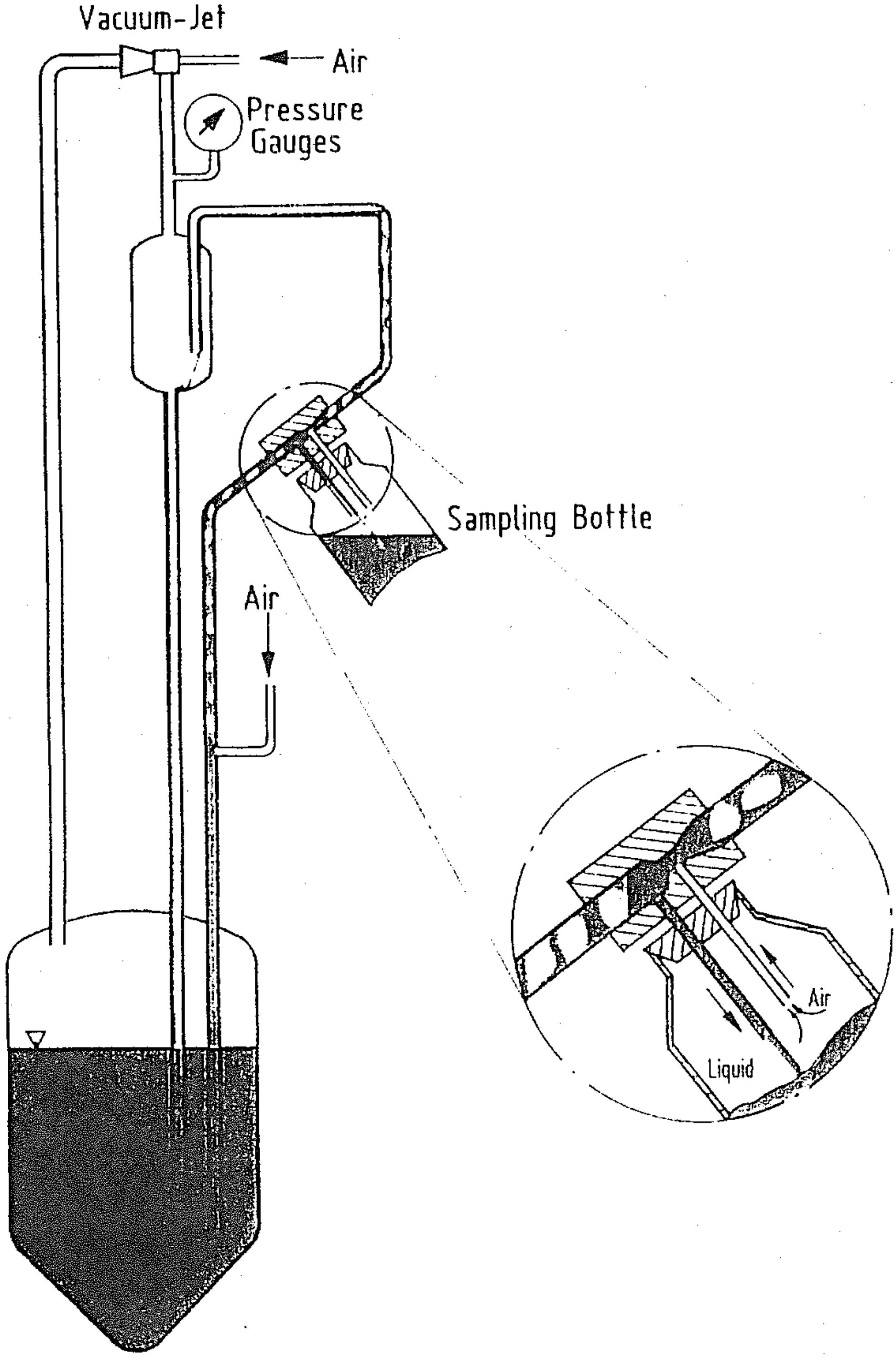
Details of valve plates



V-W1 Off-Gas Treatment: Jet Scrubber



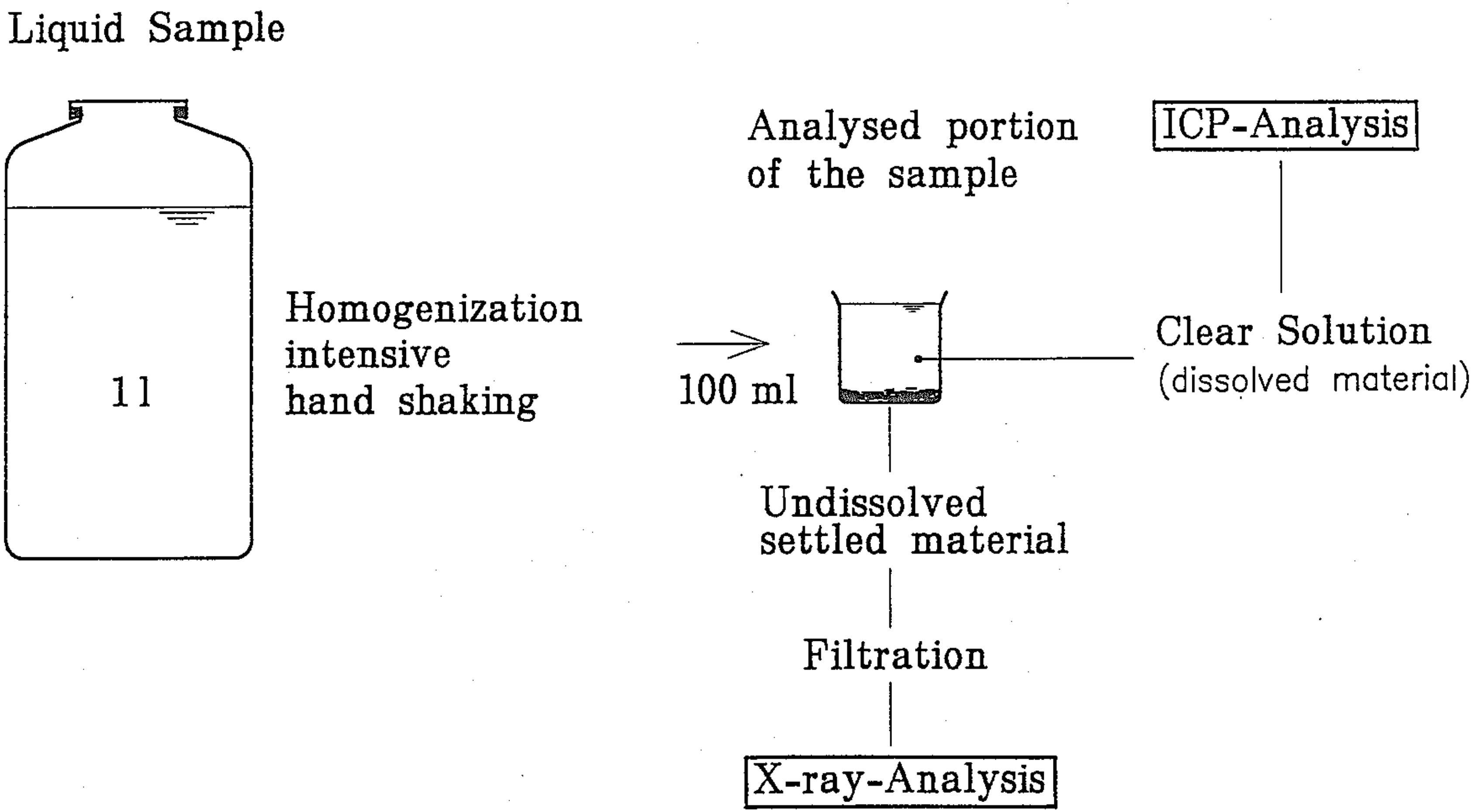
V-W1 Off-Gas Treatment: NO_x - Absorber I

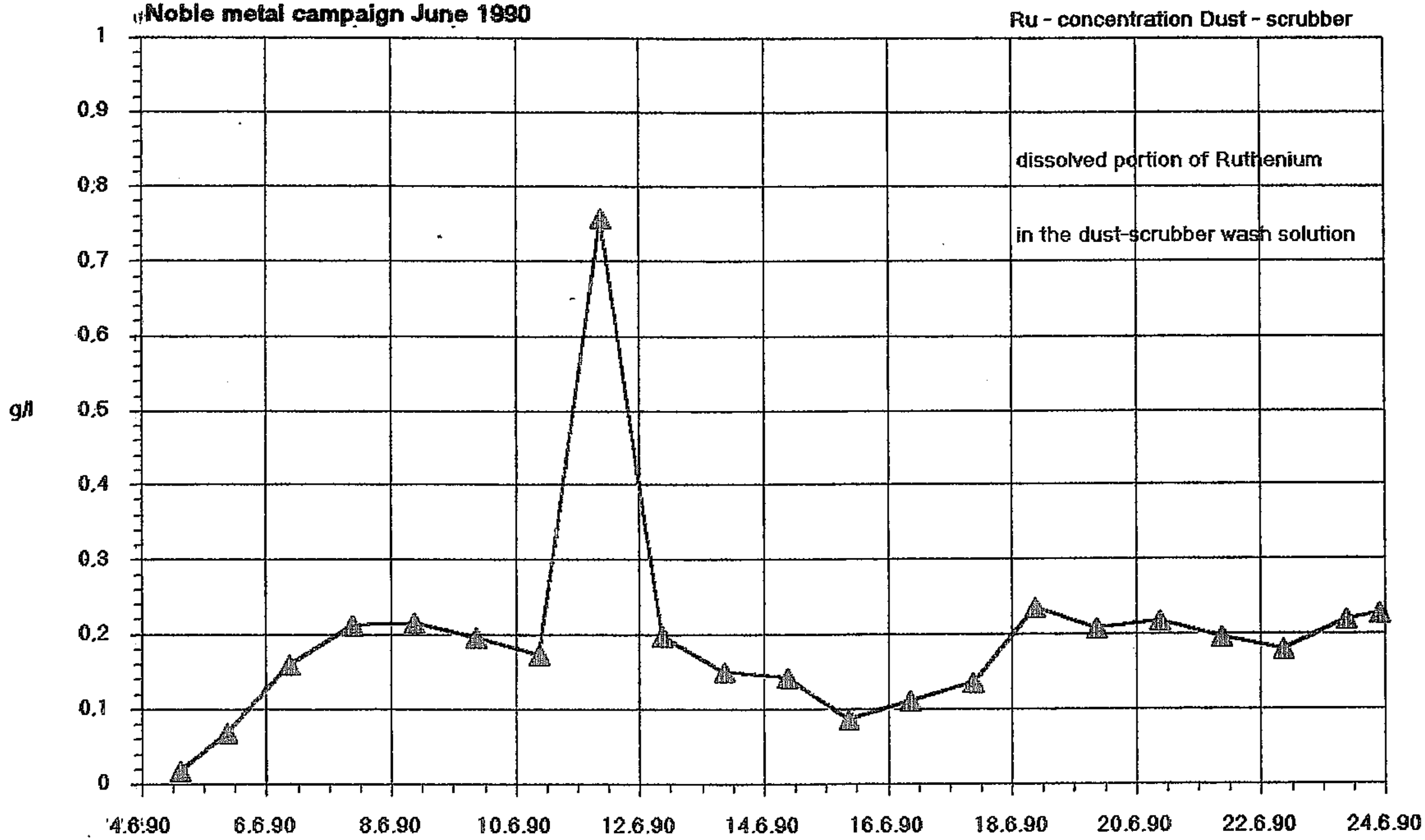

Vacuum-Jet
Air
Pressure
Gauges
Sampling Bottle
Air
Air
Liquid

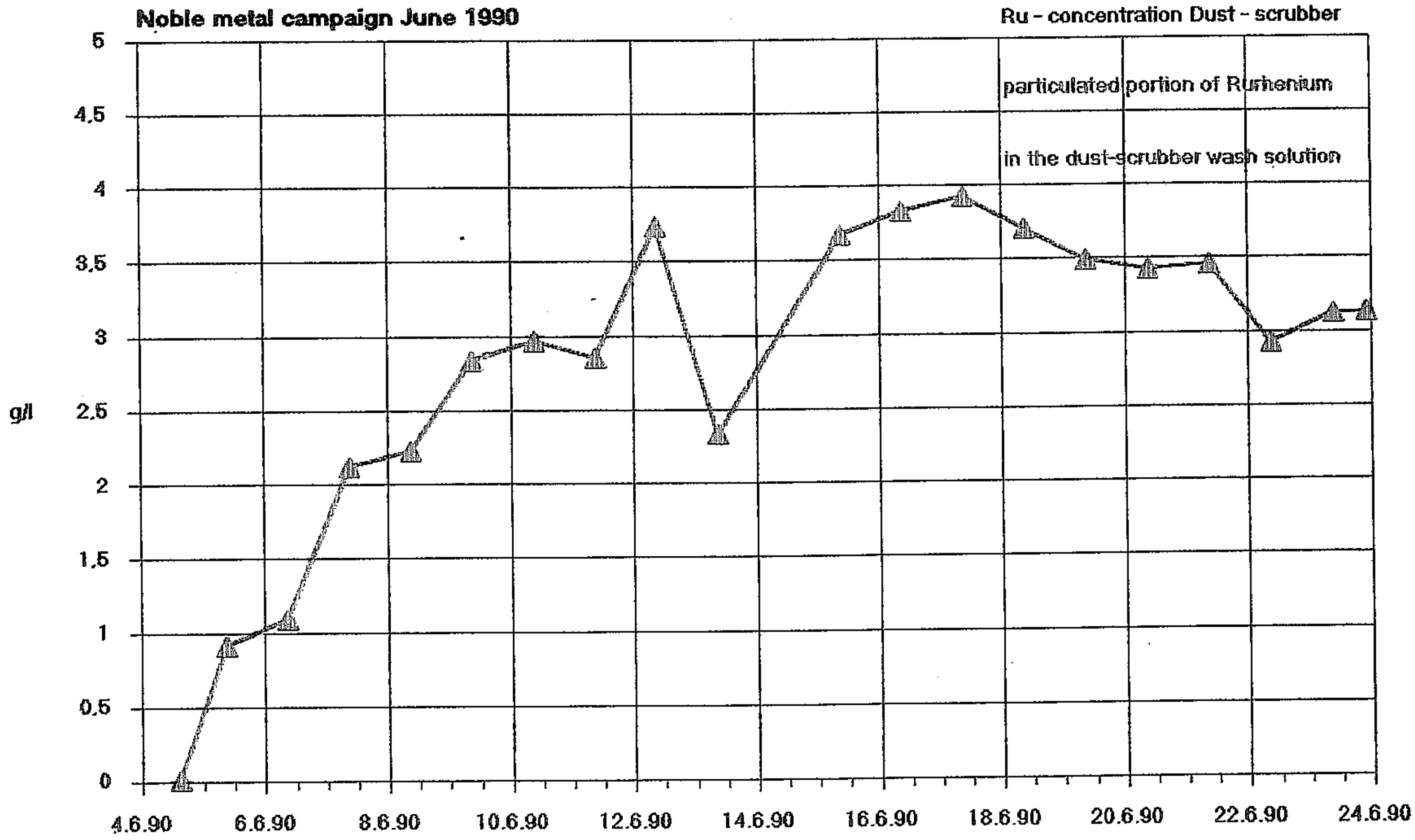
SAMPLING

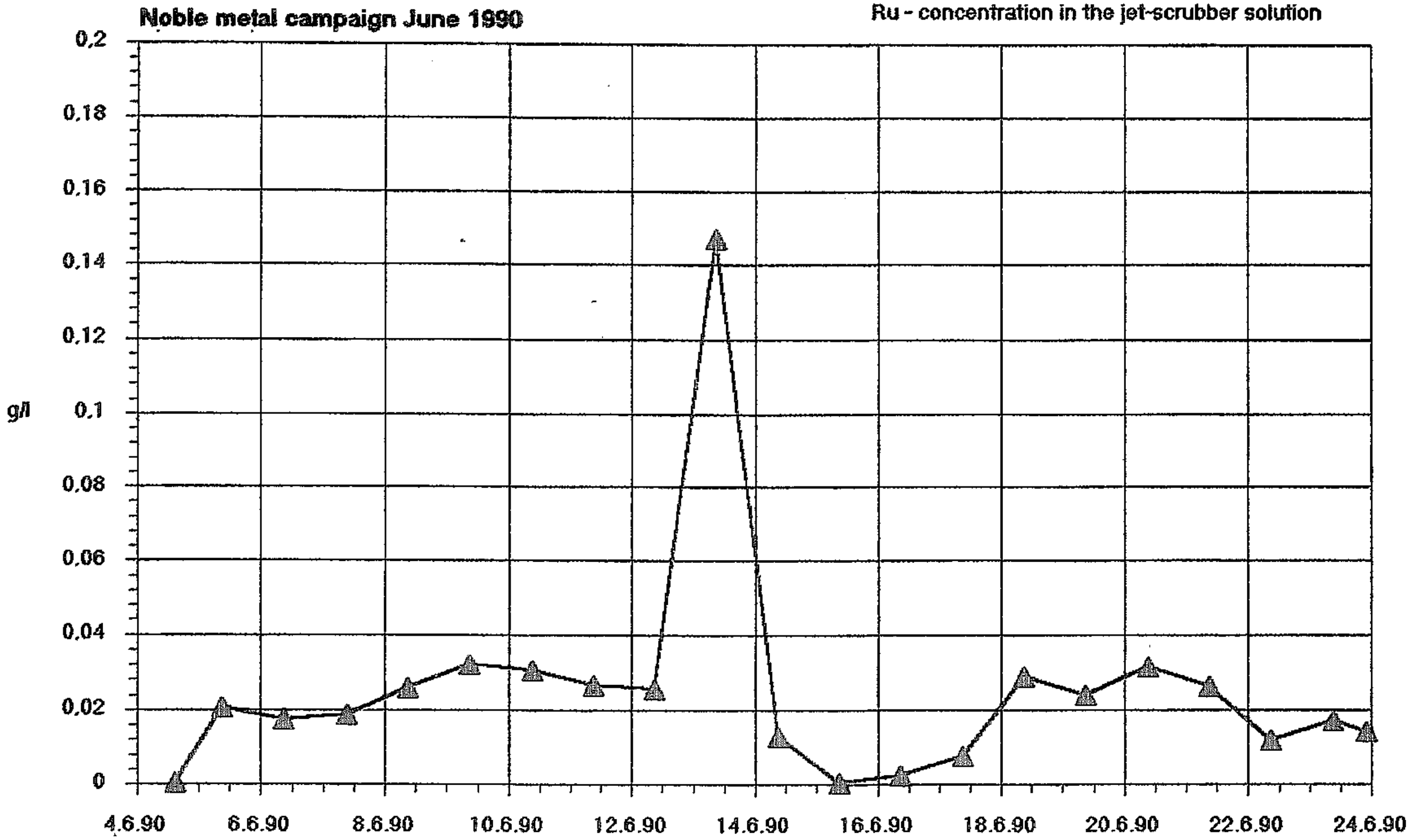
During Campaign	
Sample volume	1 l
Sampling time	- Every 24 hours of Operation - or before complete change of the scrub solution of the individual off-gas component
After Campaign	Final sampling from each off-gas component
Intermediate Objective of sample analysis	To obtain the concentration versus time curve in the solutions for individual elements like Ru, Cs, Sr, Se etc.
Final Objective of sample analysis	- To calculate the DF's for individual elements - To evaluate the off-gas system performence

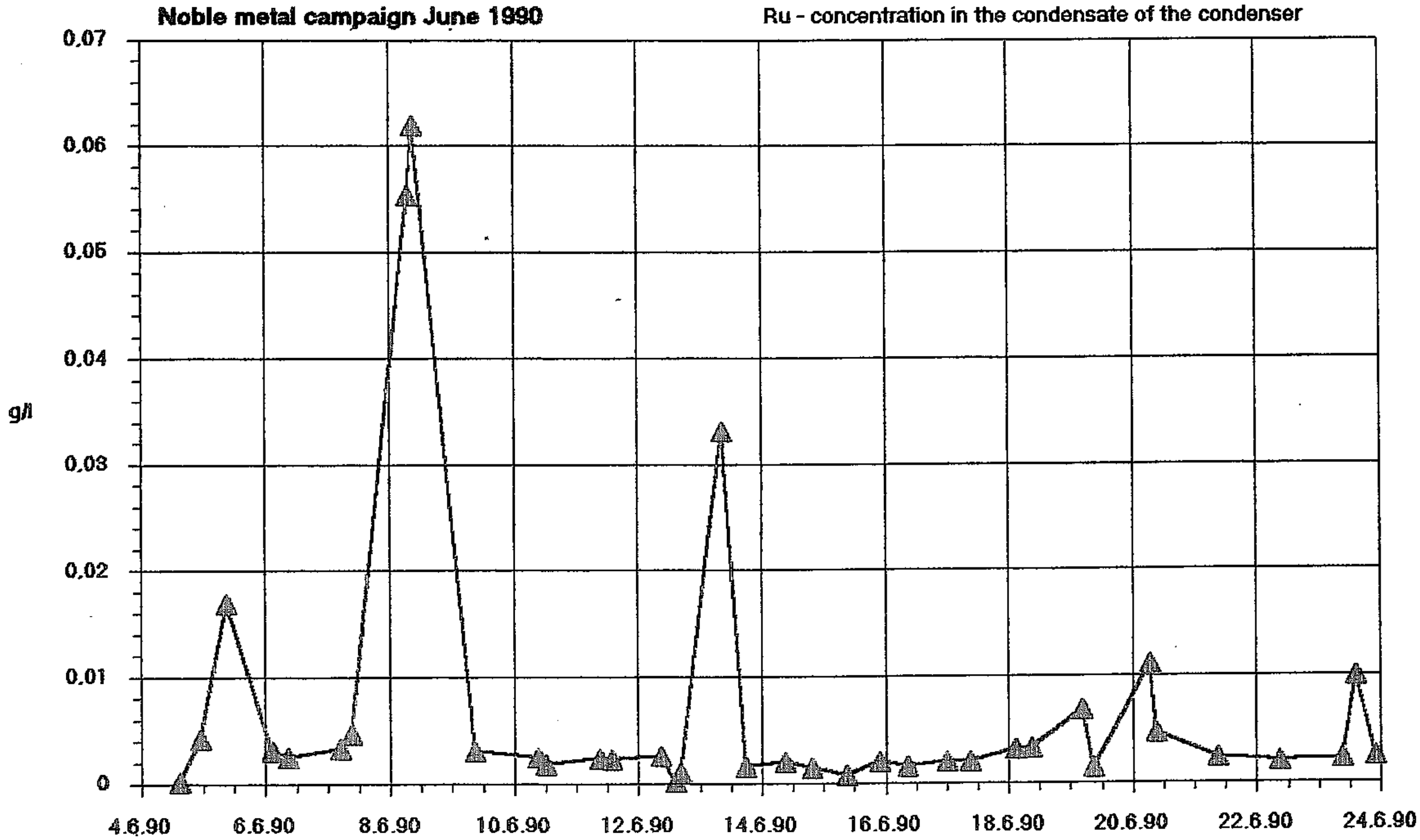
Analytical Procedure

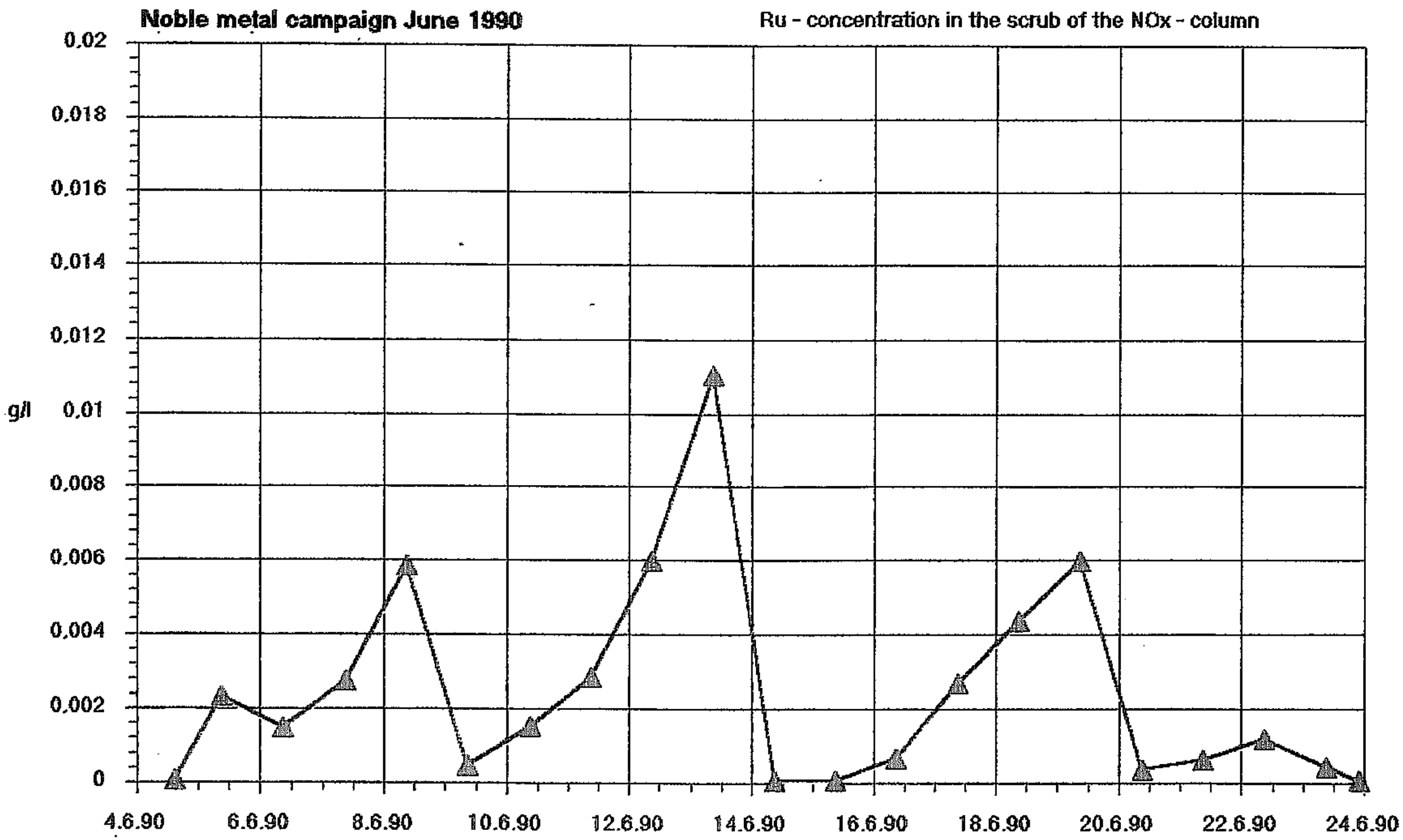


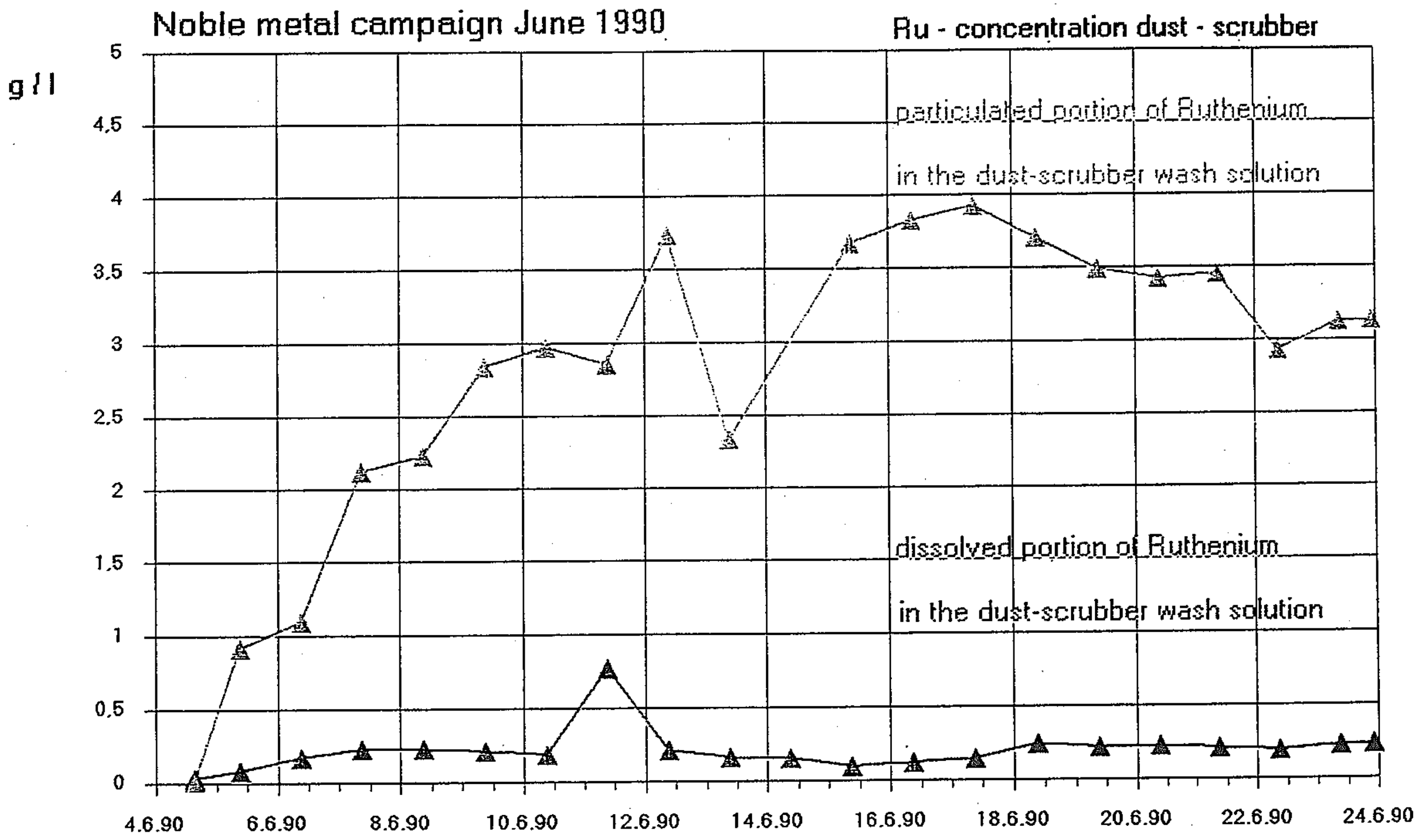

Noble metal campaign June 1990
Ru - concentration Dust - scrubber
dissolved portion of Ruthenium
in the dust-scrubber wash solution
g/l
1
0,9
0,8
0,7
0,6
0,5
0,4
0,3
0,2
0,1
0
4.6.90
6.6.90
8.6.90
10.6.90
12.6.90
14.6.90
16.6.90
18.6.90
20.6.90
22.6.90
24.6.90

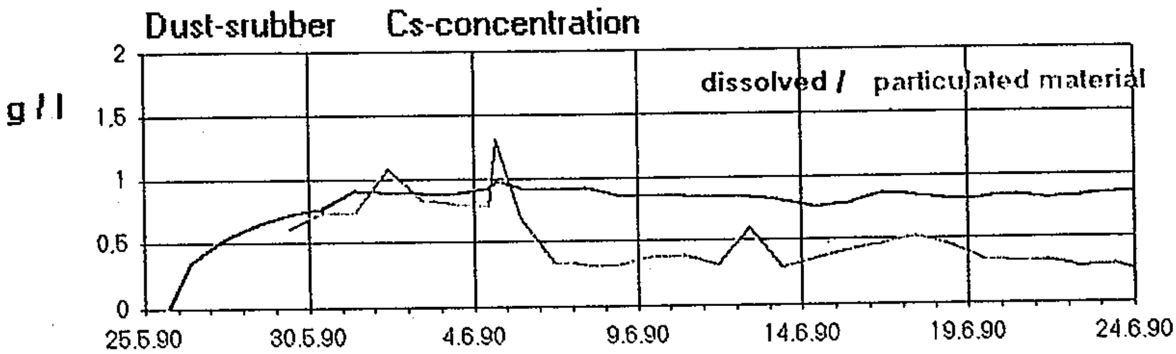

Noble metal campaign June 1990
Ru - concentration Dust - scrubber
particulated portion of Ruthenium
in the dust-scrubber wash solution
g/l
0
0,5
1
1,5
2
2,5
3
3,5
4
4,5
5
4.6.90
6.6.90
8.6.90
10.6.90
12.6.90
14.6.90
16.6.90
18.6.90
20.6.90
22.6.90
24.6.90

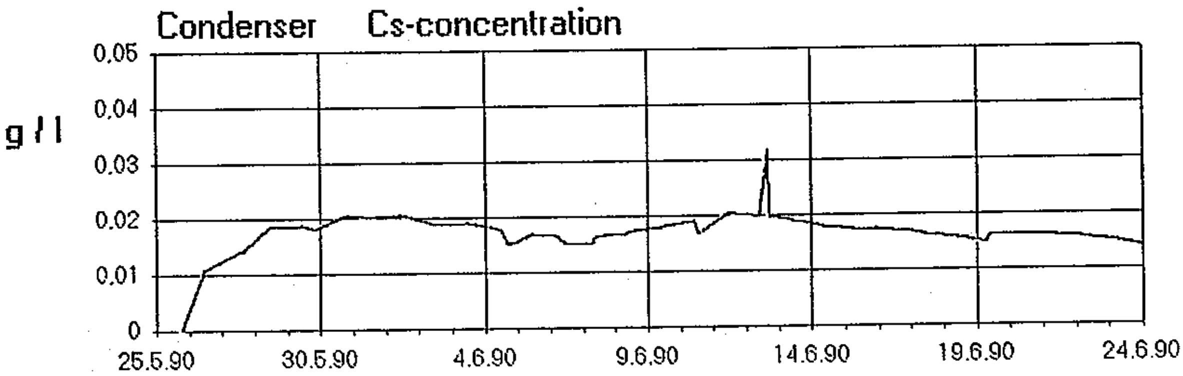

Noble metal campaign June 1990
Ru - concentration in the jet-scrubber solution
g/l
0,2
0,18
0,16
0,14
0,12
0,1
0,08
0,06
0,04
0,02
0
4.6.90
6.6.90
8.6.90
10.6.90
12.6.90
14.6.90
16.6.90
18.6.90
20.6.90
22.6.90
24.6.90

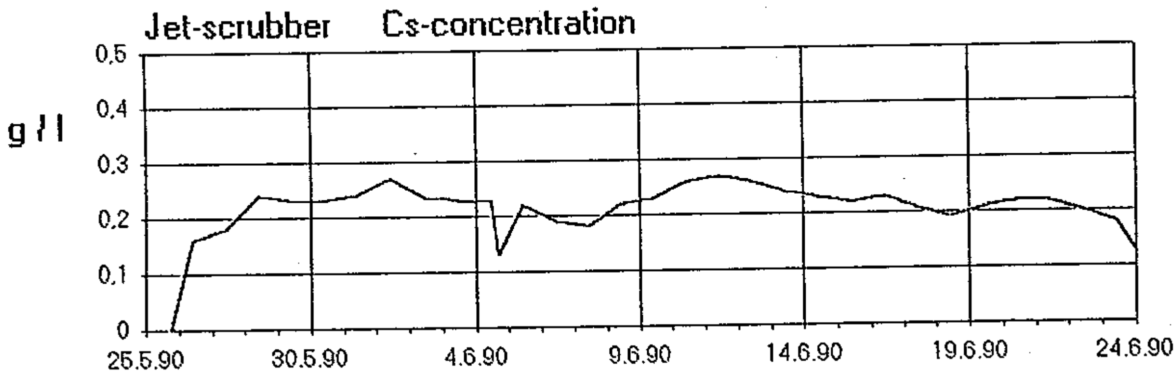

Noble metal campaign June 1990
Ru - concentration in the condensate of the condenser
g/l
0.07
0.06
0.05
0.04
0.03
0.02
0.01
0
4.6.90
6.6.90
8.6.90
10.6.90
12.6.90
14.6.90
16.6.90
18.6.90
20.6.90
22.6.90
24.6.90

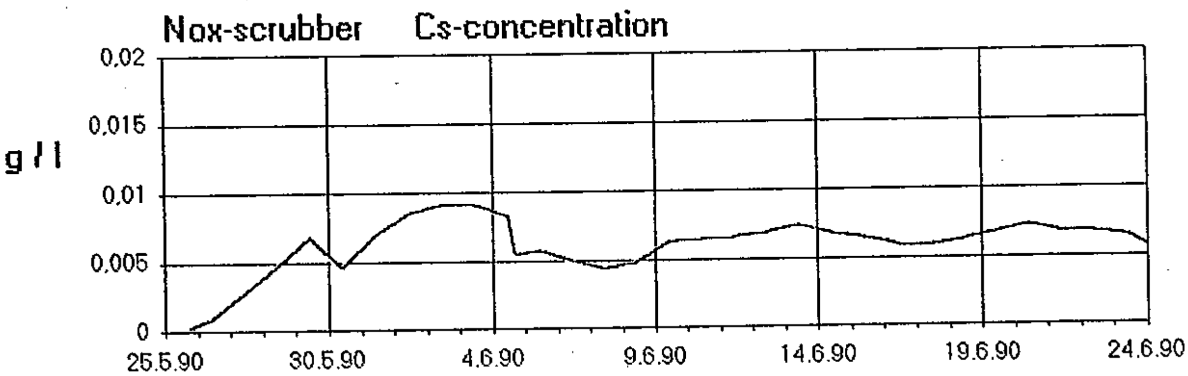

Noble metal campaign June 1990
Ru - concentration in the scrub of the NOx - column
g/l
0,02
0,018
0,016
0,014
0,012
0,01
0,008
0,006
0,004
0,002
0
4.6.90
6.6.90
8.6.90
10.6.90
12.6.90
14.6.90
16.6.90
18.6.90
20.6.90
22.6.90
24.6.90

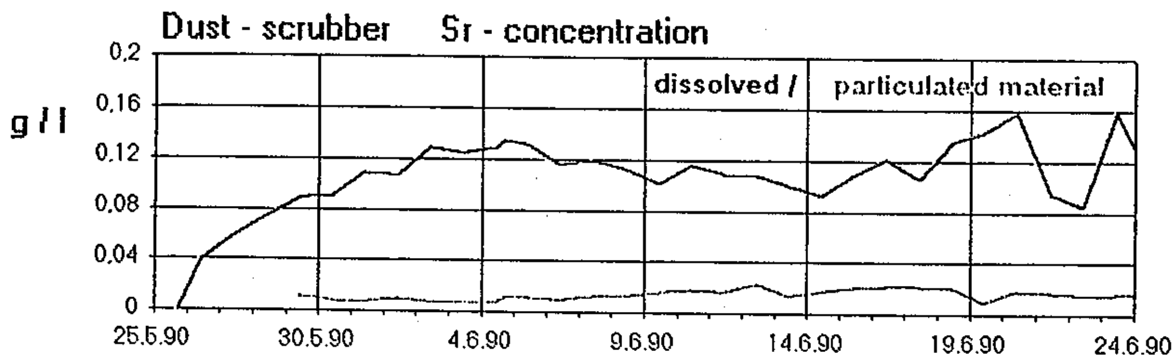

Noble metal campaign June 1990
Ru - concentration dust - scrubber
g/l
5
4,5
4
3,5
3
2,5
2
1,5
1
0,5
0
particulated portion of Ruthenium
in the dust-scrubber wash solution
dissolved portion of Ruthenium
in the dust-scrubber wash solution
4.6.90
6.6.90
8.6.90
10.6.90
12.6.90
14.6.90
16.6.90
18.6.90
20.6.90
22.6.90
24.6.90

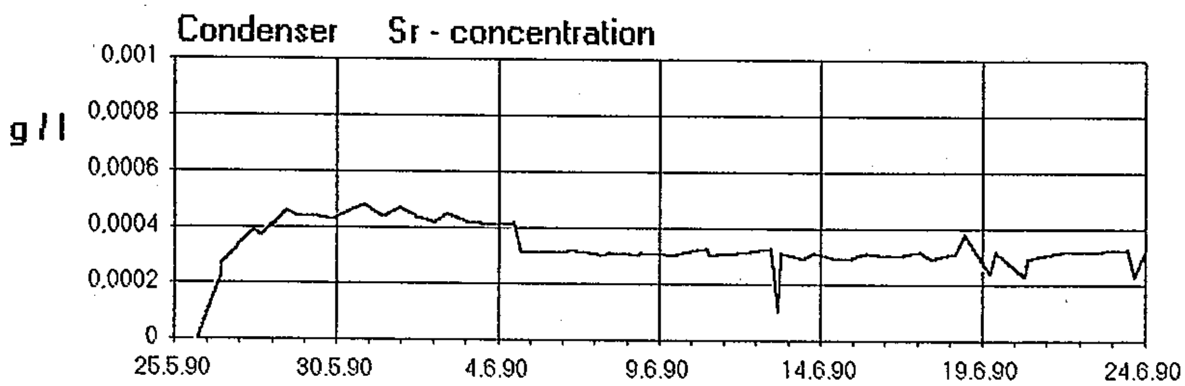

Dust-srubber Cs-concentration
dissolved / particulated material
g / l
2
1.5
1
0.5
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

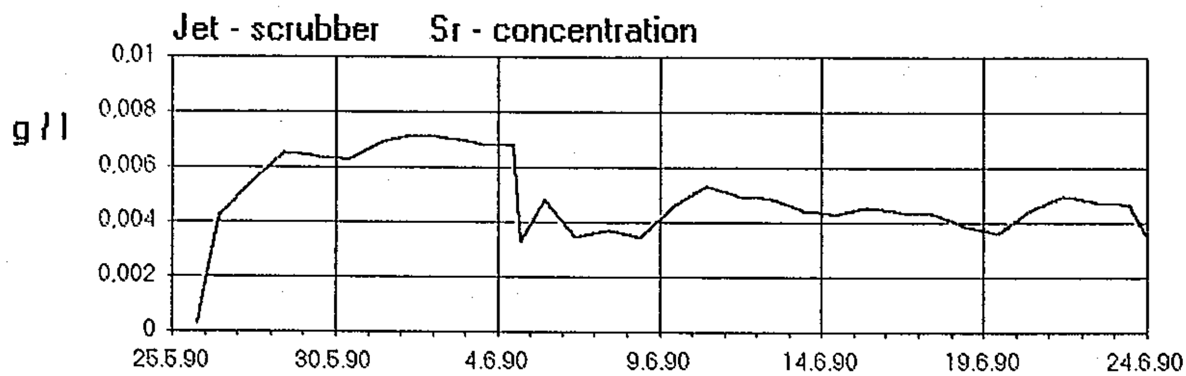

Condenser Cs-concentration
g / l
0.05
0.04
0.03
0.02
0.01
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

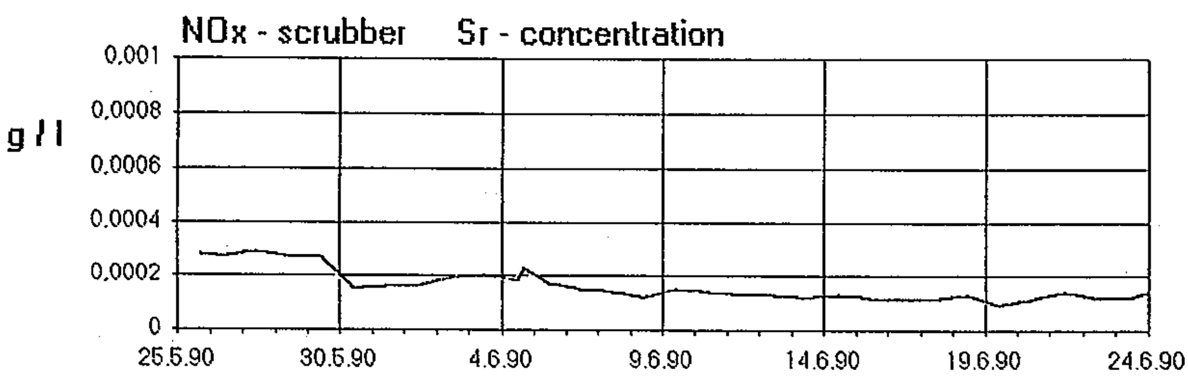

Jet-scrubber Cs-concentration
g / l
0.5
0.4
0.3
0.2
0.1
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

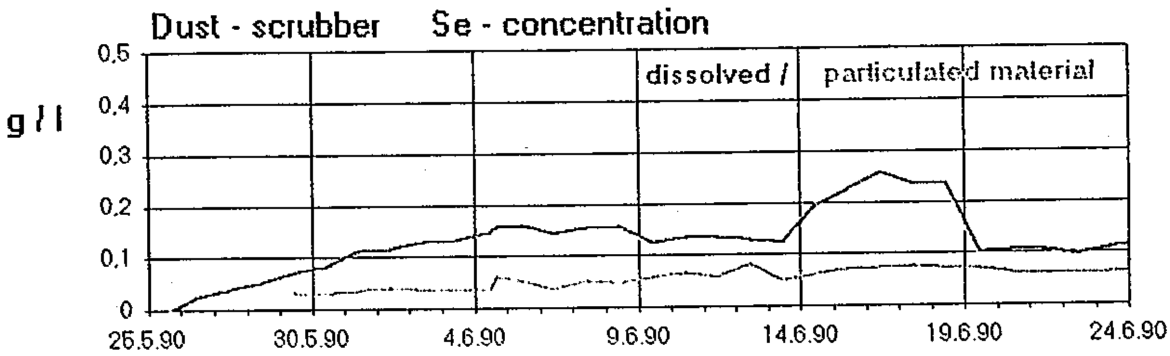

Nox-scrubber Cs-concentration
g / l
0.02
0.015
0.01
0.005
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

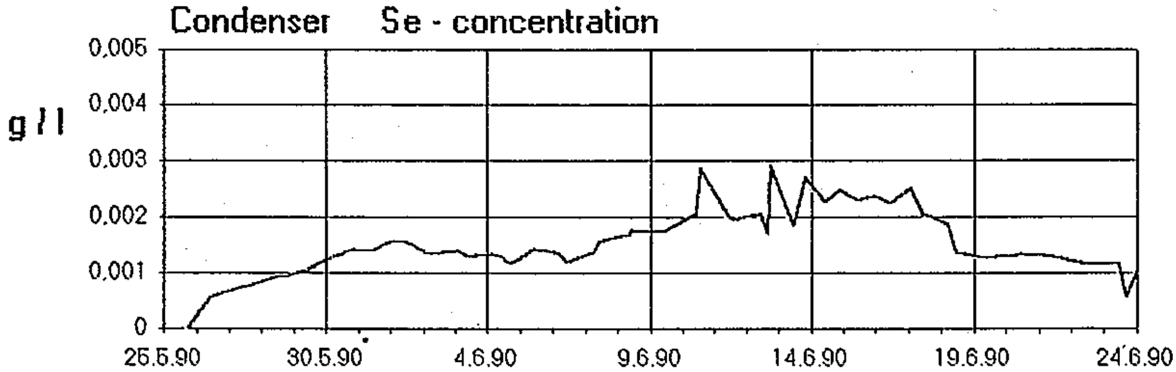

Dust - scrubber Sr - concentration
g / l
0.2
0.16
0.12
0.08
0.04
0
dissolved / particulated material
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

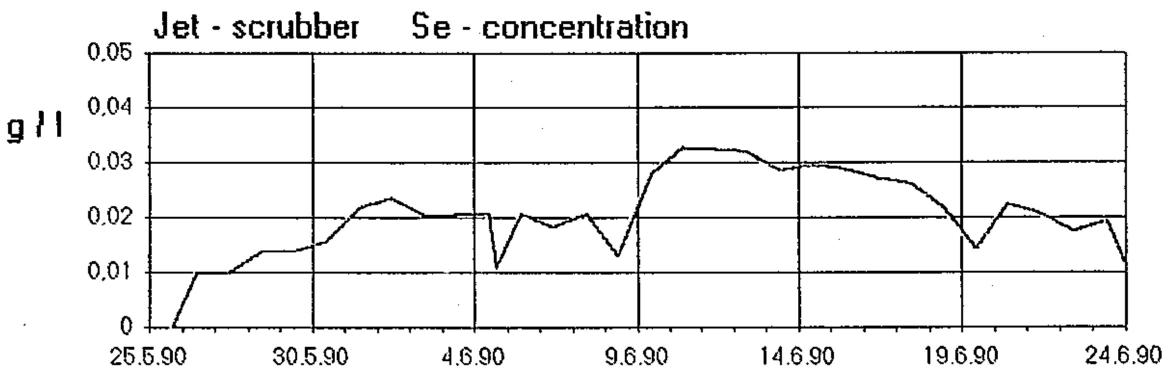

Condenser Sr - concentration
g / l
0.001
0.0008
0.0006
0.0004
0.0002
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

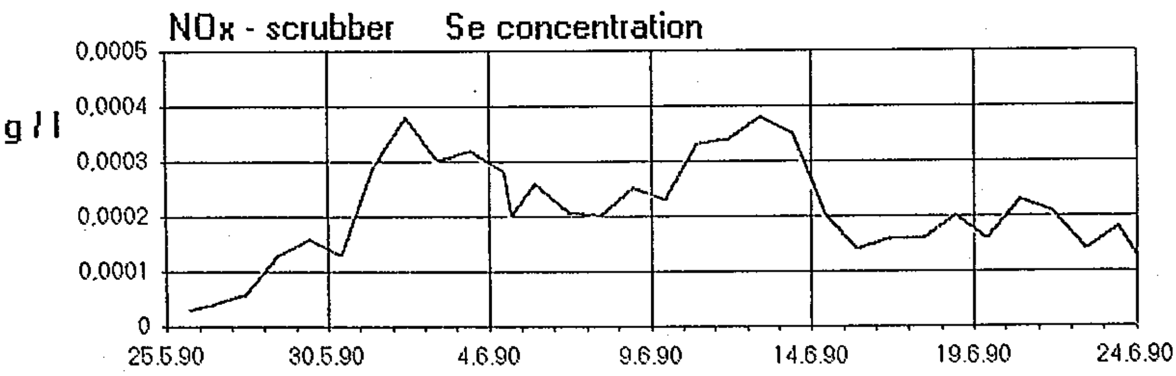

Jet - scrubber Sr - concentration
g / l
0.01
0.008
0.006
0.004
0.002
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

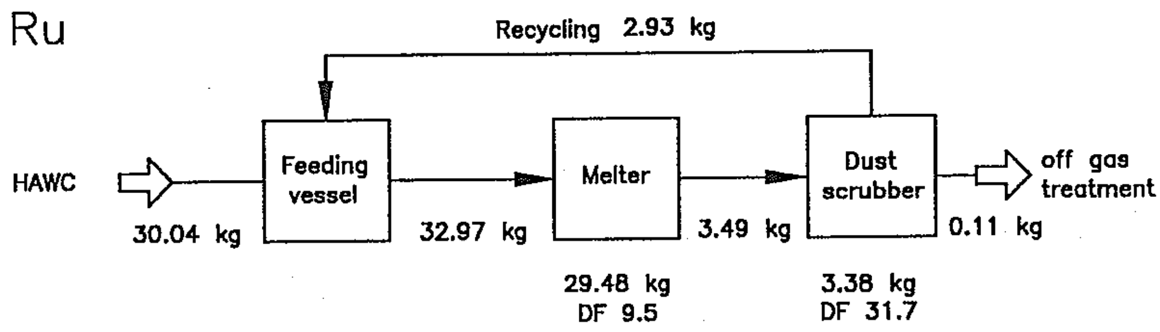

NOx - scrubber Sr - concentration
g / l
0.001
0.0008
0.0006
0.0004
0.0002
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

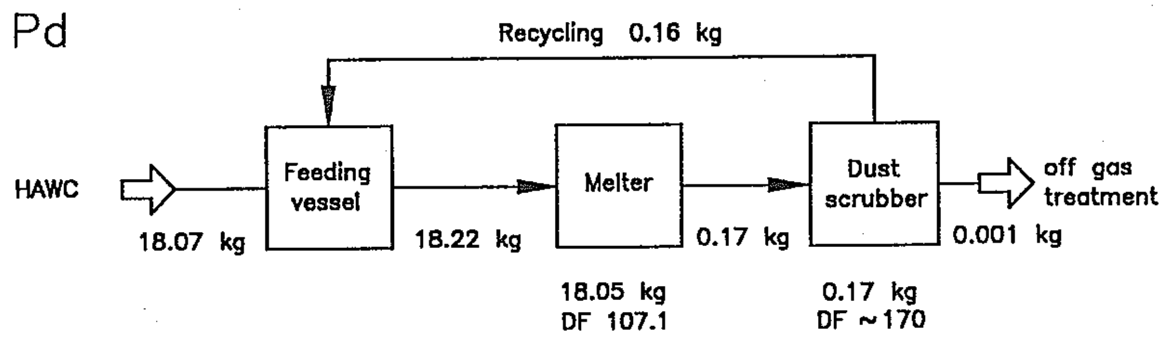

Dust - scrubber Se - concentration
dissolved / particulated material
g/l
0.5
0.4
0.3
0.2
0.1
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

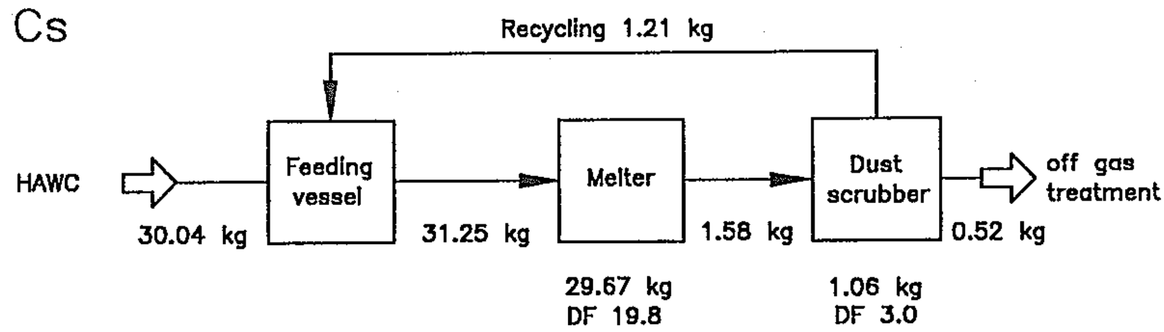

Condenser Se - concentration
g/l
0.005
0.004
0.003
0.002
0.001
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90


Jet - scrubber Se - concentration
g/l
0.05
0.04
0.03
0.02
0.01
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90


NOx - scrubber Se concentration
g/l
0.0005
0.0004
0.0003
0.0002
0.0001
0
25.5.90
30.5.90
4.6.90
9.6.90
14.6.90
19.6.90
24.6.90

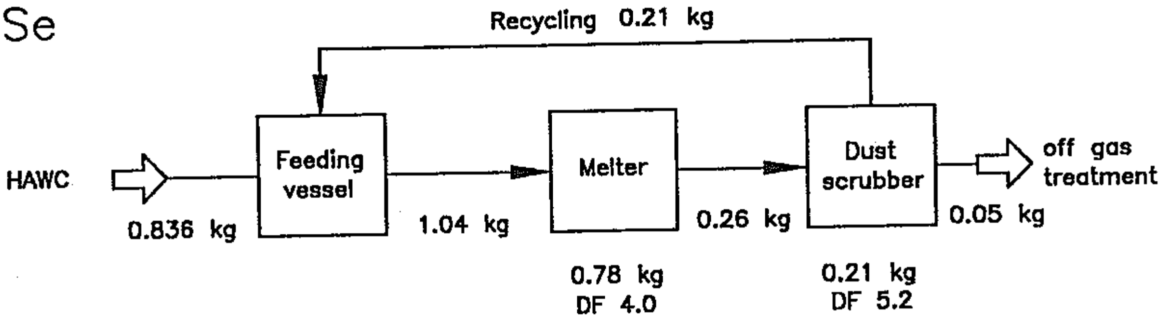




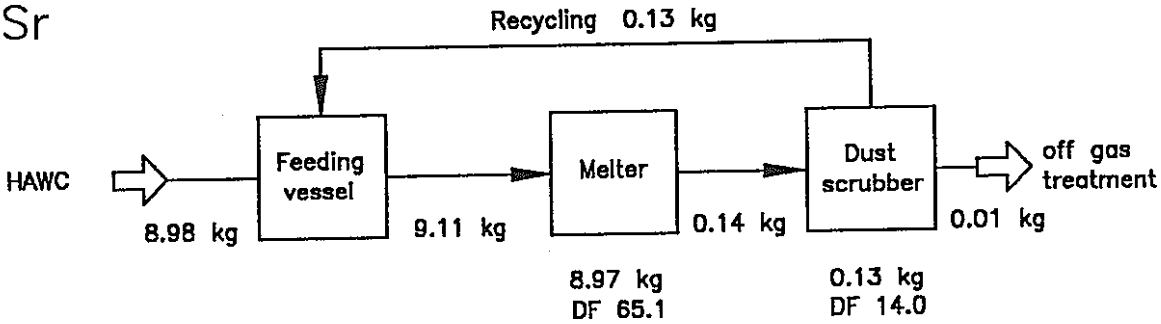


VA−WAK: Noble metal campaign D 2
June 1990

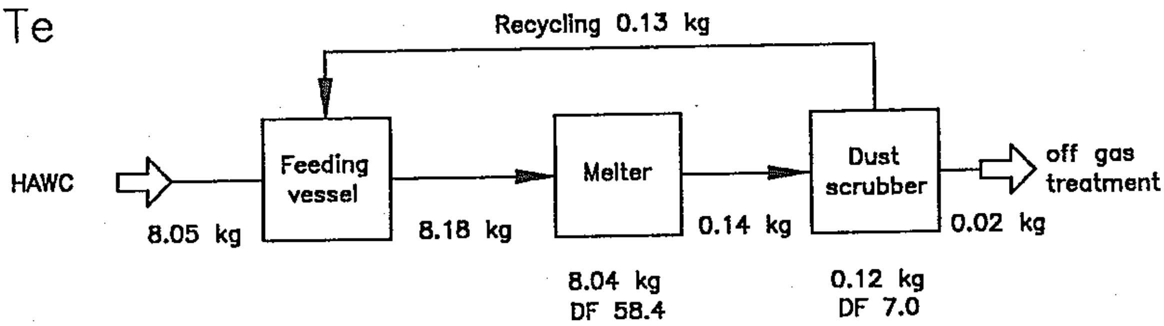
Se



Sr



Te



VA－WAK: Noble metal campaign D 2
June 1990

SUMMARY

- INE's procedure for liquid sampling from off-gas line components, and analysis by X-ray and ICP was outlined

- Concentration curves for Ru, Pd, Cs, Sr, Se in the secondary liquid waste has been presented, obtained from the noble metal campaign carried out in June 1990. In this campaign, simulated WAK-HAWC was used

- The major portion of Ru in the dust scrubber wash solution was undissolved (presumably RuO_2 particles)

- Melter DF for Ru was 9.5
  Dust scrubber DF for Ru was 31.7

- Melter DF for Cs was 19.8
  Dust scrubber DF for Cs was 3.0

- Optimized dust scrubber design was presented having 3 valve plates besides 5 bottom plates. The substantially improved DF's for Ru and other elements (except Cs) is mainly attributed to the use of three additional valve plates in the column

- The dust scrubber allows immediate recycling of removed elements, especially important for Ru, Cs and Sr, into the process

- The new dust scrubber design is especially more effective for the removal of Ru, compared to previous designs including that for the PAMELA dust scrubber

3-4 Physical and Mathematical Melter Modelling

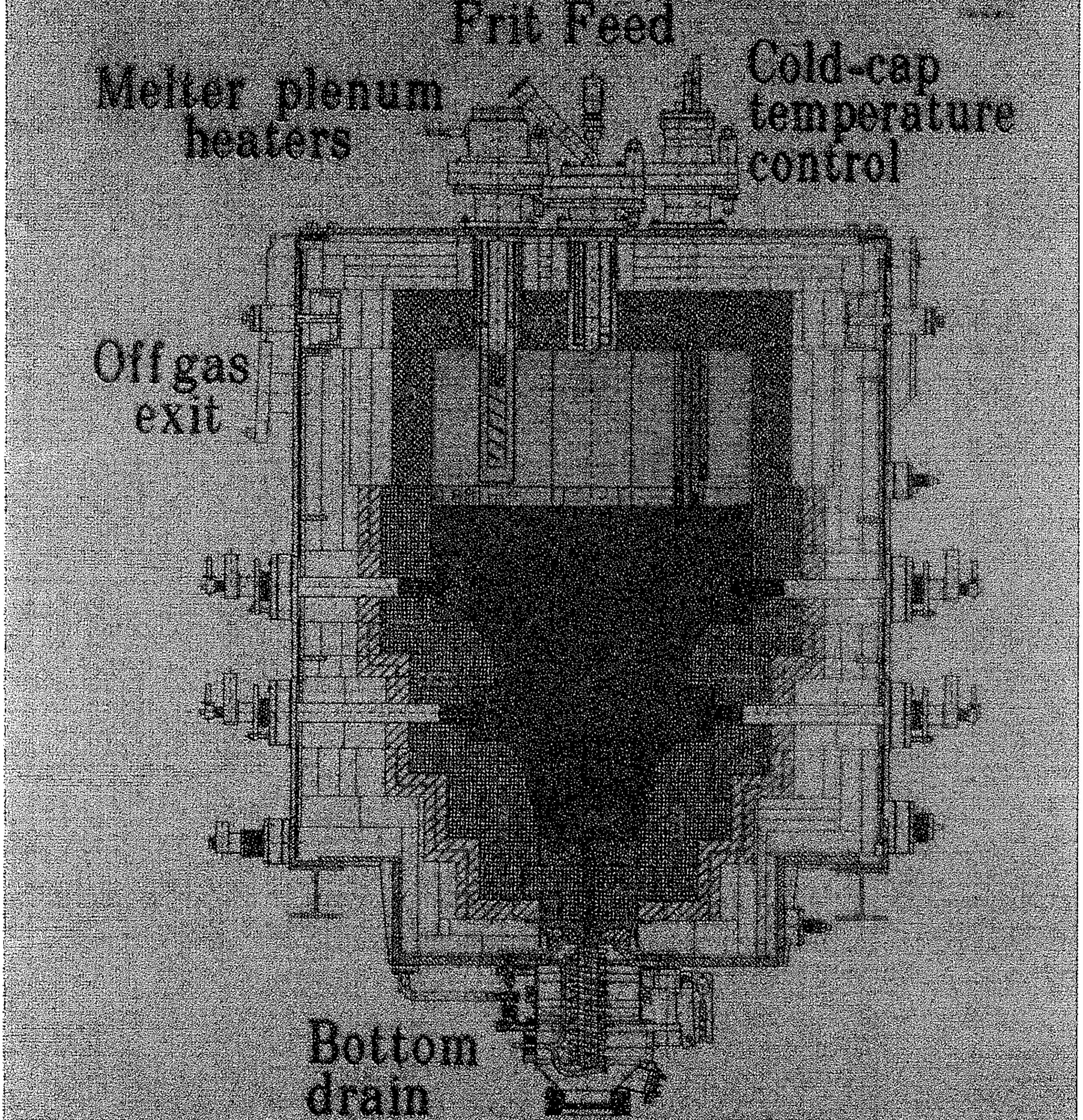
PHYSICAL AND MATHEMATICAL MELTER MODELLING

prepared for the

10th annual PNC-KfK meeting on HLLW management

held

November 18-22, 1990, Japan



Advanced melter design K-6'

MELTER MODELLING

Physical Modelling

Electric potential field
Electric field intensity

Mathematical modelling

Electric potential field
Power density distribution
Temperature field (incl. melter walls)
Convection pattern

Test program

Routine operation
Bottom electrode operation
Simulation of noble metal layer

Numerical Modelling

System definition (Geometry, physical properties)	
El. potential field $\phi\ (x,y,z)$	Conservation of el. charge ϕ (V)
El. field intensity $\vec{E}\ (x,y,z) = -\text{grad}\,\phi$.	$\vec{E}$ (V/cm)
Current density field $\vec{i}\ (x,y,z) = \sigma * \vec{E}$	$\vec{i}$ (Amp/cm^2) σ (1/Ωcm)
Power density field $P\ (x,y,z) = \sigma * \lvert\vec{E}\rvert^2$	P (W/cm^3)
Temperature field (melt + melter walls) $T\ (x,y,z)$	Conservation of mass and energy
Velocity field (glass melt) $\vec{v}\ (x,y,z)$	Conservation of momentum (Navier–Stokes equations)

MODEL BOUNDARIES/ASSUMPTIONS AND INPUT

GLASS MELT
Single-phase, incompressible Newtonian fluid
Optically thick material

COLD CAP
Heat sink, uniformly spread over melt surface
Total extent 90 % of melt surface
60 % boiling liquid (100°C), 30 % dry calcine (400°C)
10 % free surface at 900°C
No convection, temperature-dependant properties

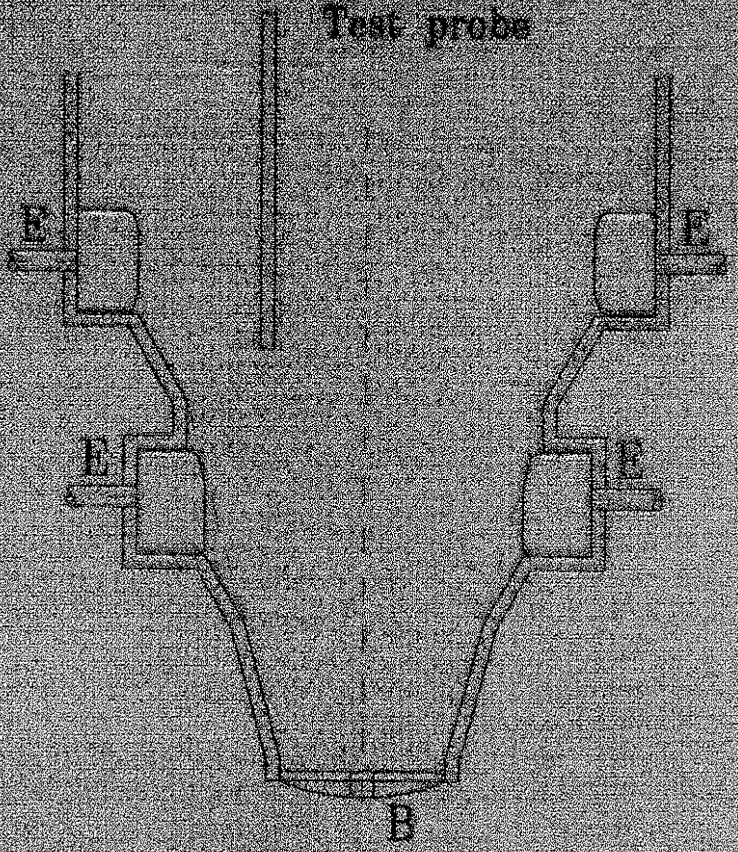
MELTER PLENUM
No convection, effective thermal conductivity (radiation)
Off-gas temperature 400°C

NOBLE METAL LAYER
Static layer, uniform distribution along inclined walls
Thickness 5-50 mm, el. conductivity 10 times higher

EXTERIOR MELTER SURFACE
Temperature distribution from linearly interpolated data

STARTING CONDITIONS
Uniform melt temperatures: 1100/1000°C upper/lower region
Initial temperature distribution (steady-state energy eq.)

DATA INPUT
El./thermal conductivity, viscosity, spec. heat,density
Electrode voltage adjustment

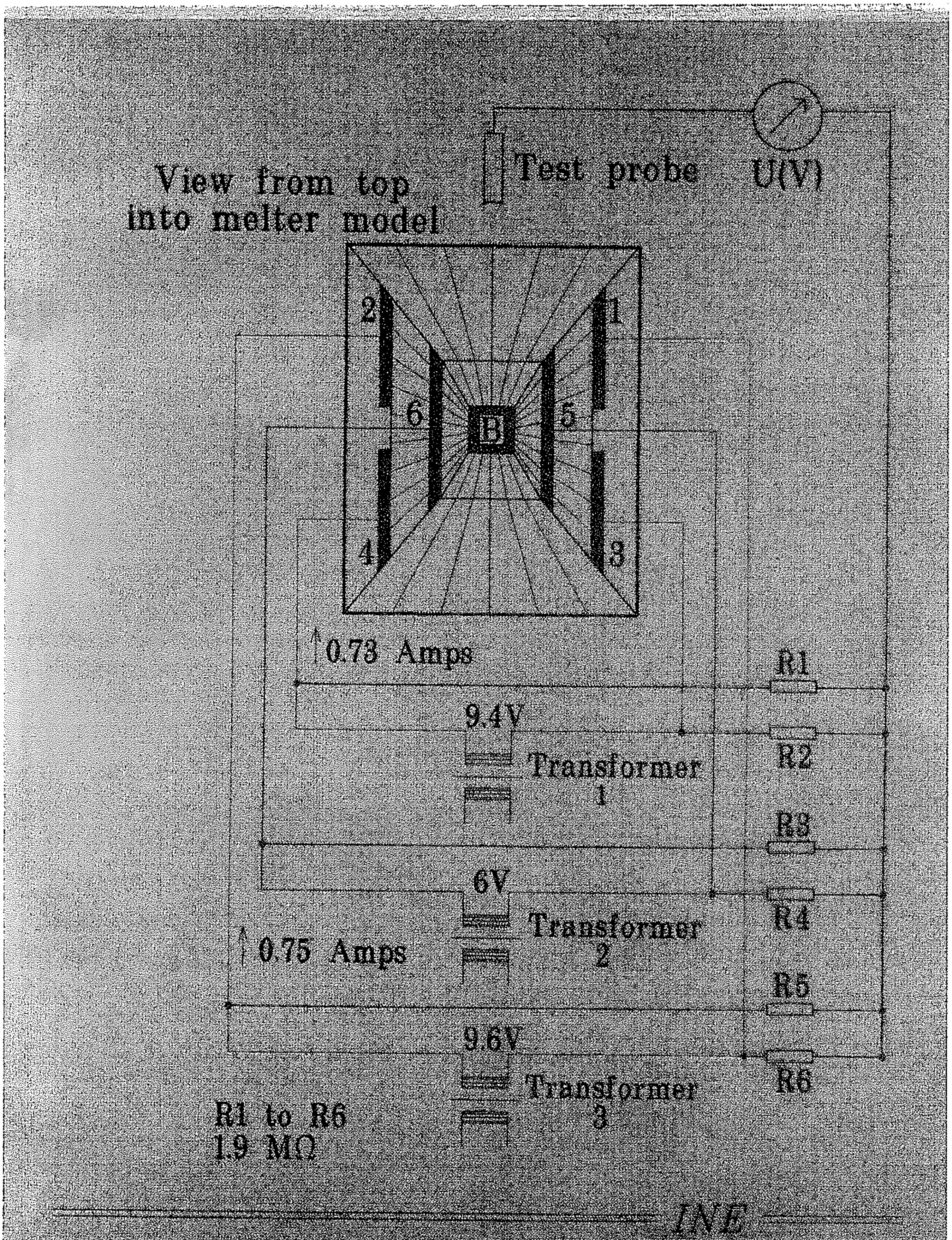


Model features

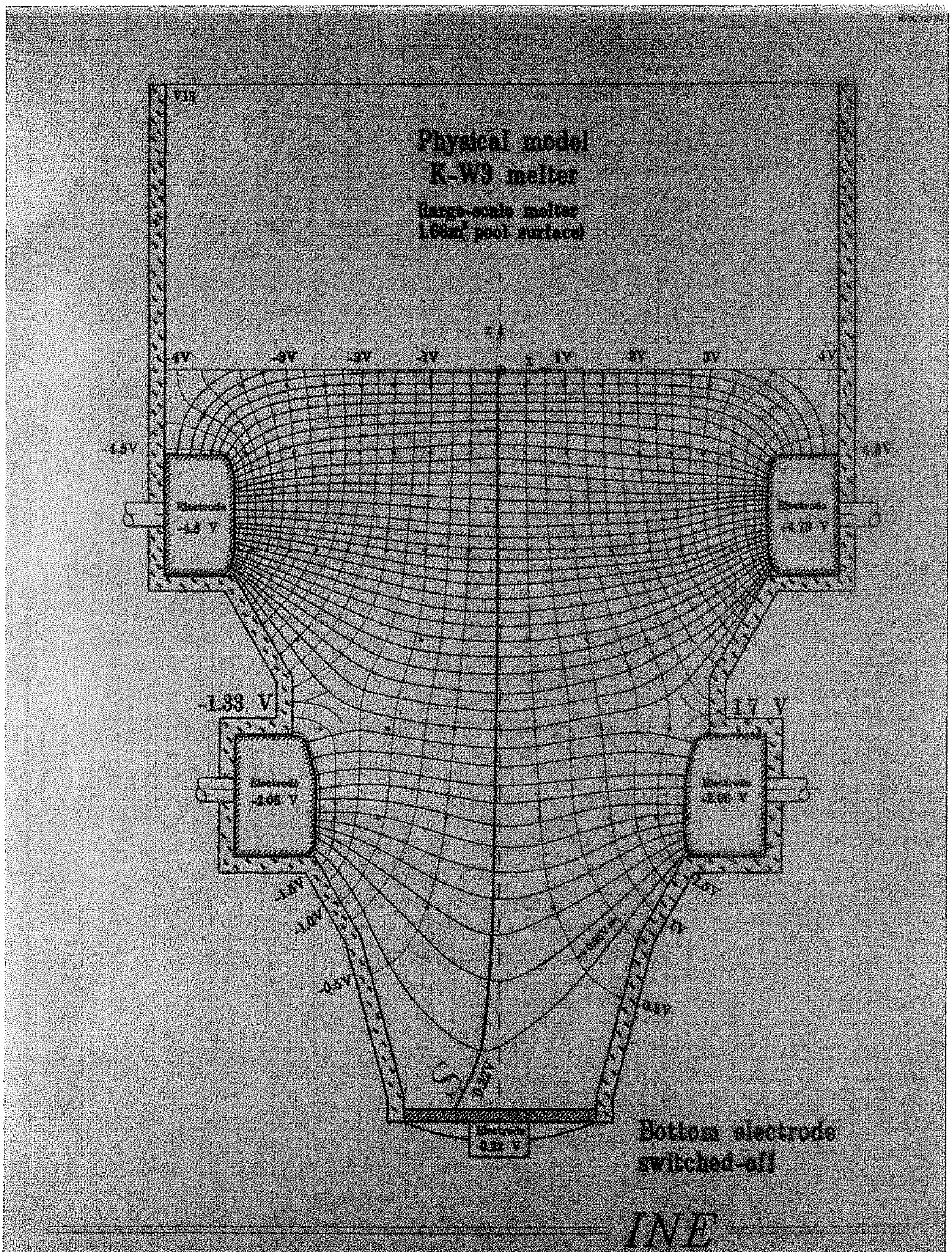
Scale	1 : 2.5
Electrodes	Copper, covered with Au-layer
Power supply	Full-scale melter system
Electrolyt	Salt water [Sodium Chloride 4.5 g/l]
El. resistivity	14.7 Ω cm
(Glass melt	6.5 Ω cm, 1150°C)
Metal-conductive layer	Aluminum foil
Thickness	0.02 mm
El. resistivity	2.86 x 10^{-6} Ω cm
(Noble metal sludge	~ 0.1 Ω cm)

INE

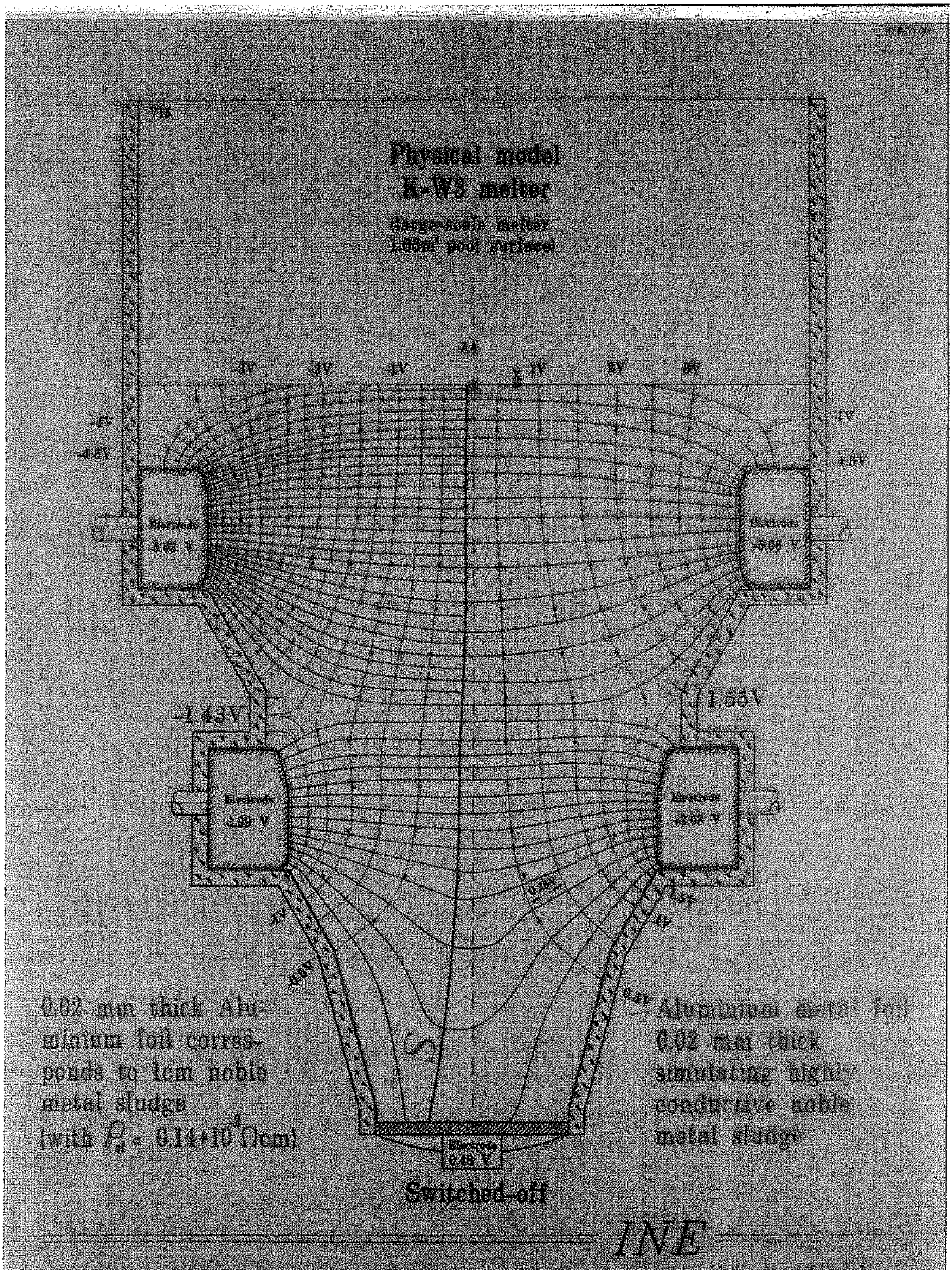
Description of physical model K-W3



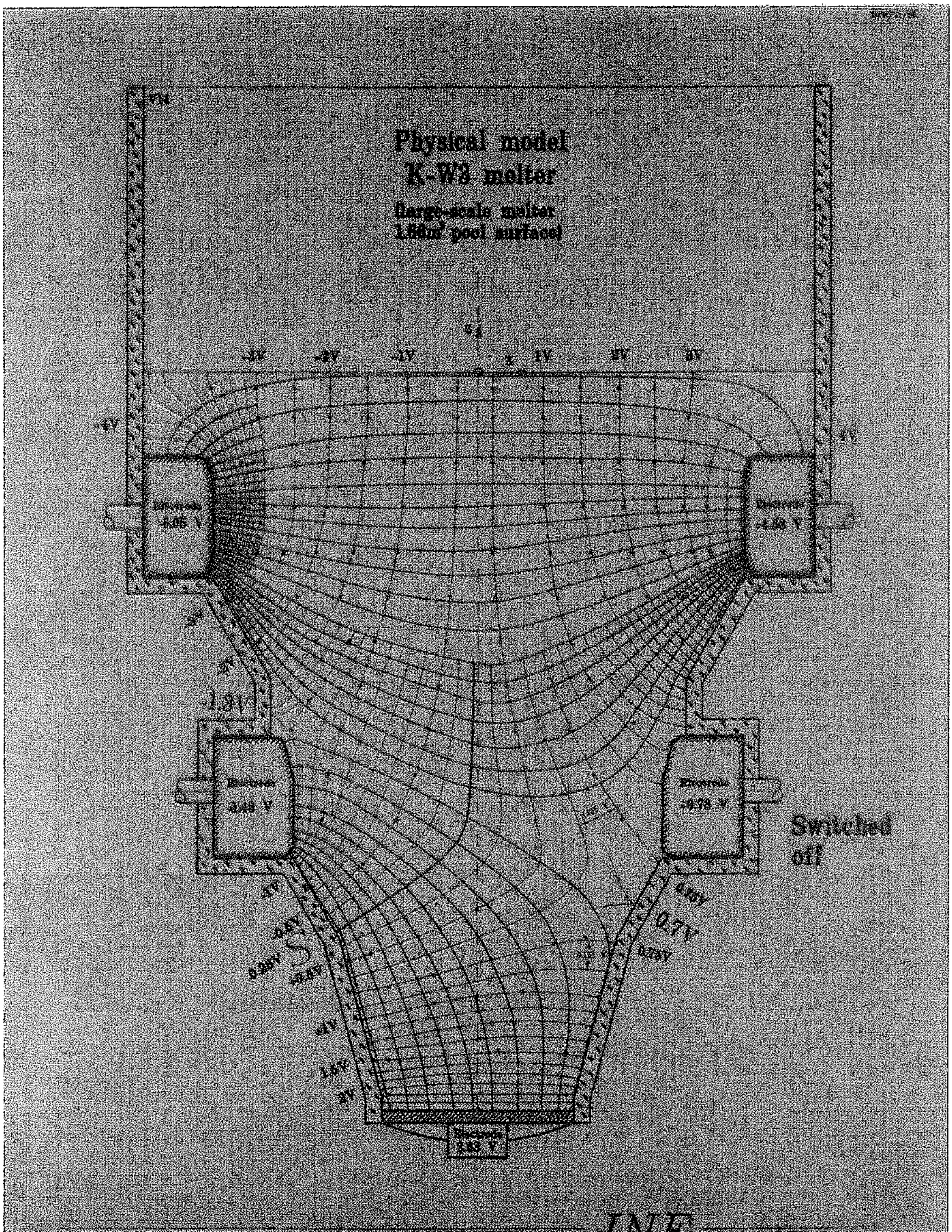
Electric supply system for the physical model of the K-W3 glass melter



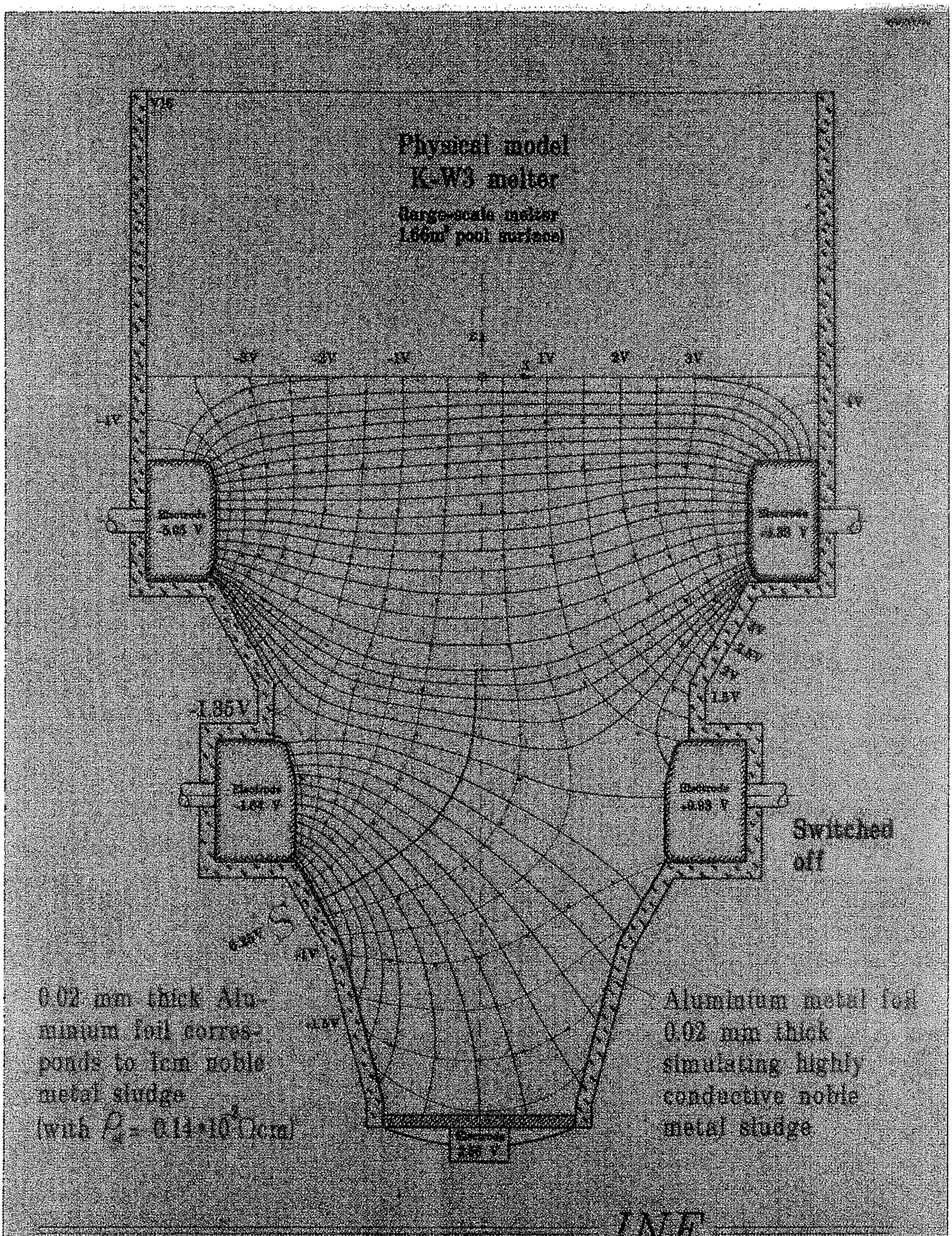
Measured electric potential field (green) and corresponding current field (red). Normal situation.



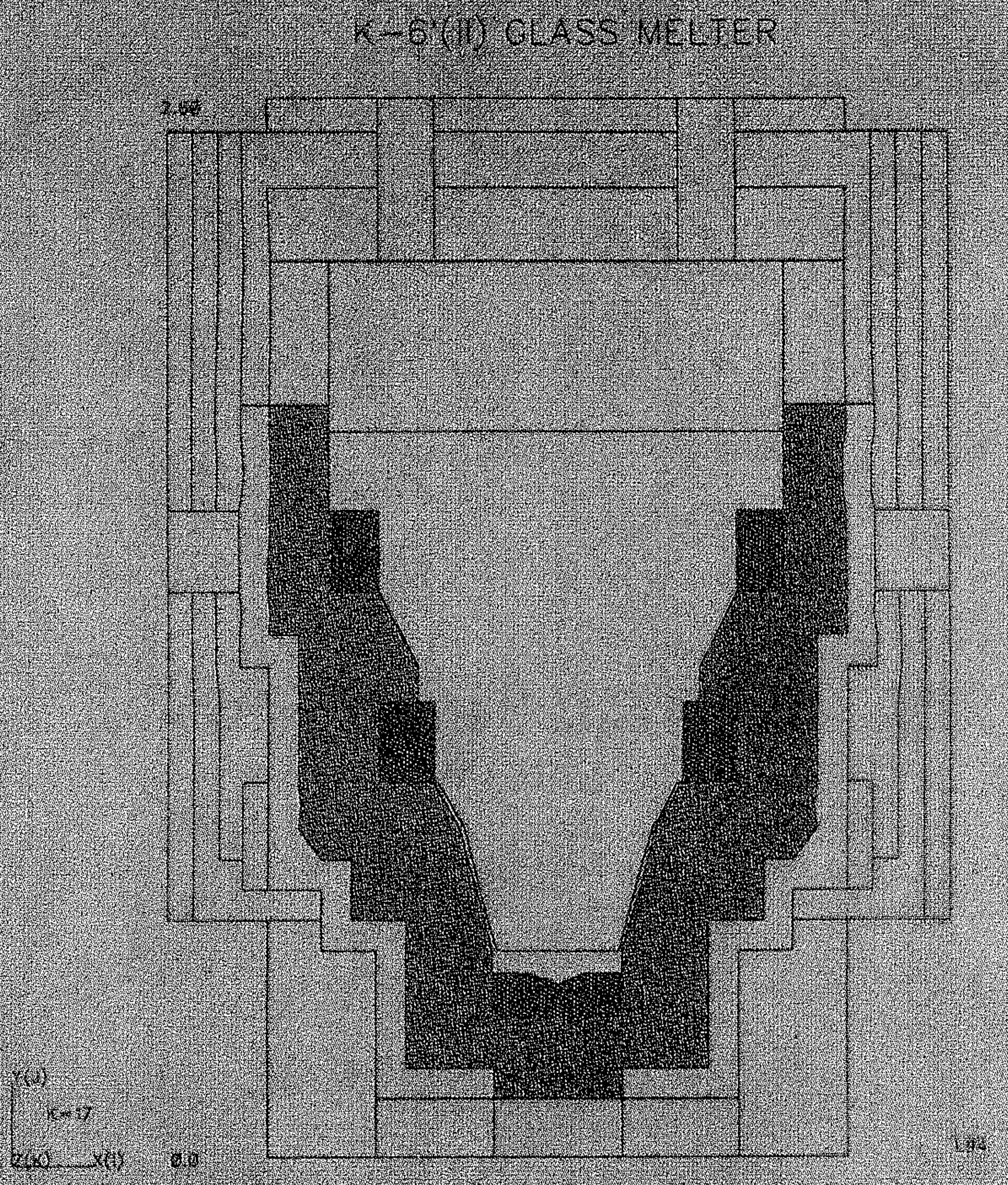
El. potential field (green) and current field (red). Lower melter walls covered with Al-metal foil. Routine electrode firing



Electric potential field (green) and current field (red)
Firing including bottom electrode



El. potential field (green) and current field (red). Lower melter walls covered with Al-metal foil. Firing incl. bottom electrode



MATERIAL BOUNDARIES

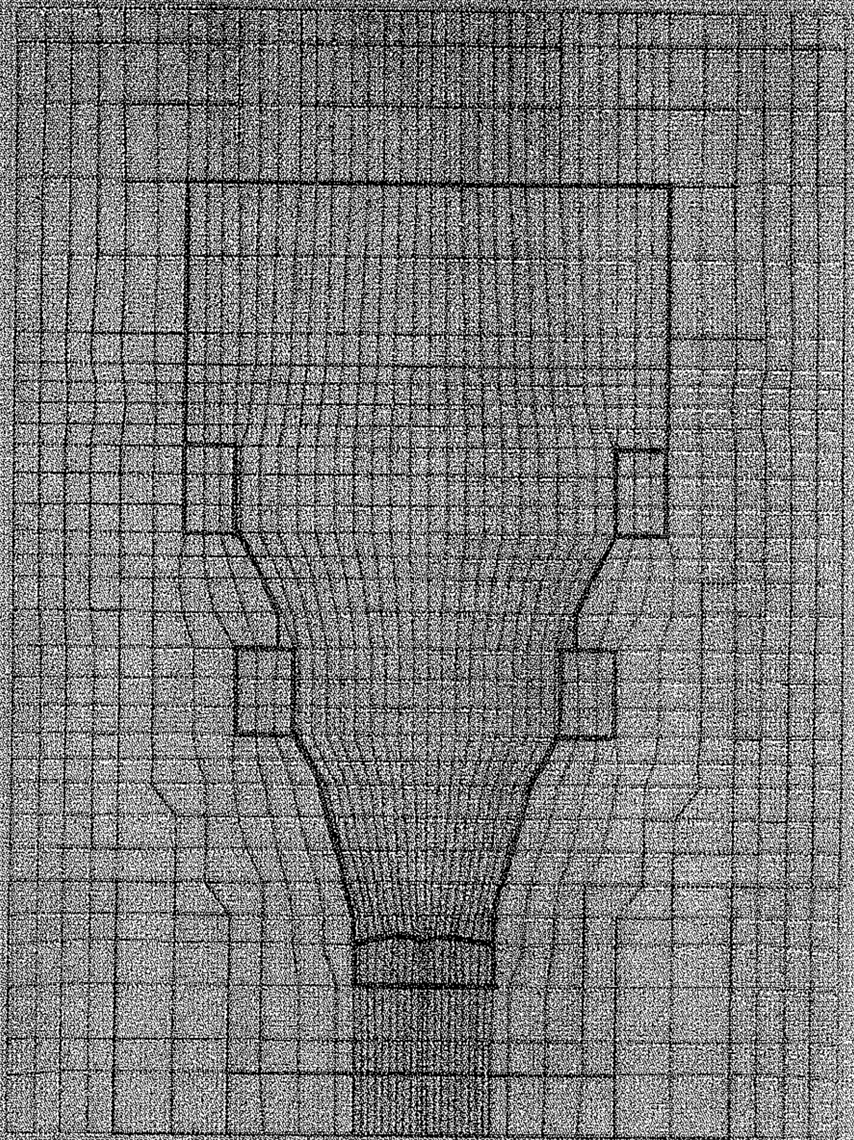


Fig. 3: NUMERICAL GRID AND MATERIAL BOUNDARIES

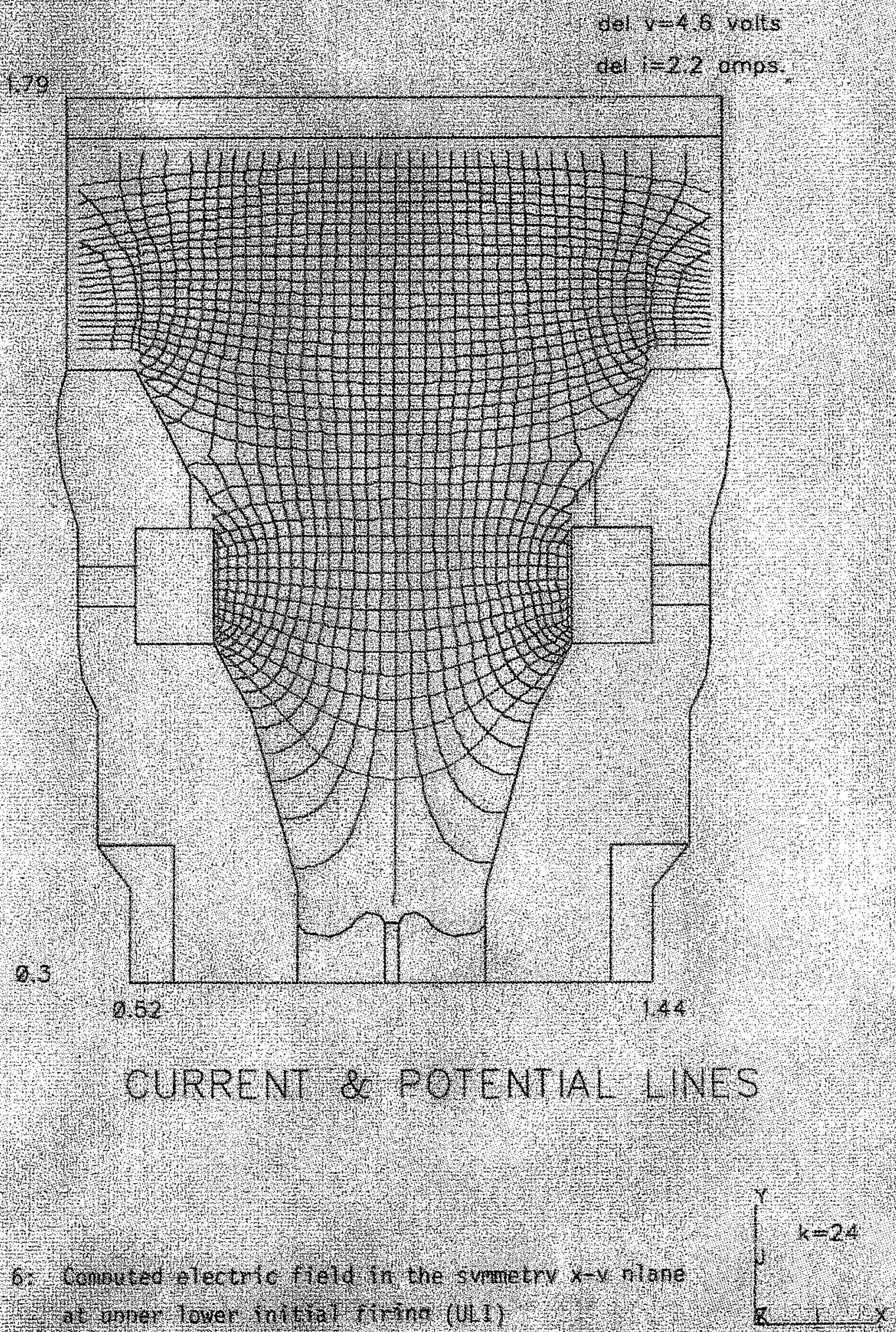


Fig 6: Computed electric field in the symmetry x-y plane at upper lower initial firing (ULI)

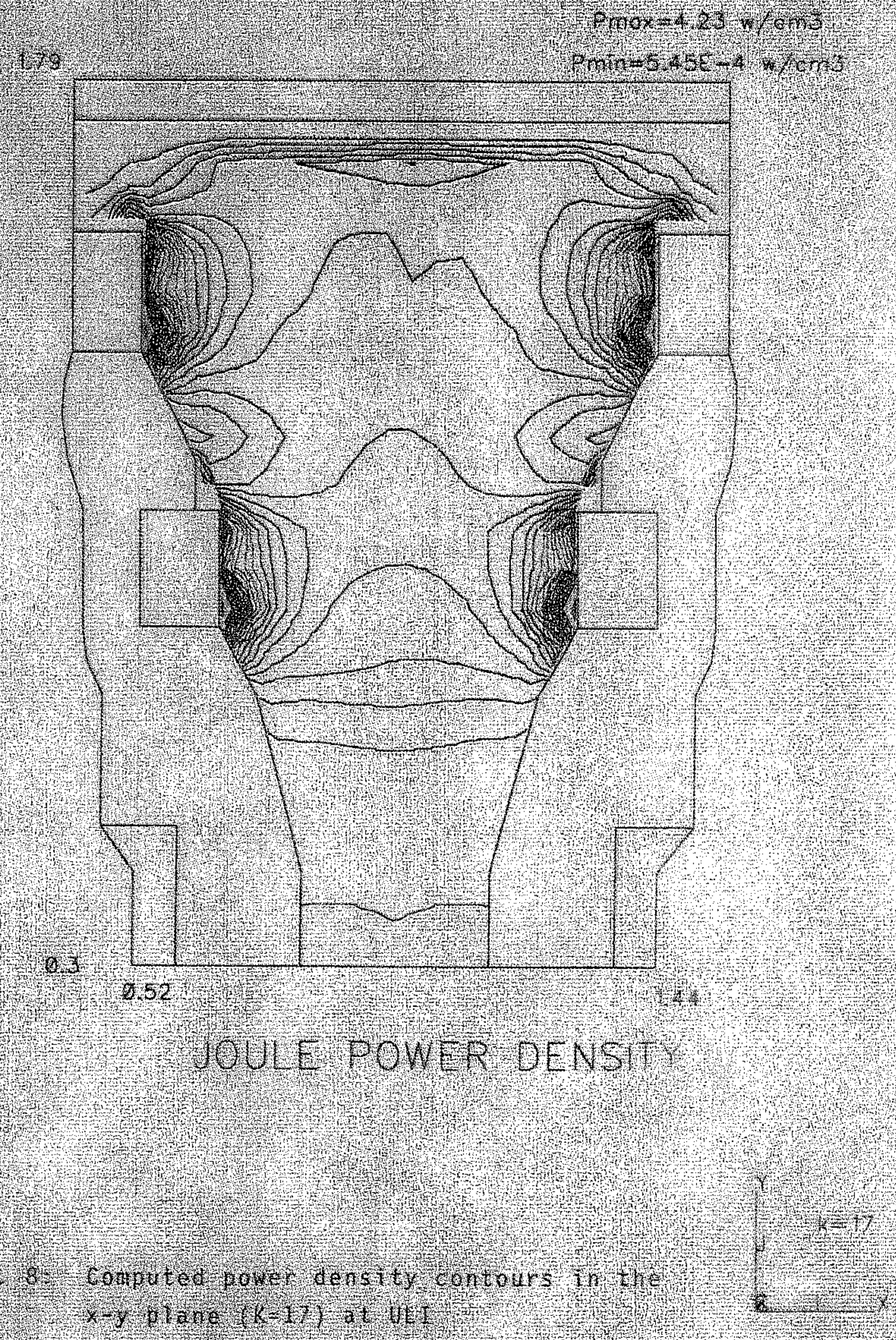


Fig. 8: Computed power density contours in the x-y plane (K=17) at UEI

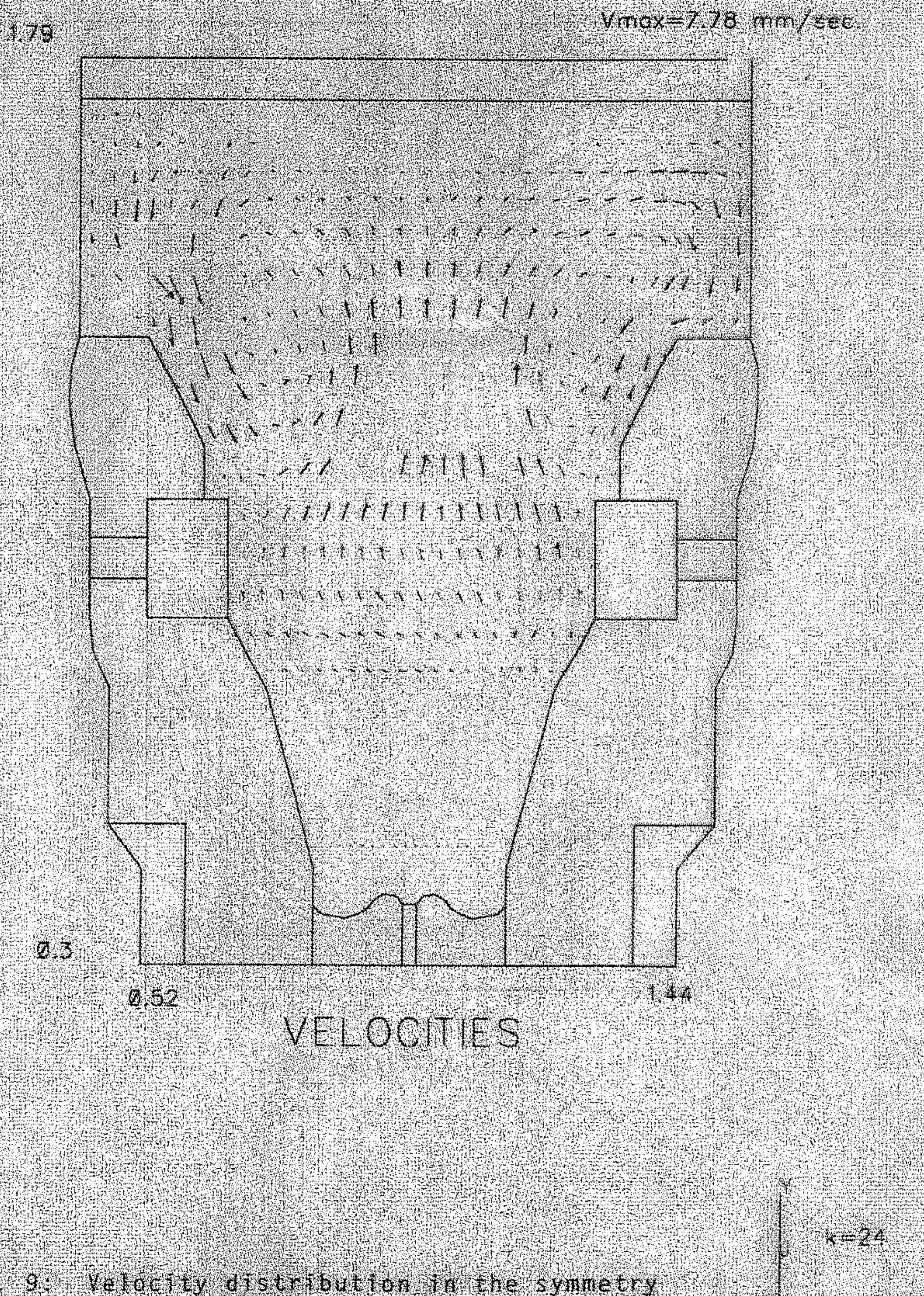


Fig. 9: Velocity distribution in the symmetry x-y plane at ULI

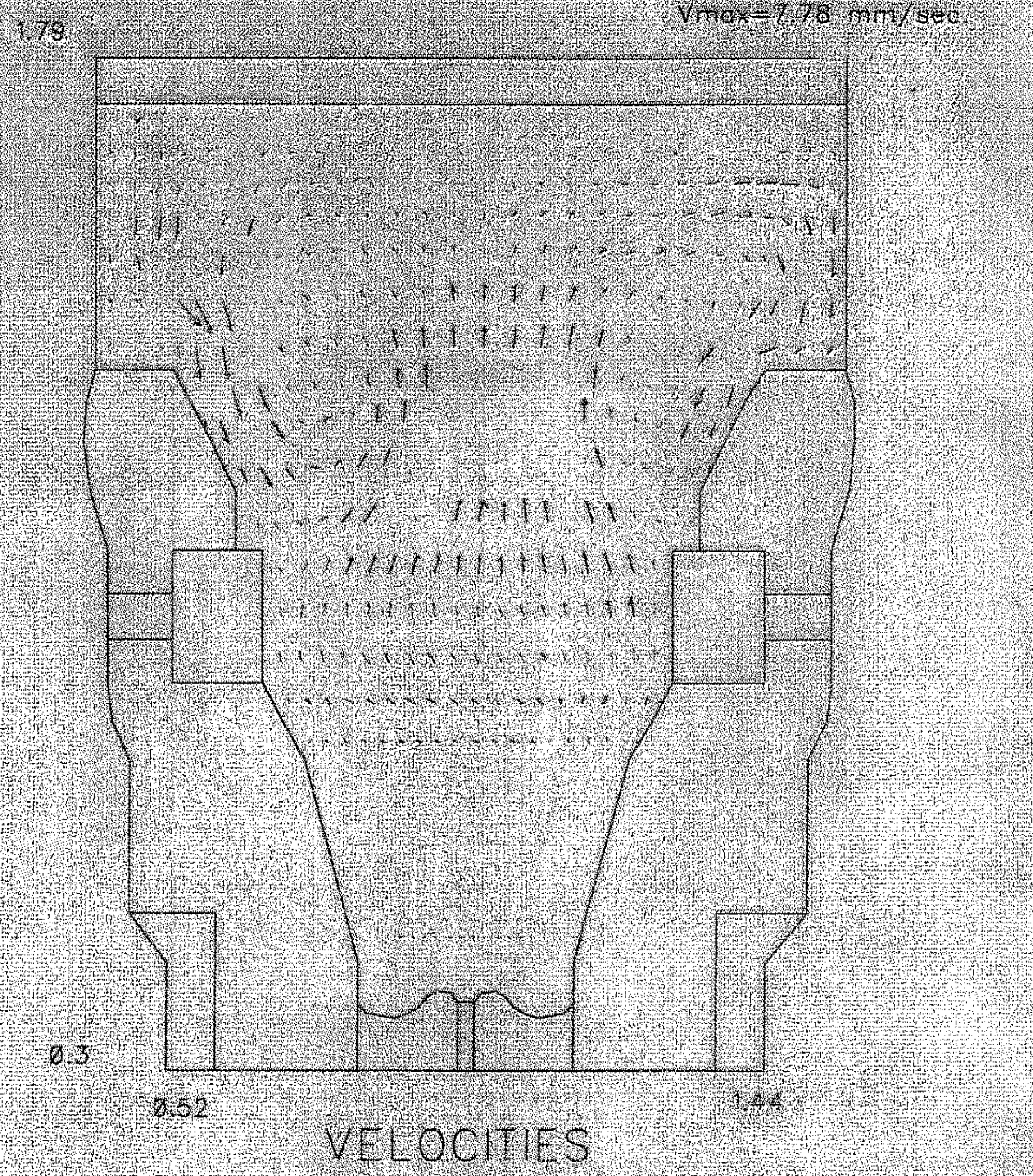


Fig. 91 Velocity distribution in the symmetry x-y plane at ULI

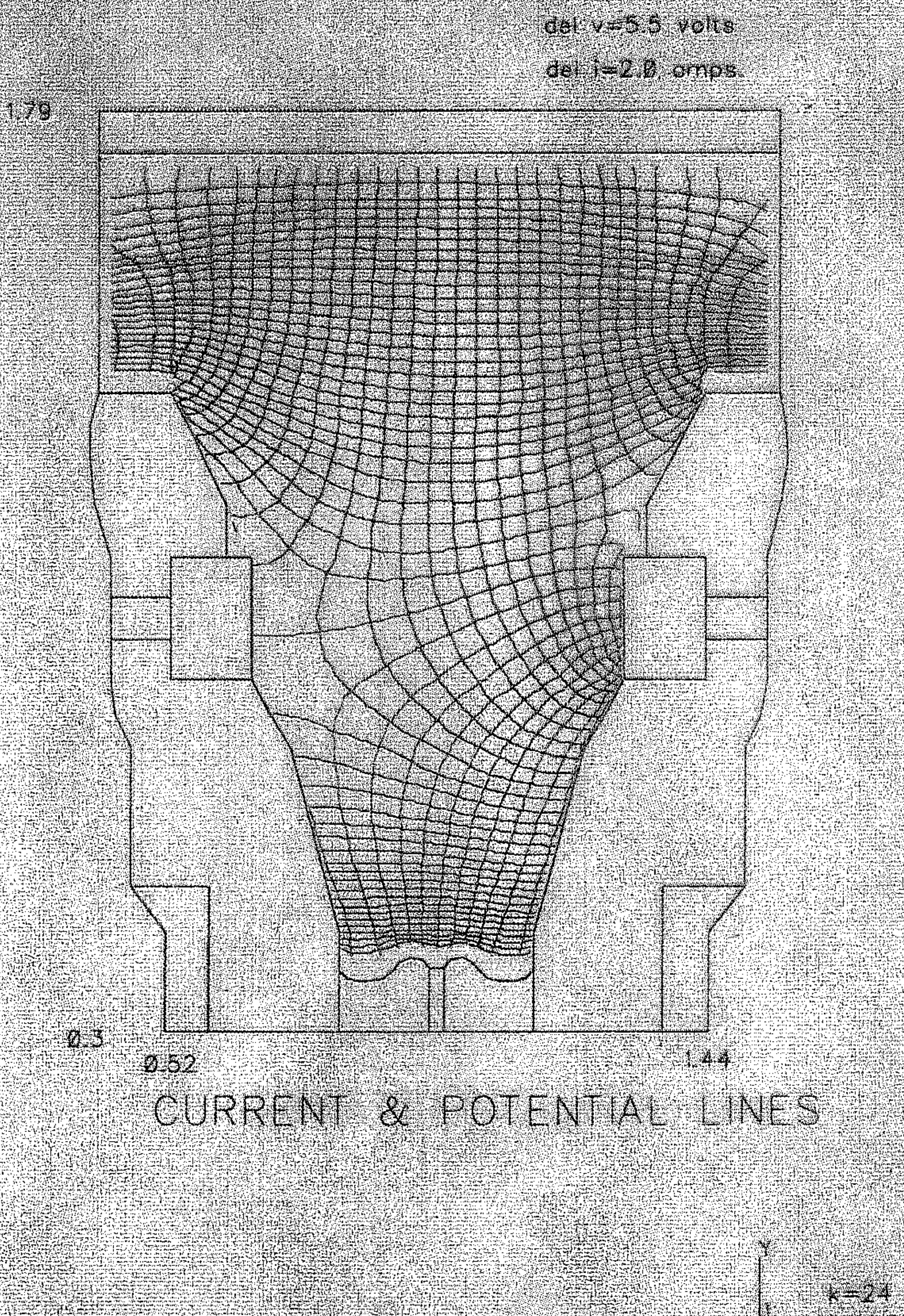


Fig. 17: Computed electric field in the symmetry x-y plane with bottom firing UBR

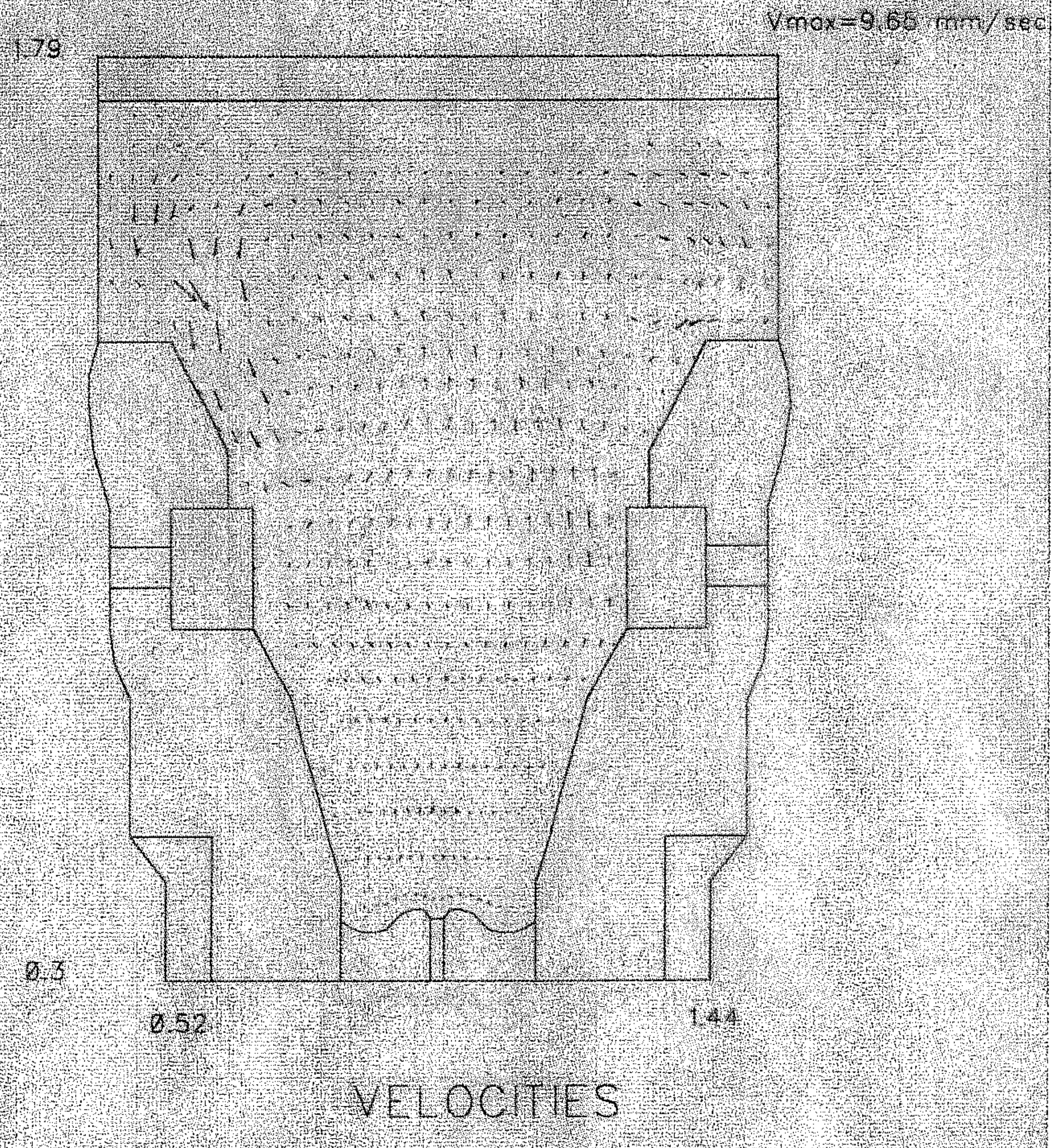


Fig. 20: Velocitiy distribution in the symmetry x-y plane with bottom firing UBR

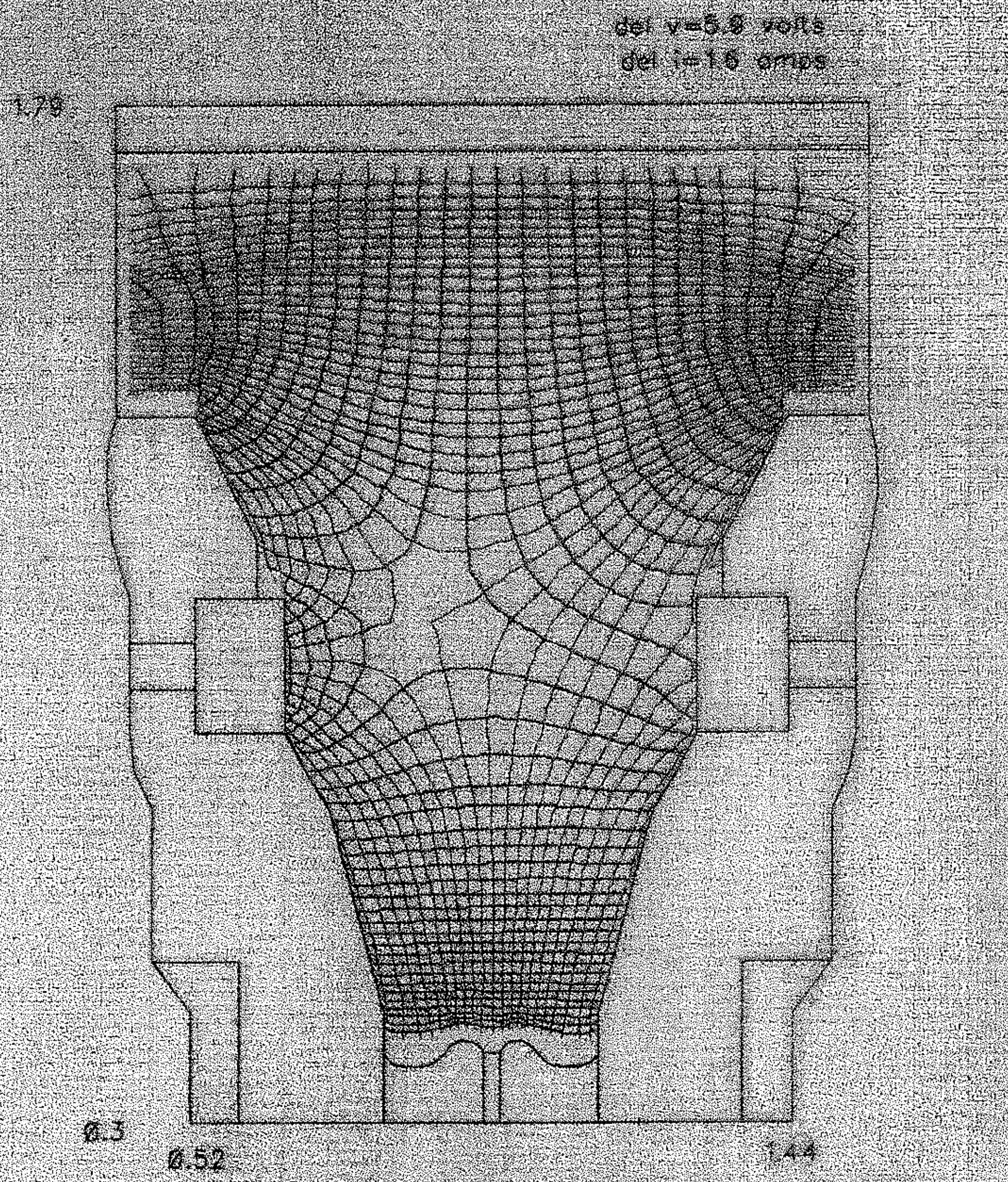


Fig. 22: Computed electric field in the symmetry x-y plane with bottom firing towards left UBL

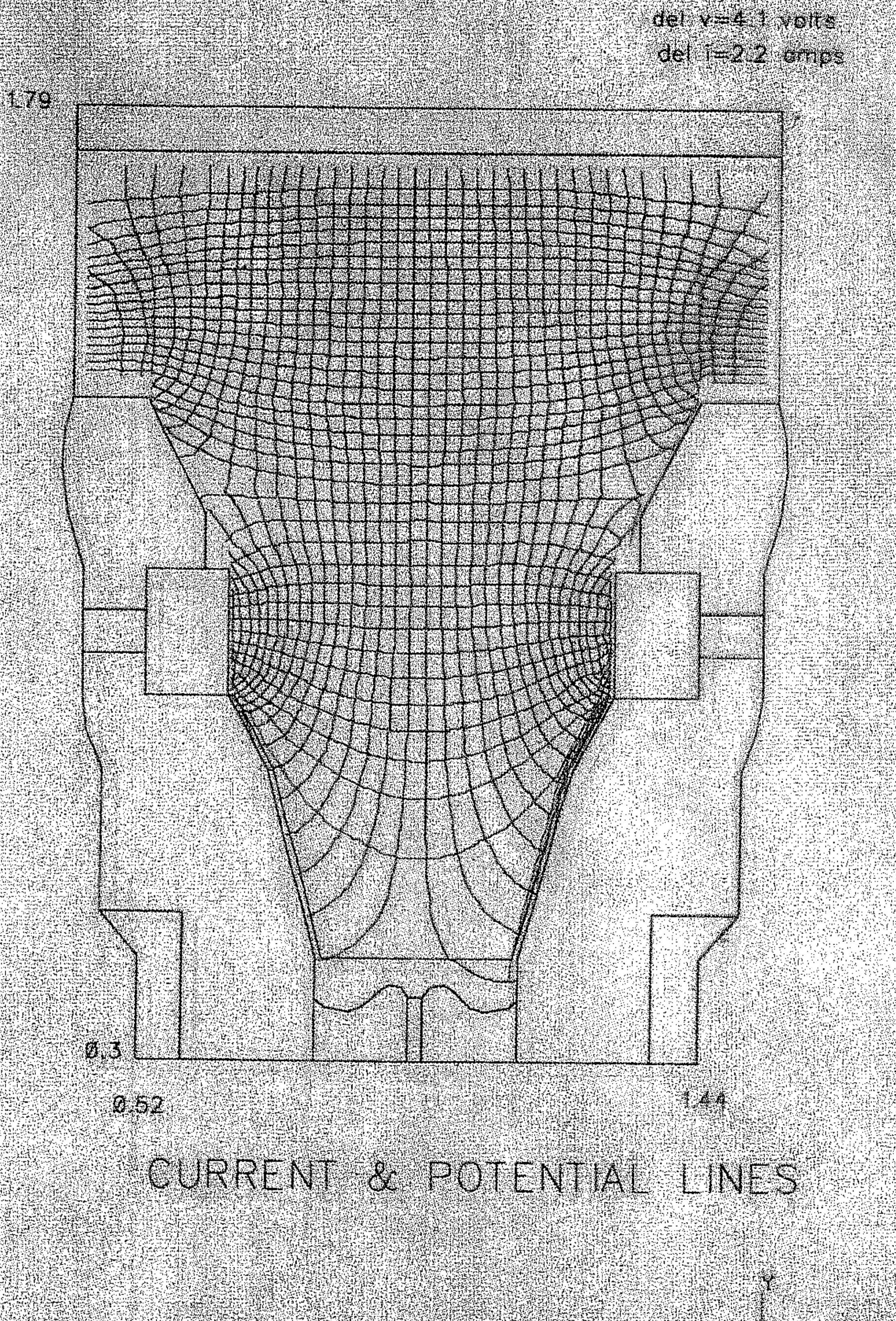


Fig. 36: Computed electric field in the symmetry x-y plane at ULI in presence of metal enriched layer

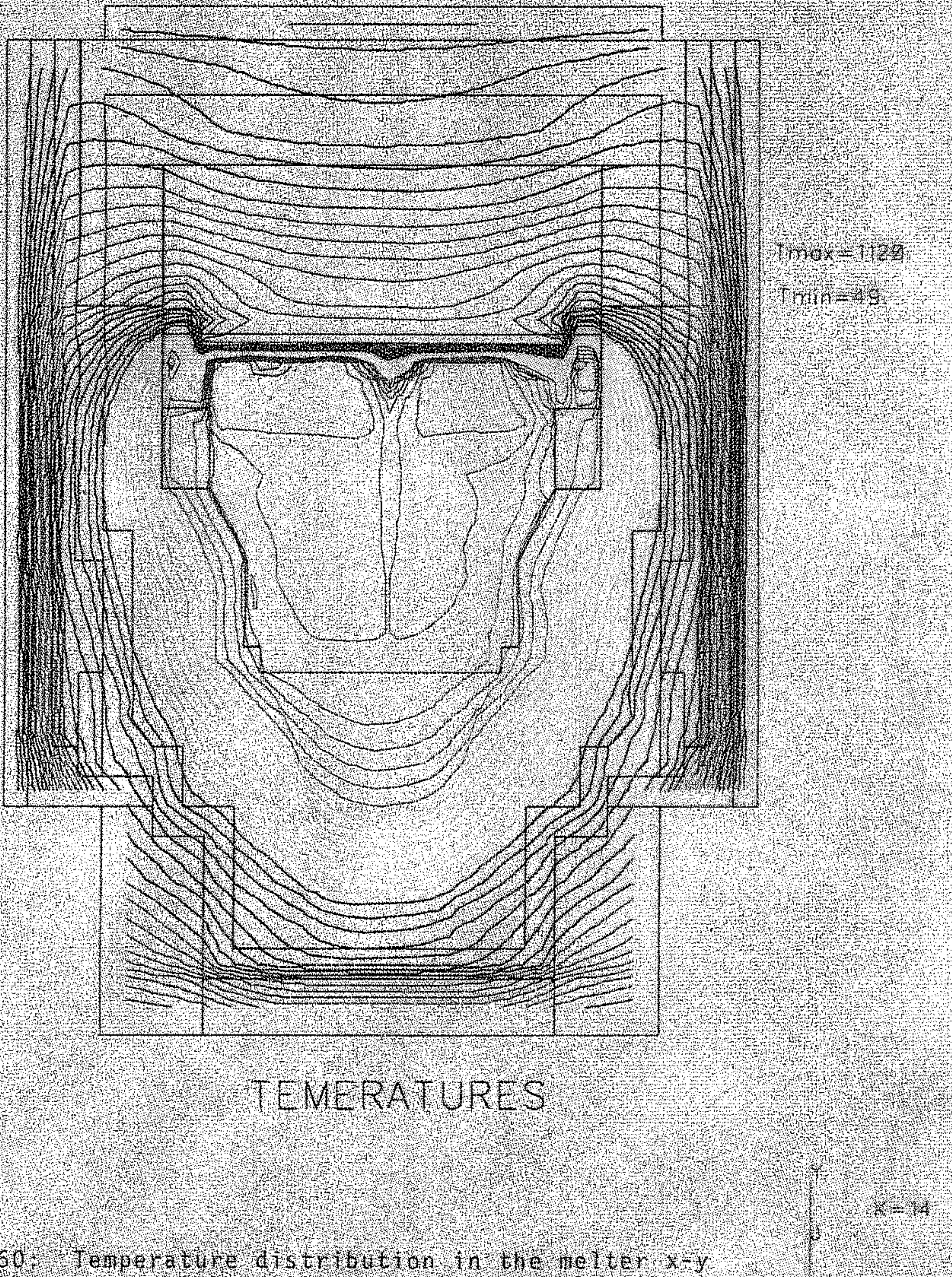


Fig. 60: Temperature distribution in the melter x-y plane (K=14) after UL8

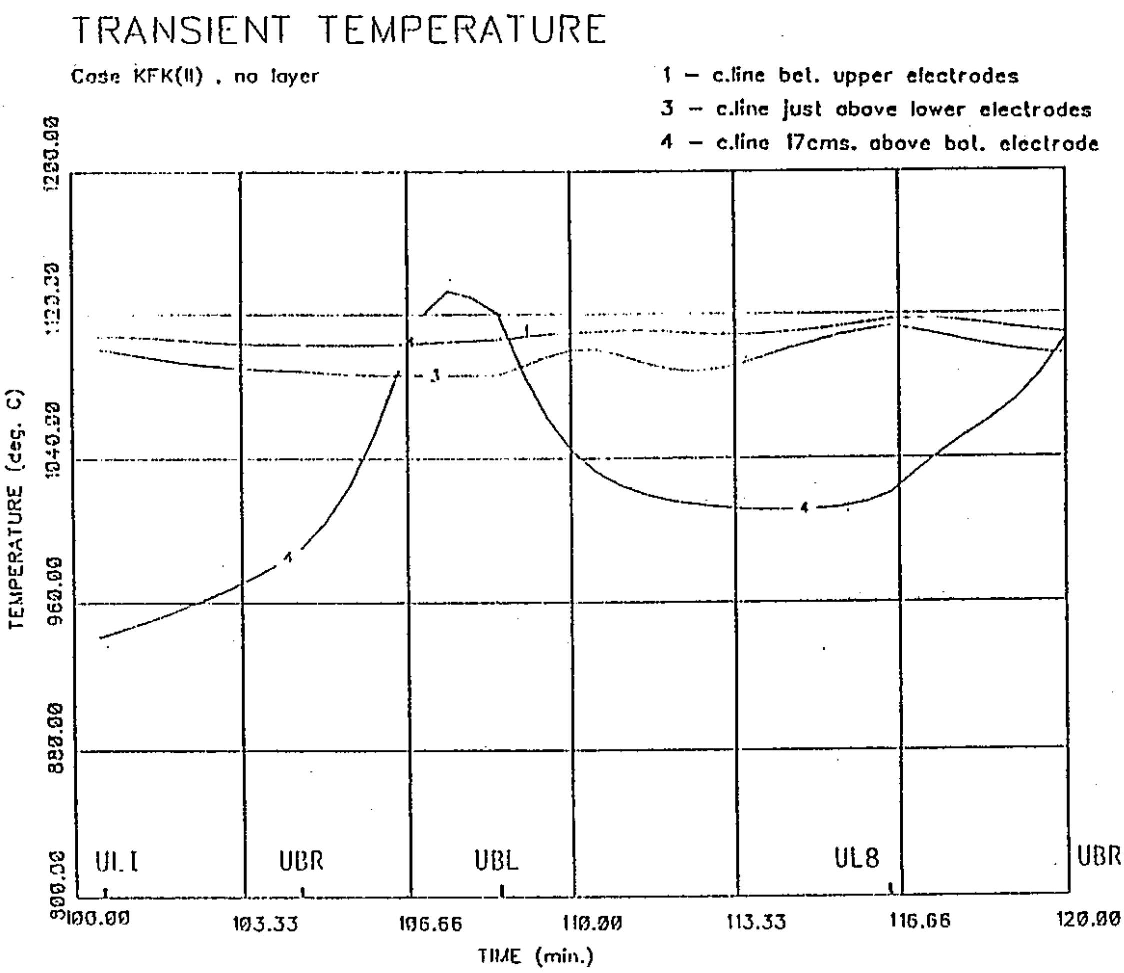
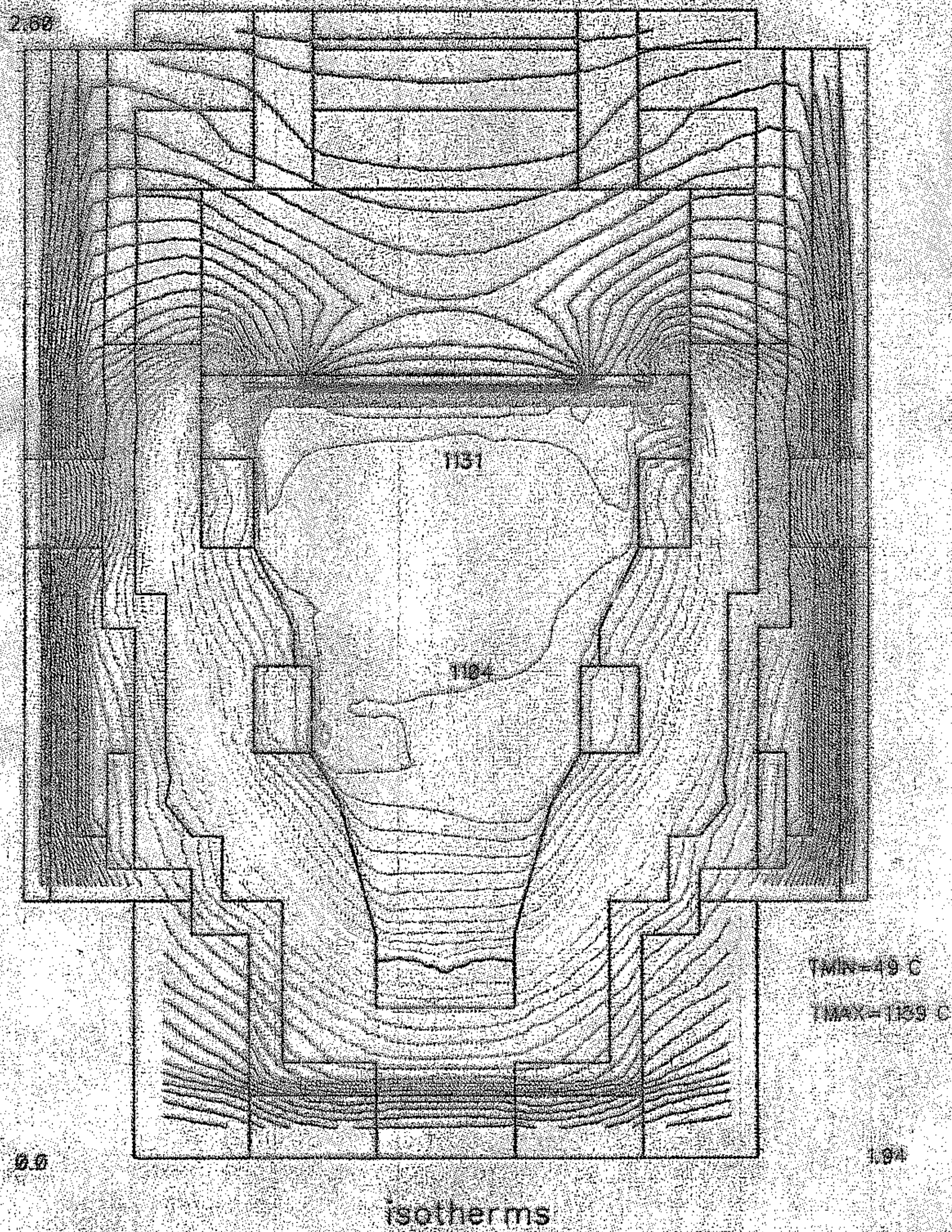


Fig. 16: Temperature variation with time at the monitor locations

K－6'(II) GLASS MELTER



3－D COMPUTED TEMPERATURE FIELD
WITHOUT NOBEL METAL ACCUMULATION

3-5 Interactions between Melter Ceramics and Glass Product Melts

Interactions between Melter Ceramics and Glass Product Melts

Dr. R. Knitter, Prof. H. Pentinghaus

Prepared for the 10th PNC/KfK Annual Meeting on HLLW-Management

November 18 - 22, 1990, Japan

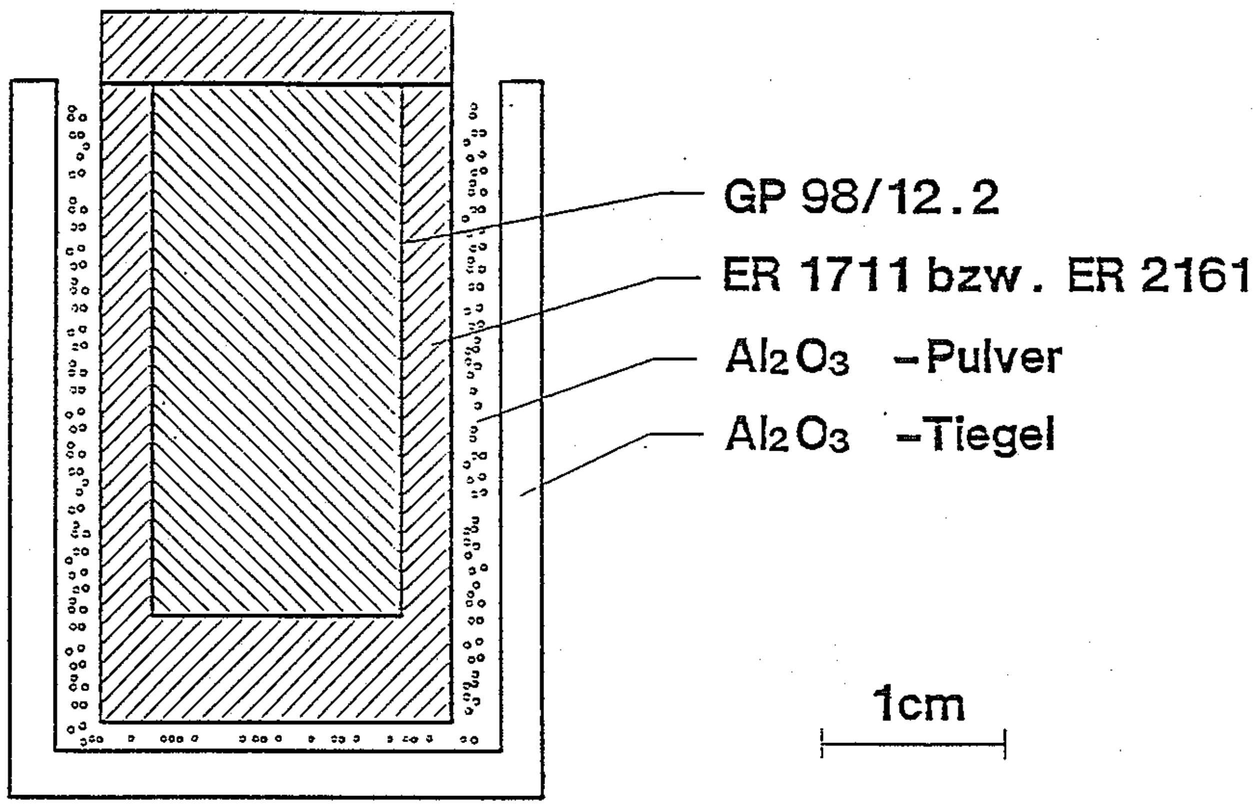
KfK

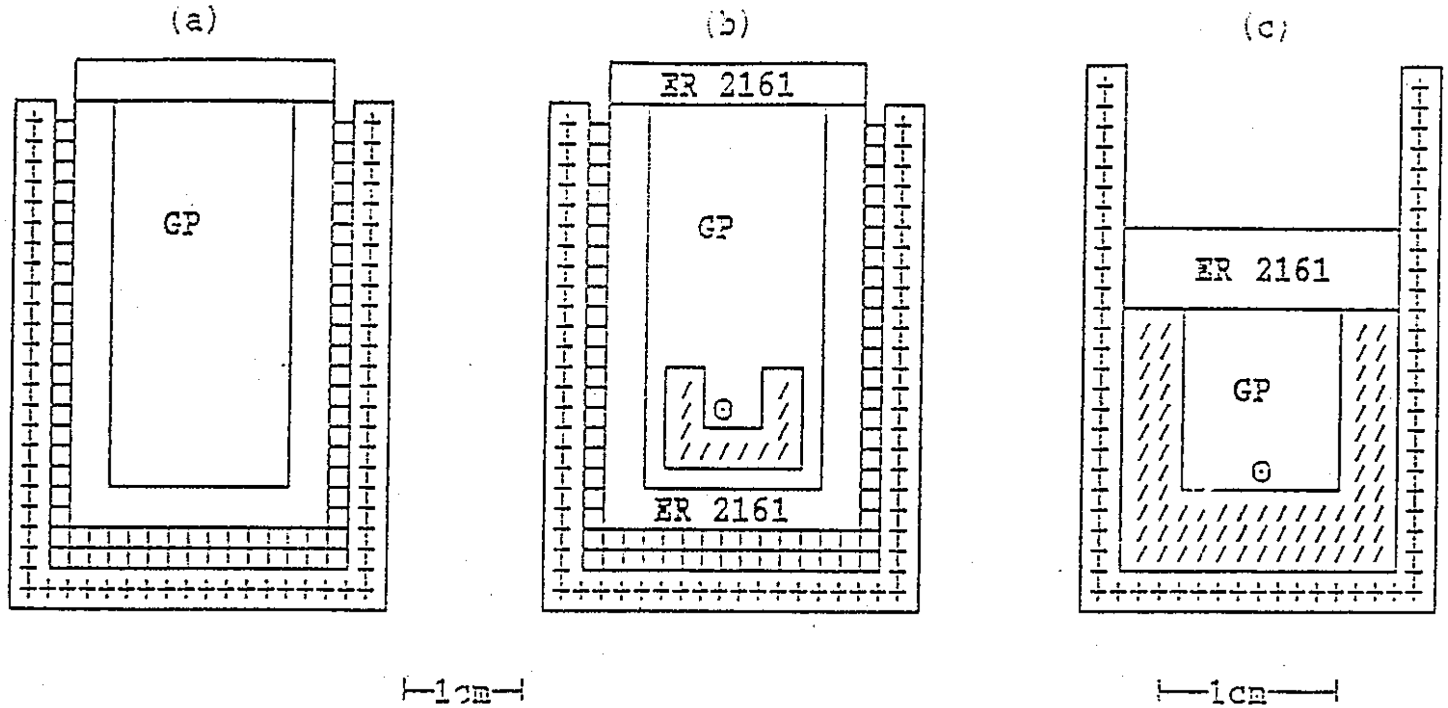
13.11.90 Pe 6

A1

Contents:

- Reaction cell
- Composition and texture of melt -cast ER 1711 and ER 2161
- Textural changes during annealing at 1000°C and 1150°C
- Microprobe analyses


GP 98/12.2
ER 1711 bzw. ER 2161
Al_2O_3 –Pulver
Al_2O_3 –Tiegel
1cm



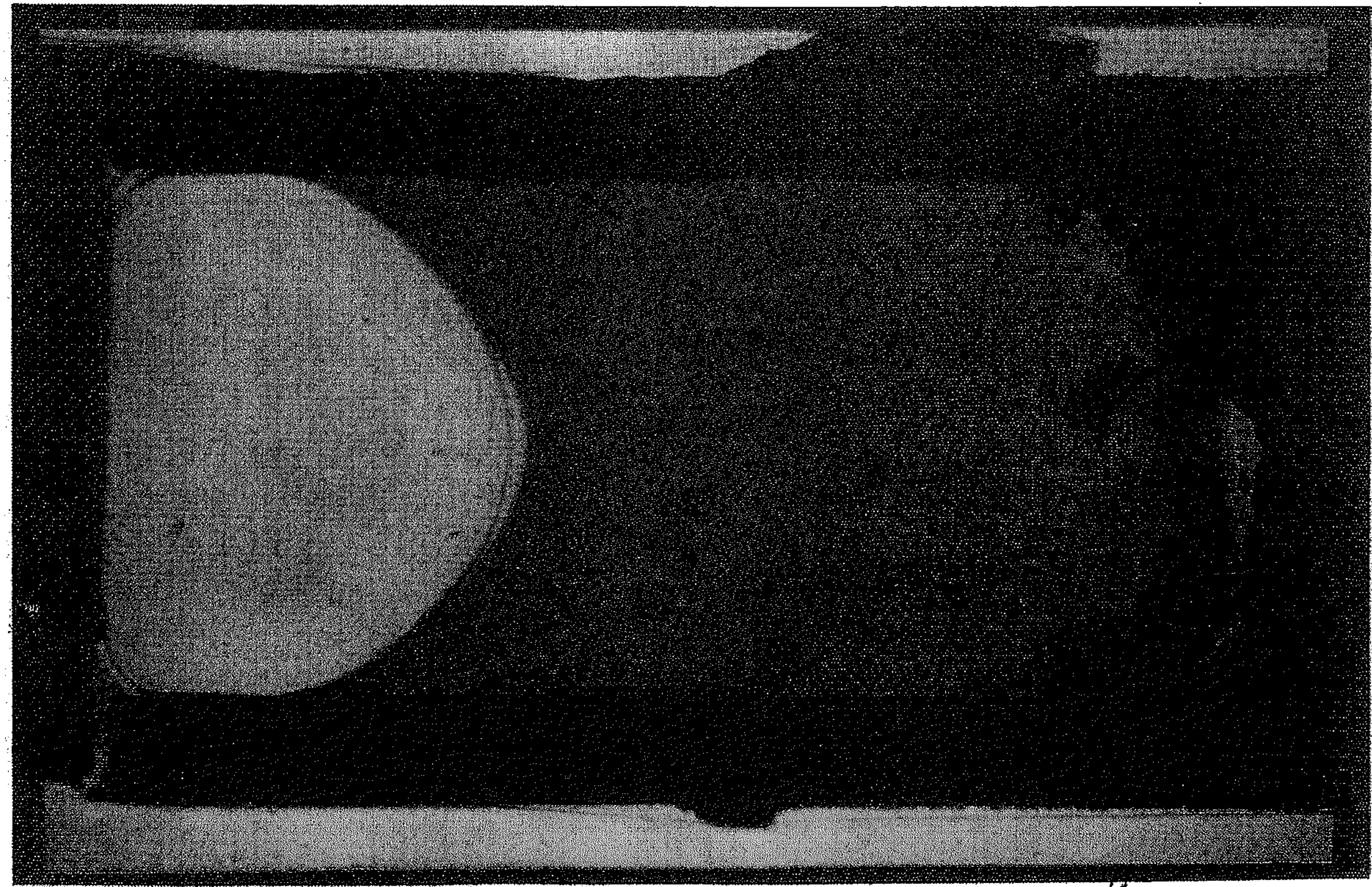
ER 2161 bzw. ER 1711, // INCONEL 690, ++ Korund-Tiegel,

Korund-Pulver, ⊙ Legierung 44 bzw. 37, GP Glasprodukt.

Schematische Darstellung der Meßzellen für die Wechselwirkungsexperimente zwischen (a) Keramik und Glasprodukt, (b) ER 2161, GP 98/12.2, INCONEL 690 und *Legierung 37, (c) INCONEL 690 und GP 98/12.2* und *Legierung 44 bzw 37.*

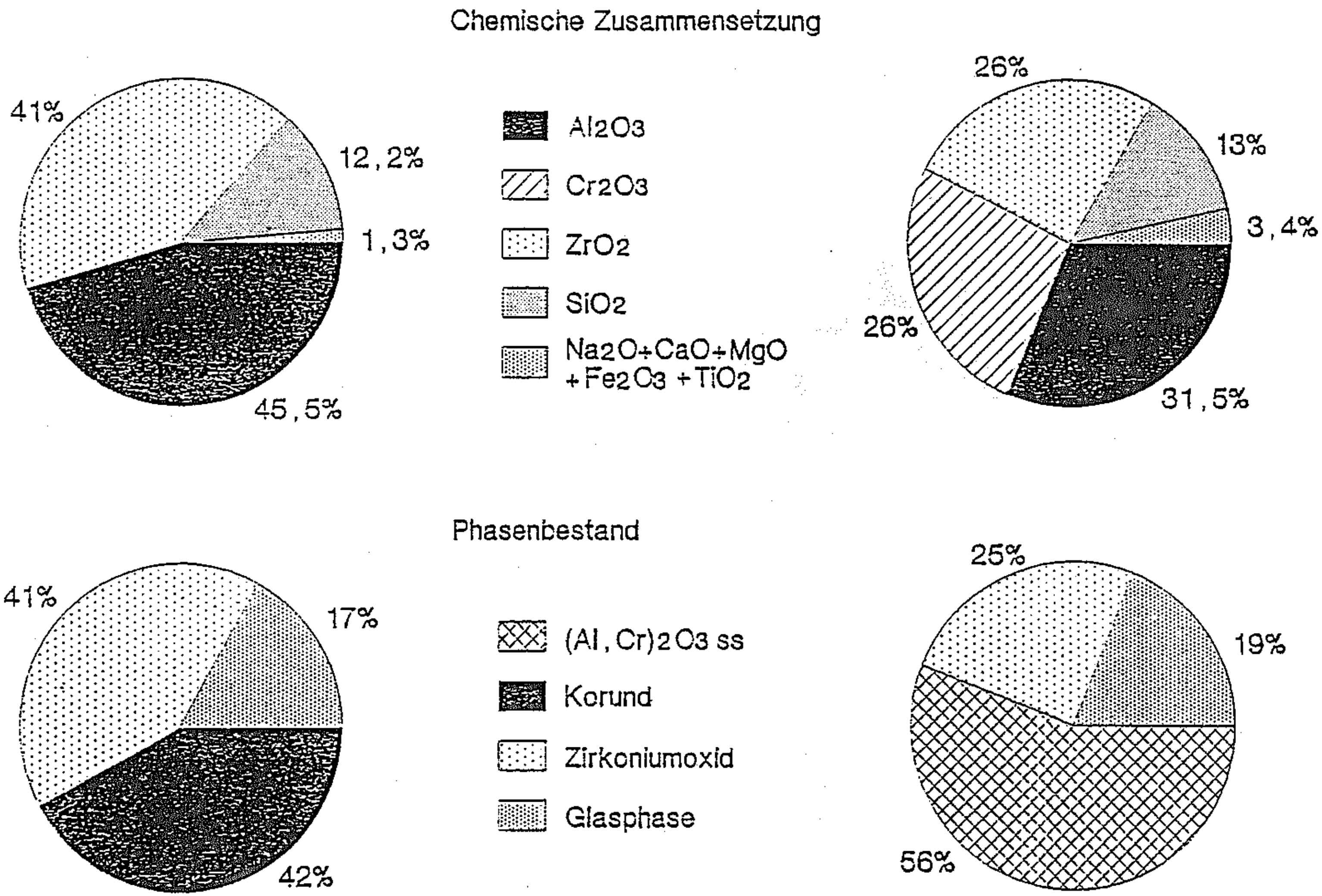
KfK 7.11.89 Pe 8

C 1 Reaction cells for interaction experiments

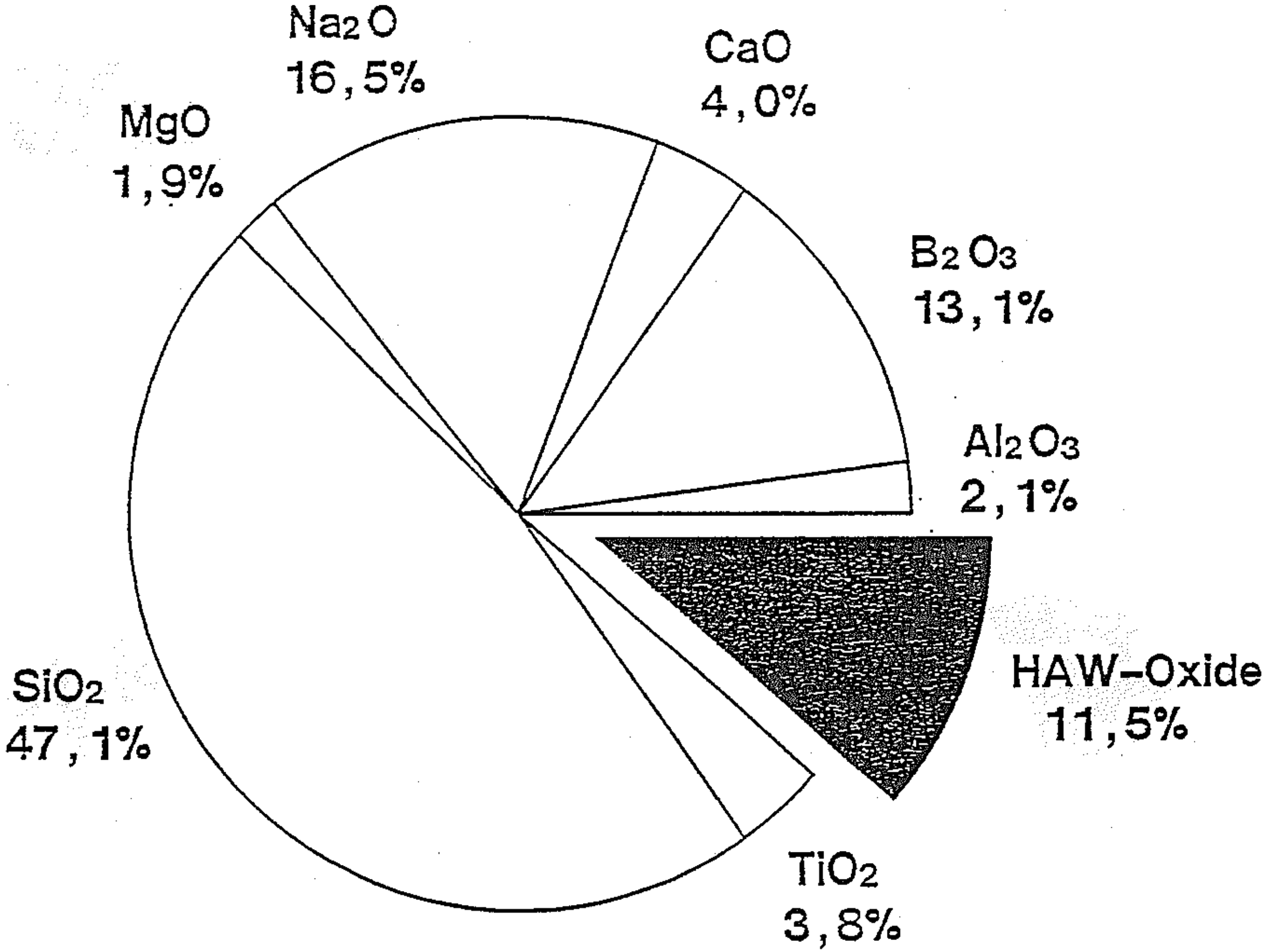


KfK 7.11.89 Pa 9

C 2 Thick section of an ER 2161 crucible after annealing

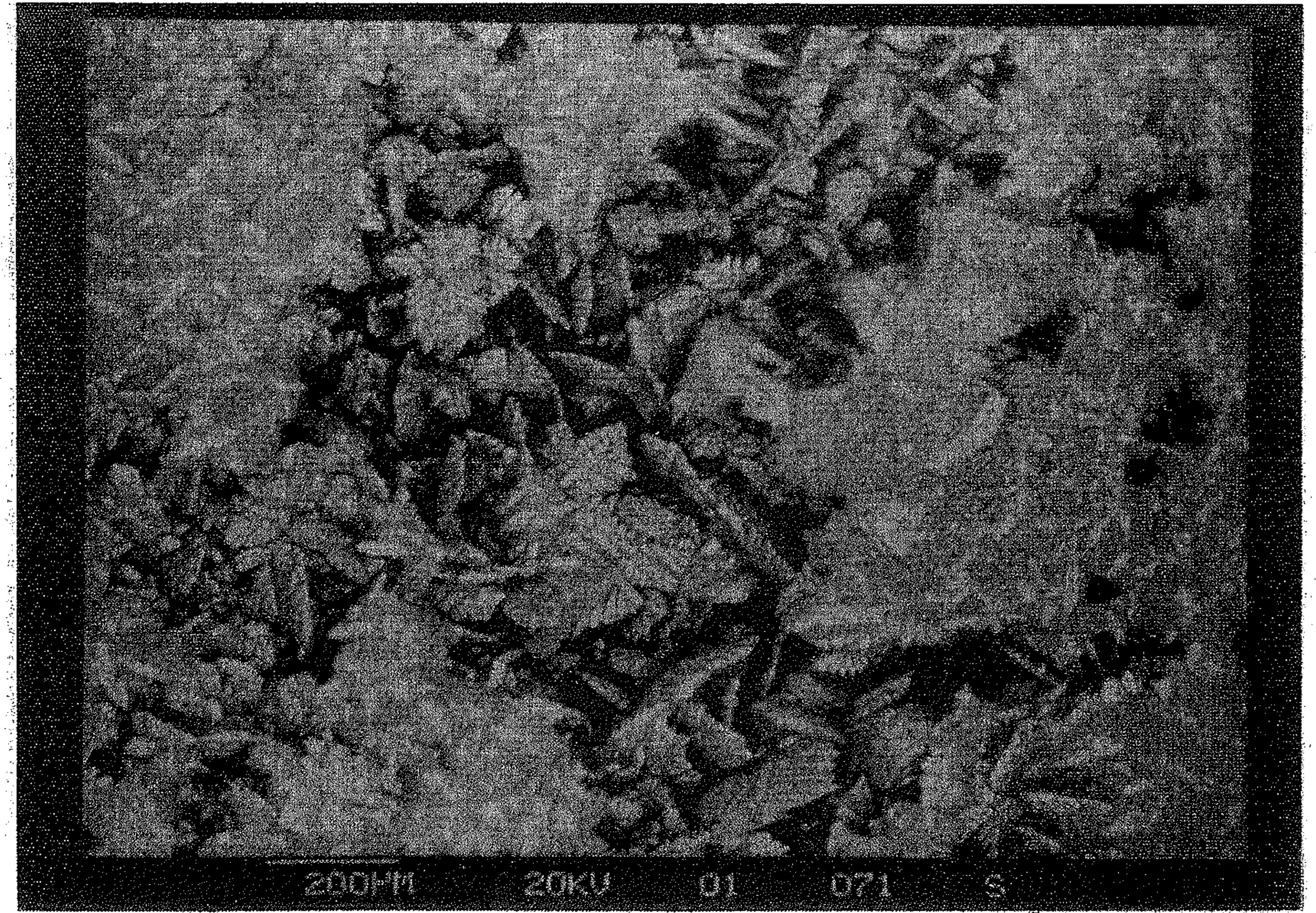

Chemische Zusammensetzung
41%
12,2%
1,3%
45,5%
Al2O3
Cr2O3
ZrO2
SiO2
Na2O+CaO+MgO
+Fe2O3 +TiO2
26%
13%
3,4%
26%
31,5%
Phasenbestand
41%
17%
42%
(Al, Cr)2O3 ss
Korund
Zirkoniumoxid
Glasphase
25%
19%
56%

25.10.89



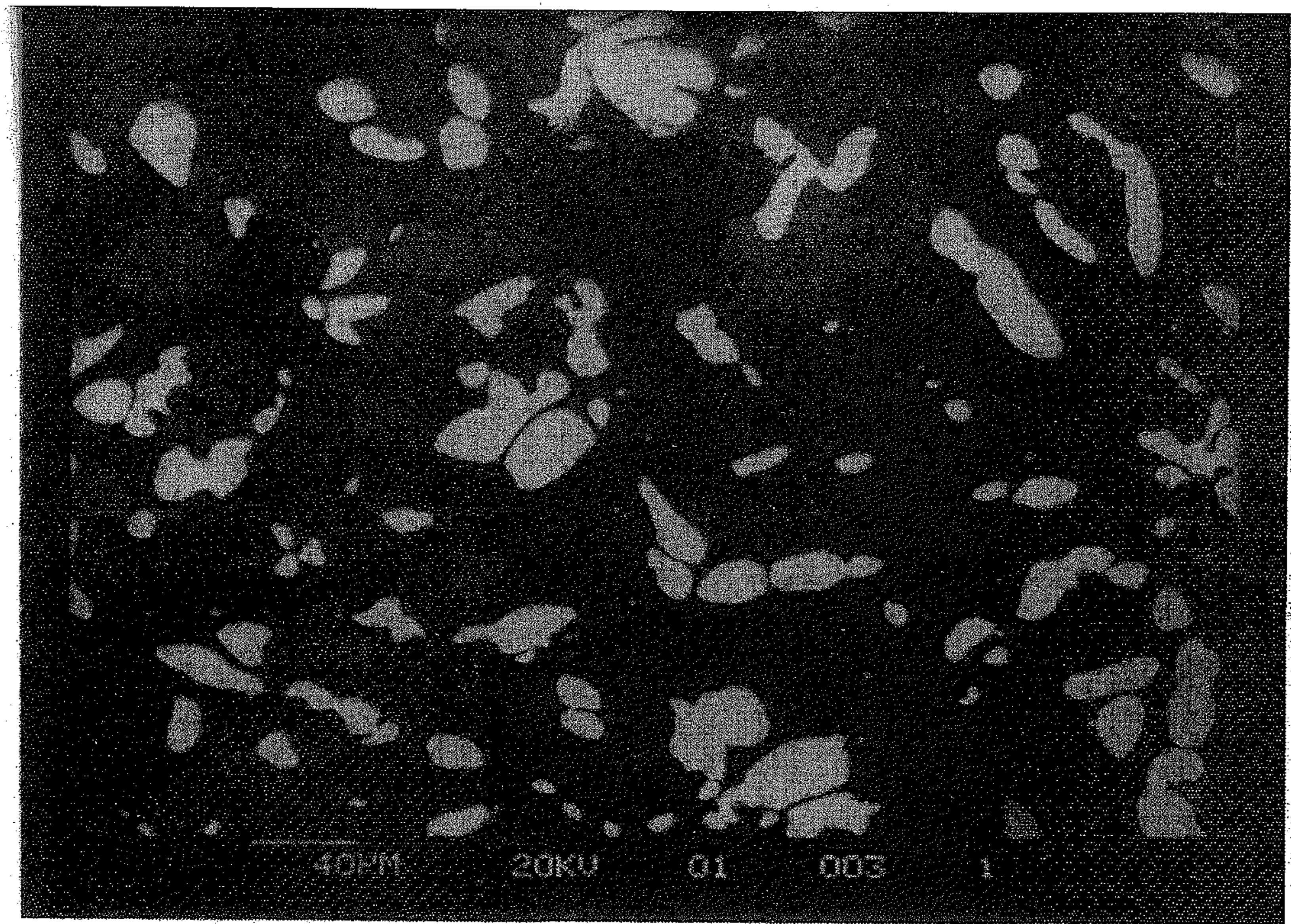
25.10.89

GP 98/12.2



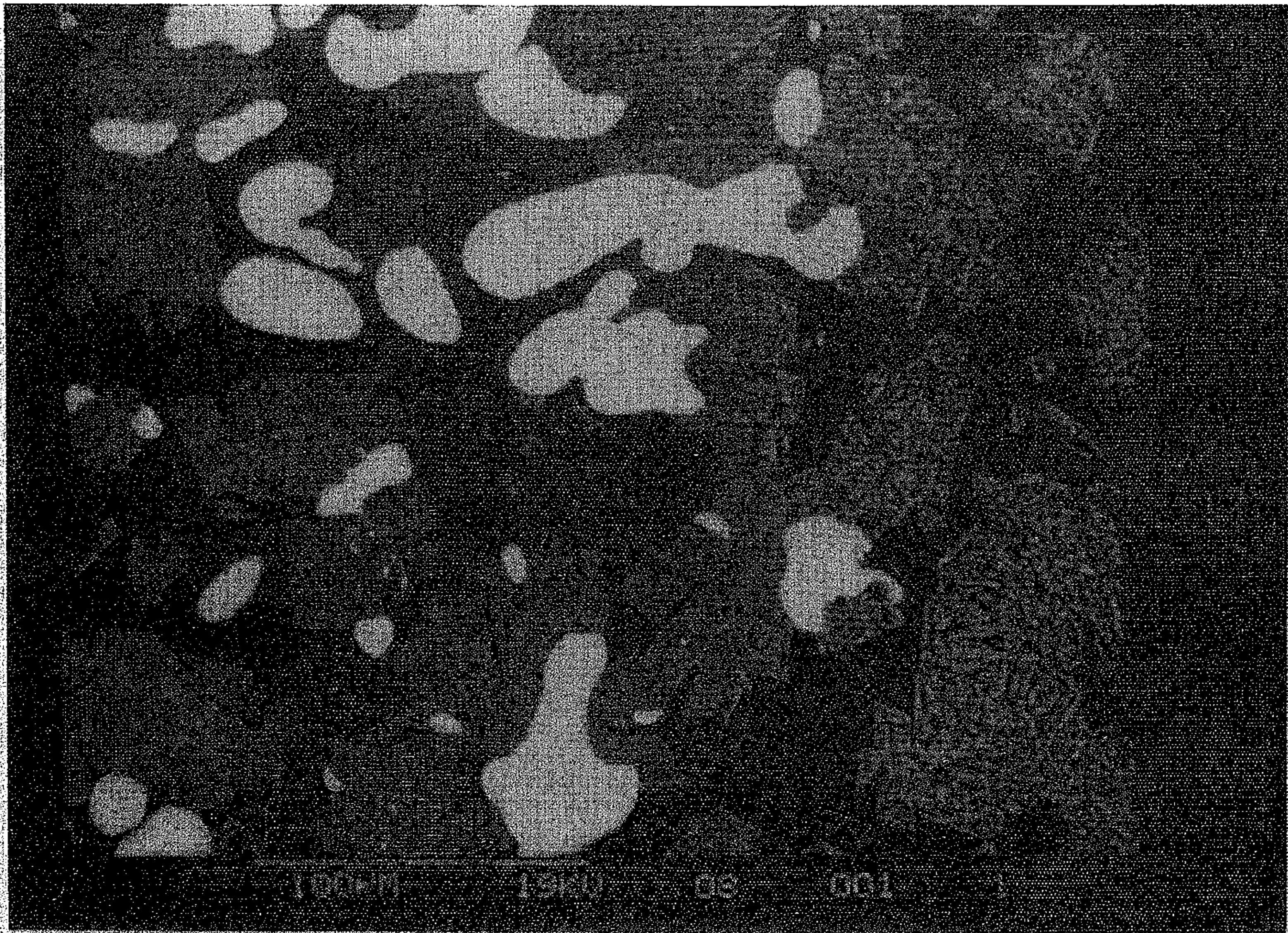
7.11.89 Pe 11

D 13 Texture of a fractured surface of ER 2161. ZrO_2 dendrites in pores



7.11.89 Pe 10

D 12 Texture of a polished sample of ER 2161. Porosity is due to shrinking of crystallizing melt

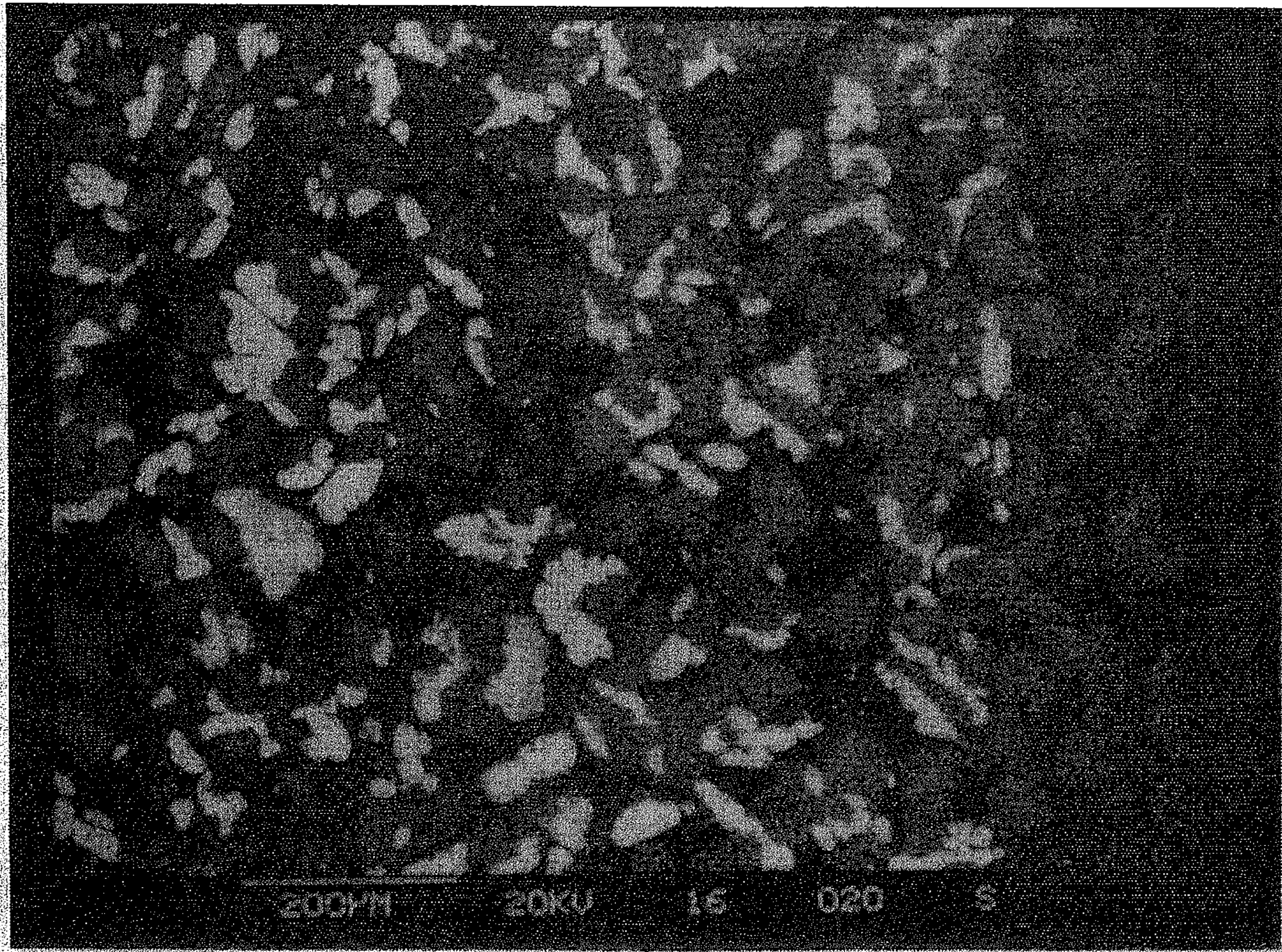


KfK

7.11.89 Pe13

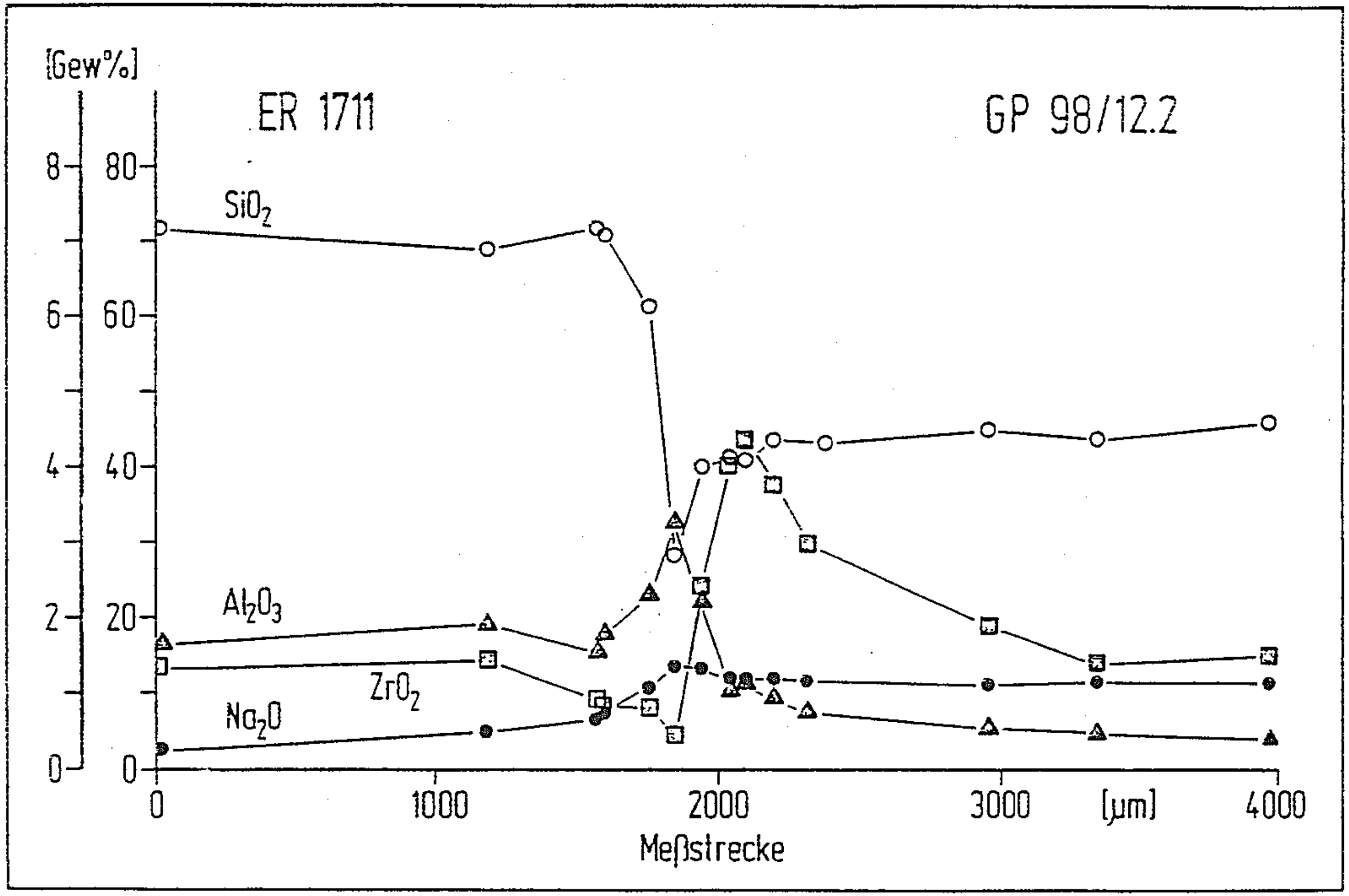
E 13 Corroded region of ER 2161, secondary Cr2O3-crystals outside in the melt (glass) (8/1000) RE

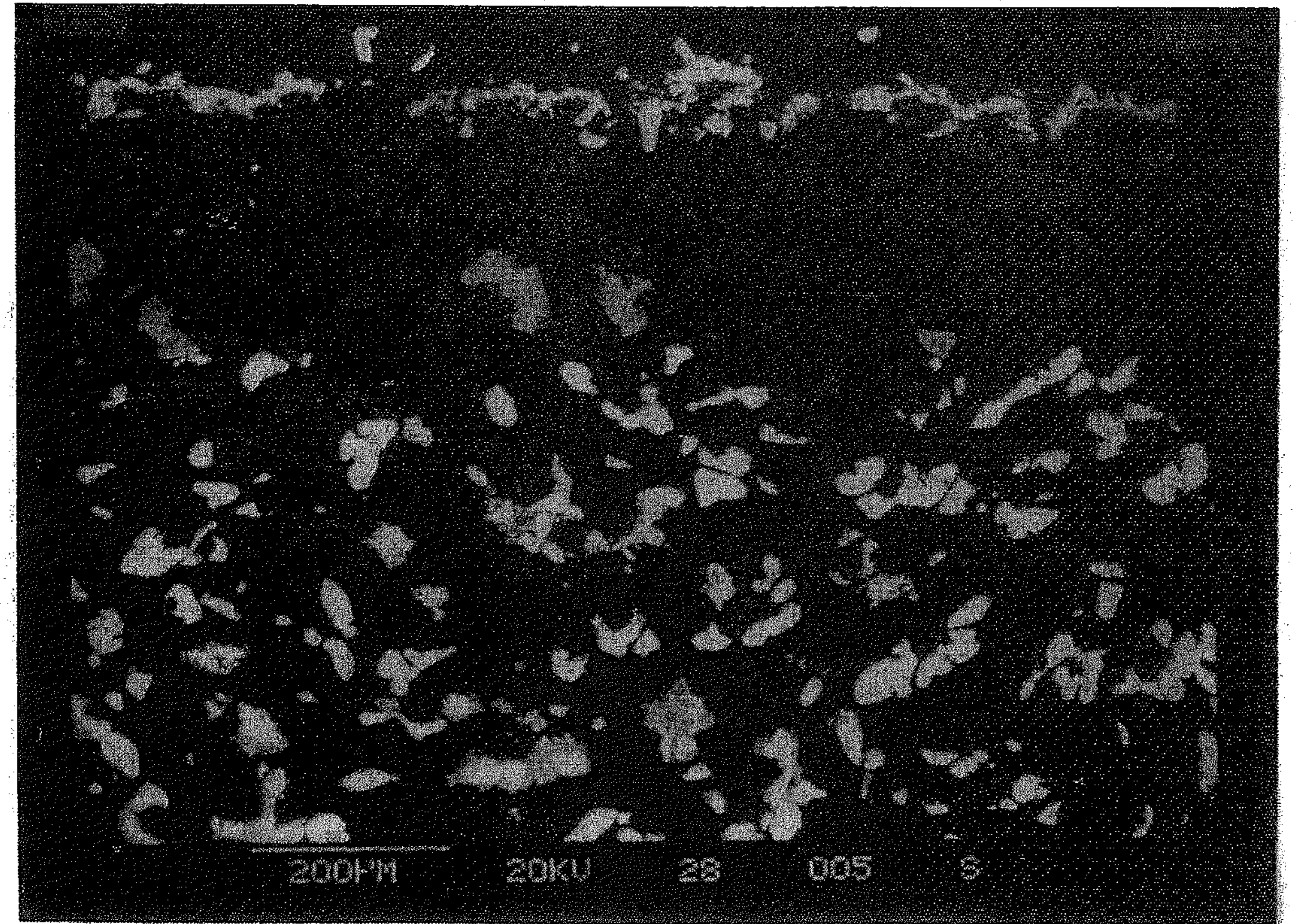
A10



7.11.89 Pe 14

E 14 Rim of corroded ER 2161 showing three different zones. RE


[Gew%]
ER 1711
GP 98/12.2
SiO2
Al2O3
ZrO2
Na2O
0
2
4
6
8
0
20
40
60
80
0
1000
2000
3000
[µm]
4000
Meßstrecke



8.11.89 Pe 23

E 18 ER 2161, corrosion after 128 days (1000°C); Zircon in zone (3) and between zones (1) and (3). Chevkinites on top (bottom of the crucible!)

Sekundär gebildete Phasen

		ER 1711		ER 2161	
	T [°C]	1000	1150	1000	1150
"Phase X"	$(Ca,Nd,Pr,Ce)_4(Ti,Zr)_4Cr[Si_2O_7]_2O_8$	4		2	
Eskolait	Cr_2O_3				2
Titanat	mit Ti,Zr,Ca,Ce,Fe	2			
Ceroxid	$(Ce,Zr)_xO_y$	3			
Baddeleyit	ZrO_2		1		3
Sodalith	$Ca_{8-x}Na_xAl_{12-x}Si_xO_{24}[(Mo,Cr)O_4]_2$	1			
Spinell	$(Al,Cr)_2(Mg,Fe,Ni)O_4$		2		4
"Rubin"	$(Al,Cr)_2O_3$		3		
Zirkon	$ZrSiO_4$			1	1

Basic features of ceramic/melt interactions

- The corrosion proceeds homogeneously

- The corrosion rate is low and determined by thermal gradients within the bricks, by the viscosity of Cr_2O_3-suspensions on their surfaces and by Zr^{4+}- and $Cr3^{+}$-concentrations in the melt originating from the waste

- Reaction path for the corrosion is the amorphous phase (glass/melt) in the ceramic. Transport of matter is by diffusion

- Secondary phases are formed within the corroding rim

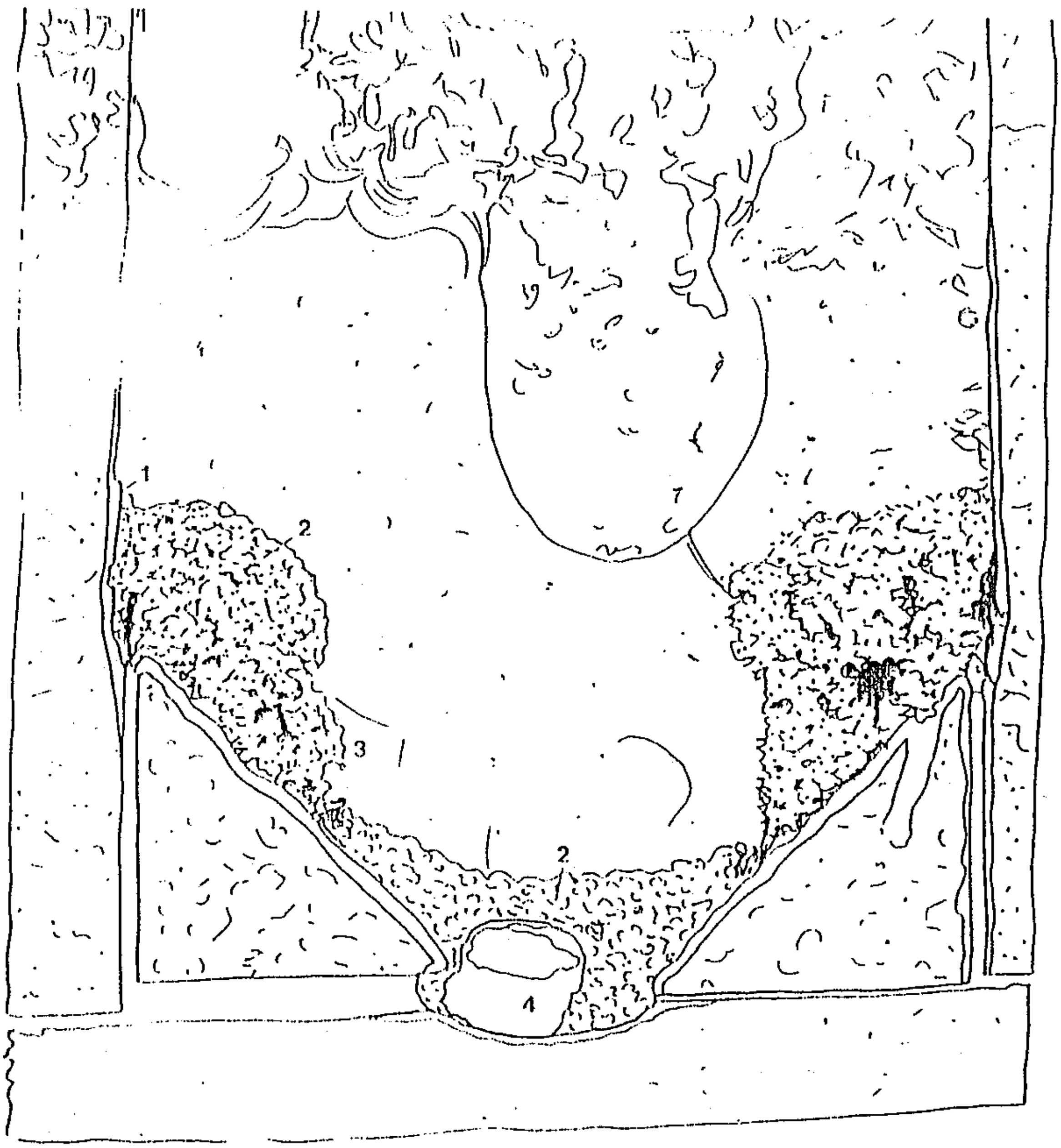


Abb. F 2 *Schematische Darstellung eines Schnittes durch einen mit Glasprodukt gefüllten Laborschmelzer.*
Die gelbe Phase am Boden des Ofens hat in KR 2161 Tropfenbohren verursacht. Die große Blase in der Mitte des Glasproduktes ist auf die Volumenverringerung beim Abkühlen zurückzuführen.
(1) Cr_2O_3-Schicht,
(2) lockere Suspension von RuO_2 und Edelmetalltelluriden,
(3) gealterte Suspension von RuO_2 und Edelmetalltelluriden,
(4) gelbe Phase.

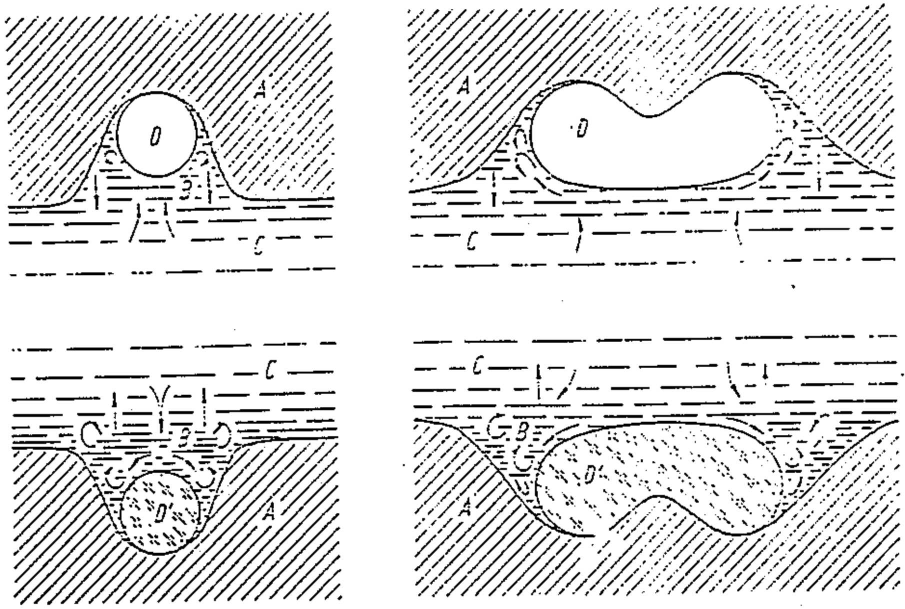


Bild 7.8. Schematische Darstellung der bevorzugten Korrosion in vertikaler Richtung durch Grenzflächenkonvektion um kleine und große Gasblasen und Flüssigkeitstropfen (Zapfenbildung bei großen Blasen und Tropfen). *A* Festkörper, *C* korrodierende Flüssigkeit bzw. Schmelze, *B* Lösung aus *A* und *C*, *D* Gasblase, *D'* Mit *C* nicht oder nicht vollständig mischbarer Flüssigkeitstropfen

8.11.89 Pe 24

B 2 Schema of corrosion by interfacial convection around bubbles (D) and drops (D')

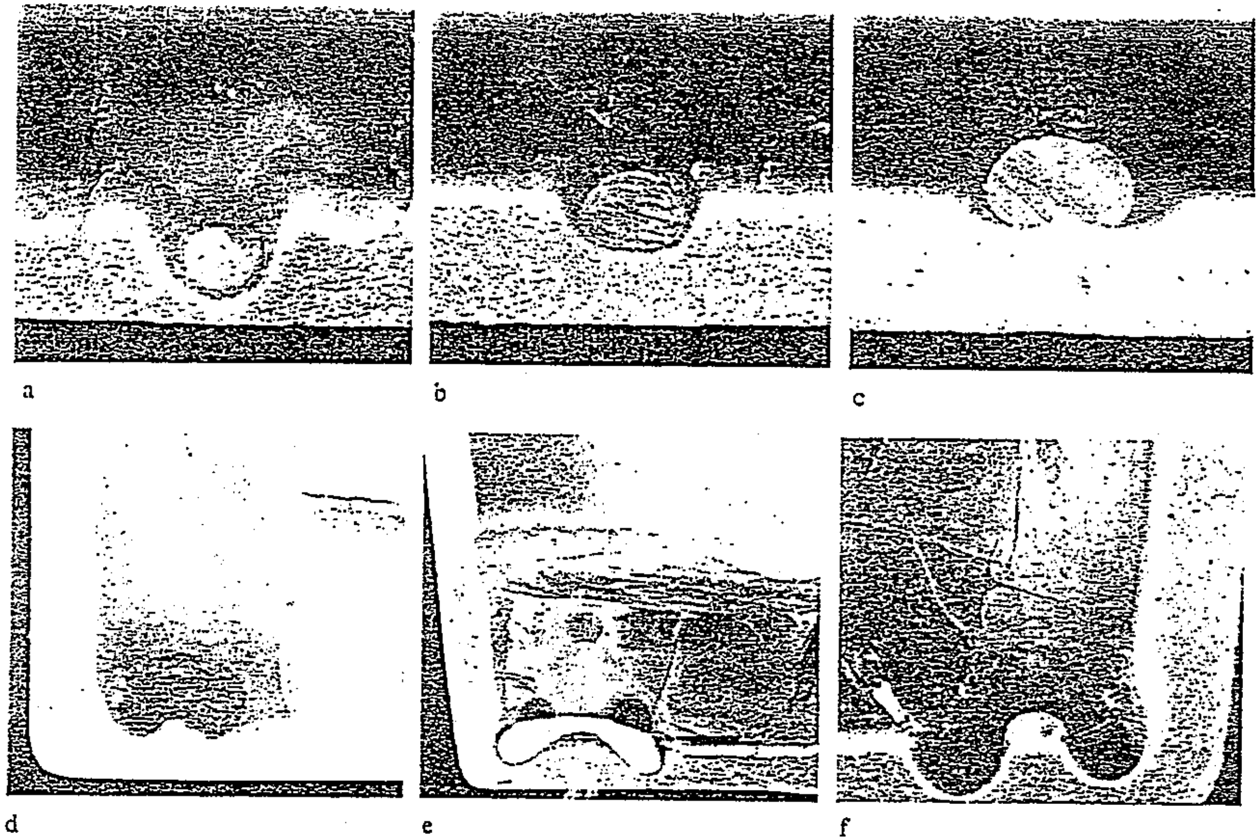


Bild 7.11 a–f. Korrosionsprofil bei abwärts gerichteter bevorzugter Korrosion von SKX-Tiegeln und eines Sinterkorundtiegels (d) durch Grenzflächenkonvektion. Temperatur: 1400°C, Zeit: 6 bis 8 h. a kleiner Silbertropfen; b kleiner Goldtropfen; c Goldtropfen, bei dessen Größe gerade Zapfenbildung einsetzt; d Goldtropfen mit Zapfenbildung im Sinterkorundtiegel; e großer Silbertropfen; f großer Bleitropfen, der sich so tief in den Tiegelboden eingefressen hat, daß er Ringform angenommen hat

KfK

8.11.89 Pe 20

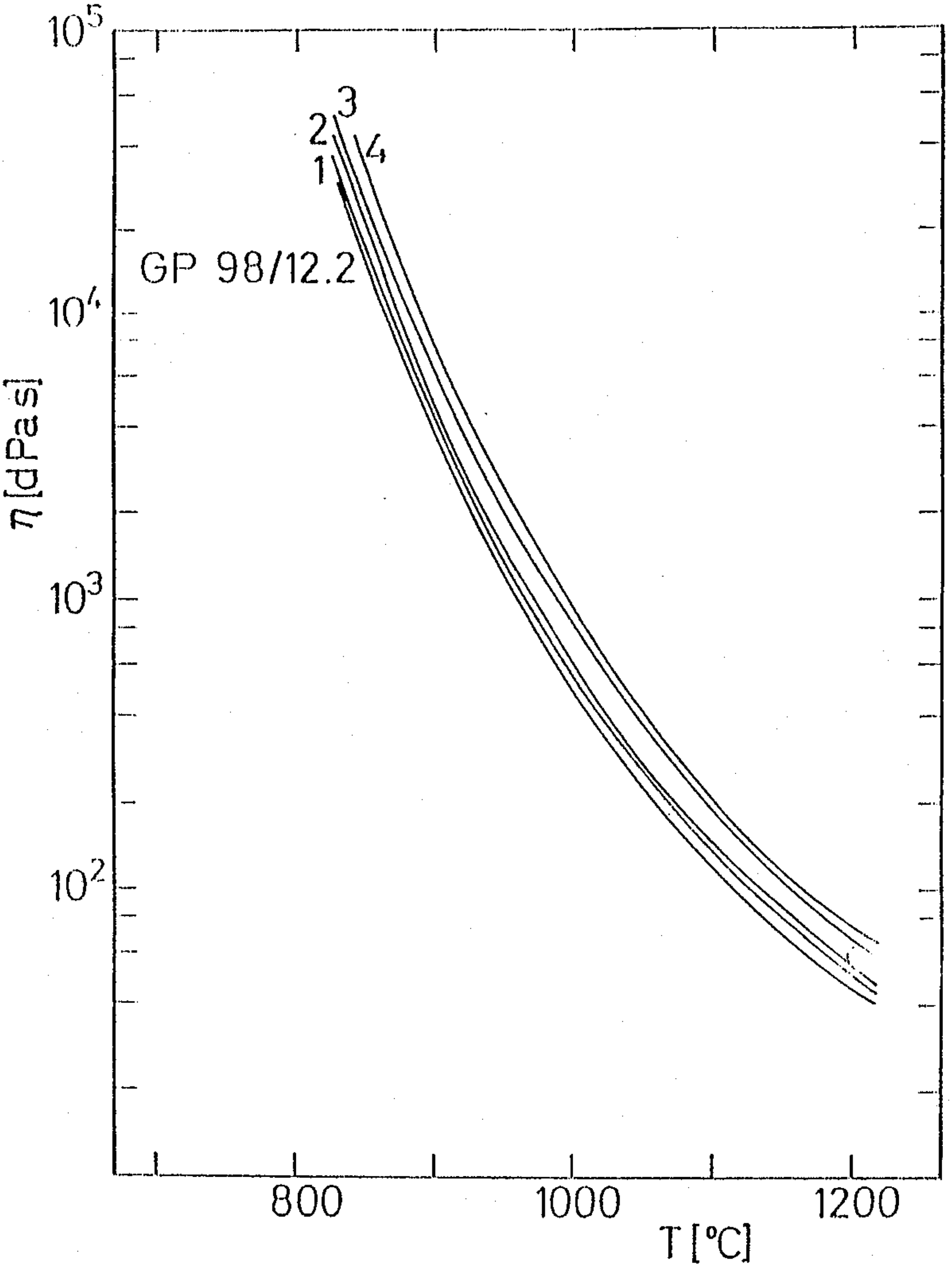
Corrosion profiles due to interfacial convection by metallic drops of different size and/or composition: a,e = Ag; b,c,d = Au; f = Pb

A 19

Sample	wt% Cr_2O_3	Size (μm)	Φ calc.
1	5 (3)	5	2.7 (1.5)
2	5 (3)	0.4	2.7 (1.5)
3	10 (8)	5	5.6 (3.5)
4	10 (8)	0.4	5.6 (3.5)

KfK 30.10.89

Dilute Cr_2O_3-Suspension in GP 98/12



KfK 8.11.89 Pe 28

F 1 η = F(T); GP 98/12.2 and Cr_2O_3-Suspensions

(1) 5 wt%, 5 µm; (2) 5 wt% 0.4 µm; (3) 10 wt%, 5 µm; (4) 10 wt%, 0,4 µm

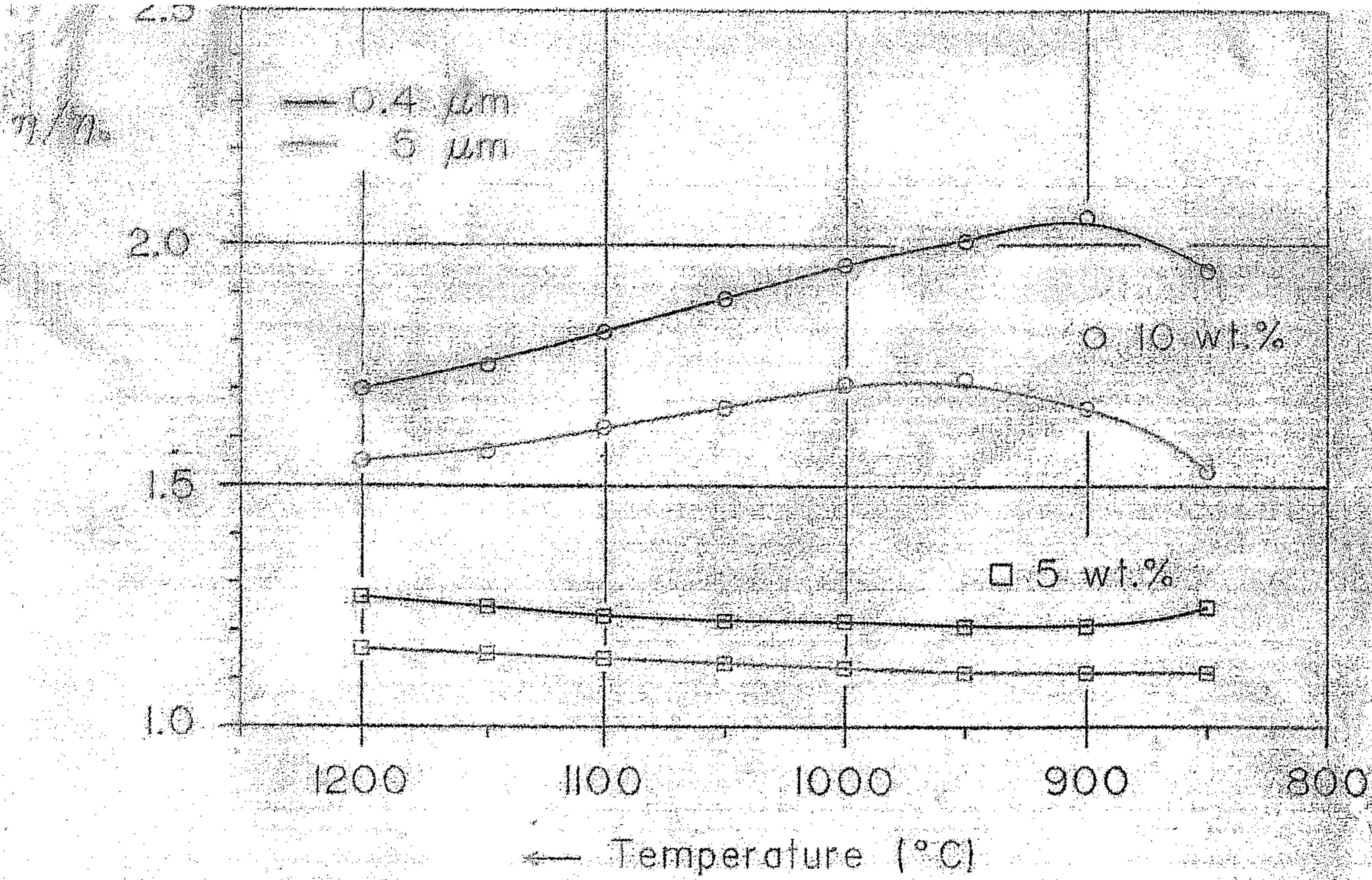
Sample	T (°C)	1200	1150	1100	1050	1000	950	900	850
1	η/η_o	1.159	1.145	1.144	1.133	1.121	1.105	1.081	1.049
2	η/η_o	1.273	1.246	1.231	1.221	1.217	1.213	1.220	1.246
3	η/η_o	1.545	1.565	1.620	1.668	1.709	1.718	1.672	1.530
4	η/η_o	1.705	1.754	1.818	1.889	1.957	2.006	2.020	1.947

KfK

30.10.89

Dilute Cr_2O_3-Suspensions in GP 98/12

A 21



KfK

Cr_2O_3 - Suspension in glass product GP98/12.2

A22

3.11.89 Pe 34 A 23

Cr_2O_3-Suspensions, texture 5wt% 5µm

A24

Cr_2O_3-Suspensions, texture 5wt%, 0.4 mm

A25

KfK

Cr_2O_3-Suspensions, texture 10 wt%, 5 μm

A26

KfK

8.11.89 Pe 34

Cr_2O_3-Suspensions, texture 10 wt% , 0.4 um

Einstein's Law of Viscosity: Experimental

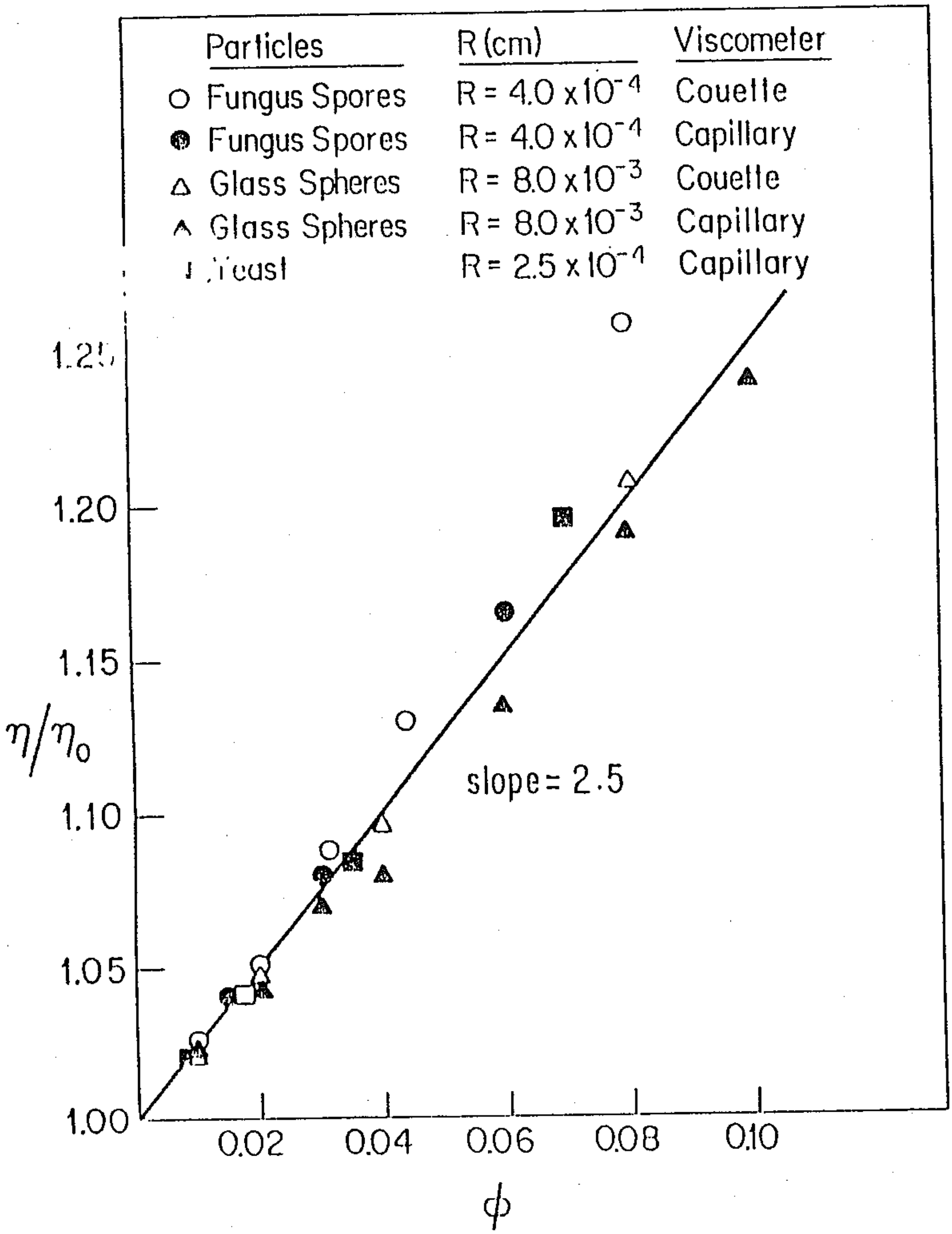


FIGURE 2.11 *Experimental verification of Einstein's law of viscosity for spherical particles of several different sizes and materials. [Data from F. Eirich, M. Bunzl, and H. Margaretha,*

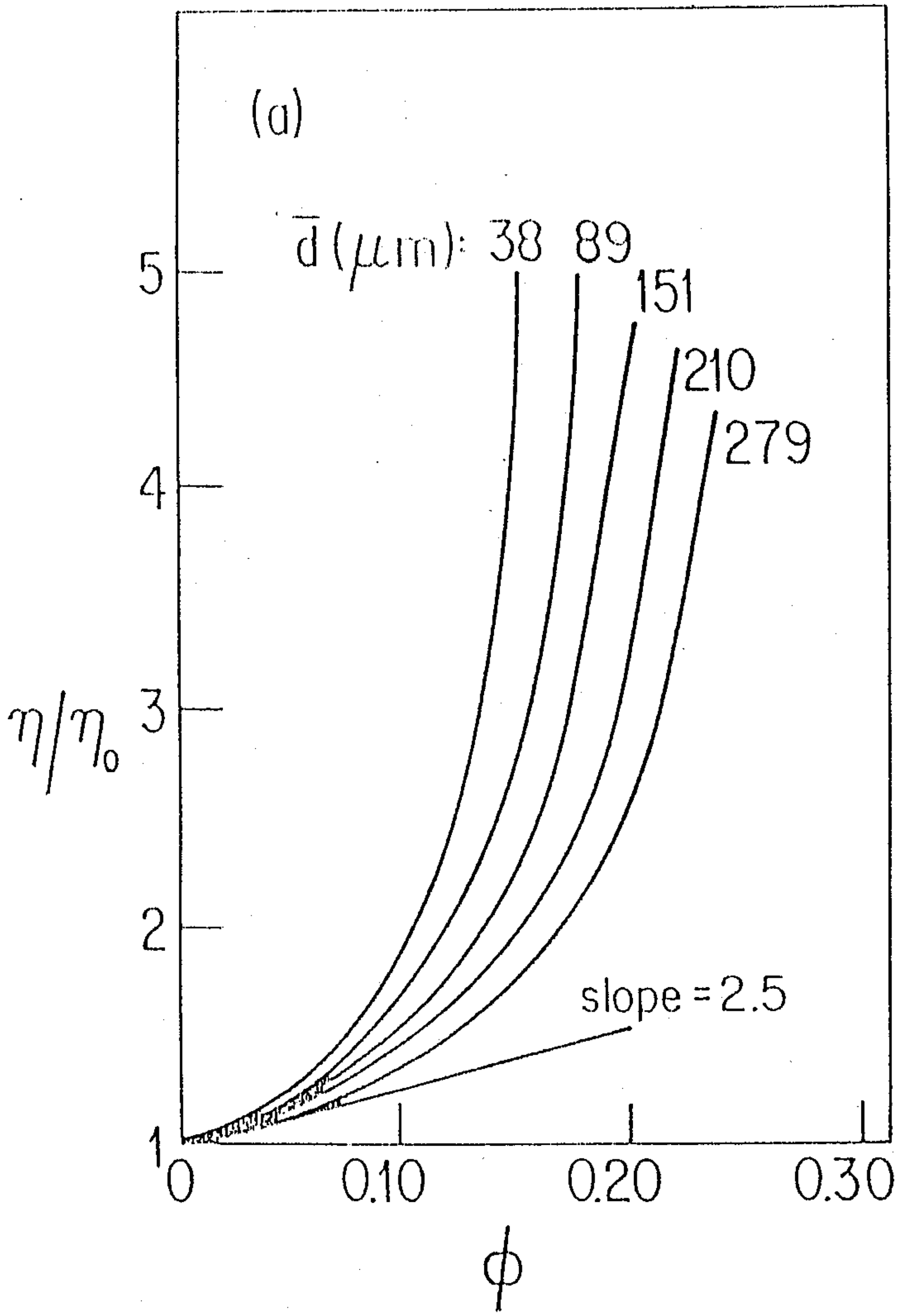
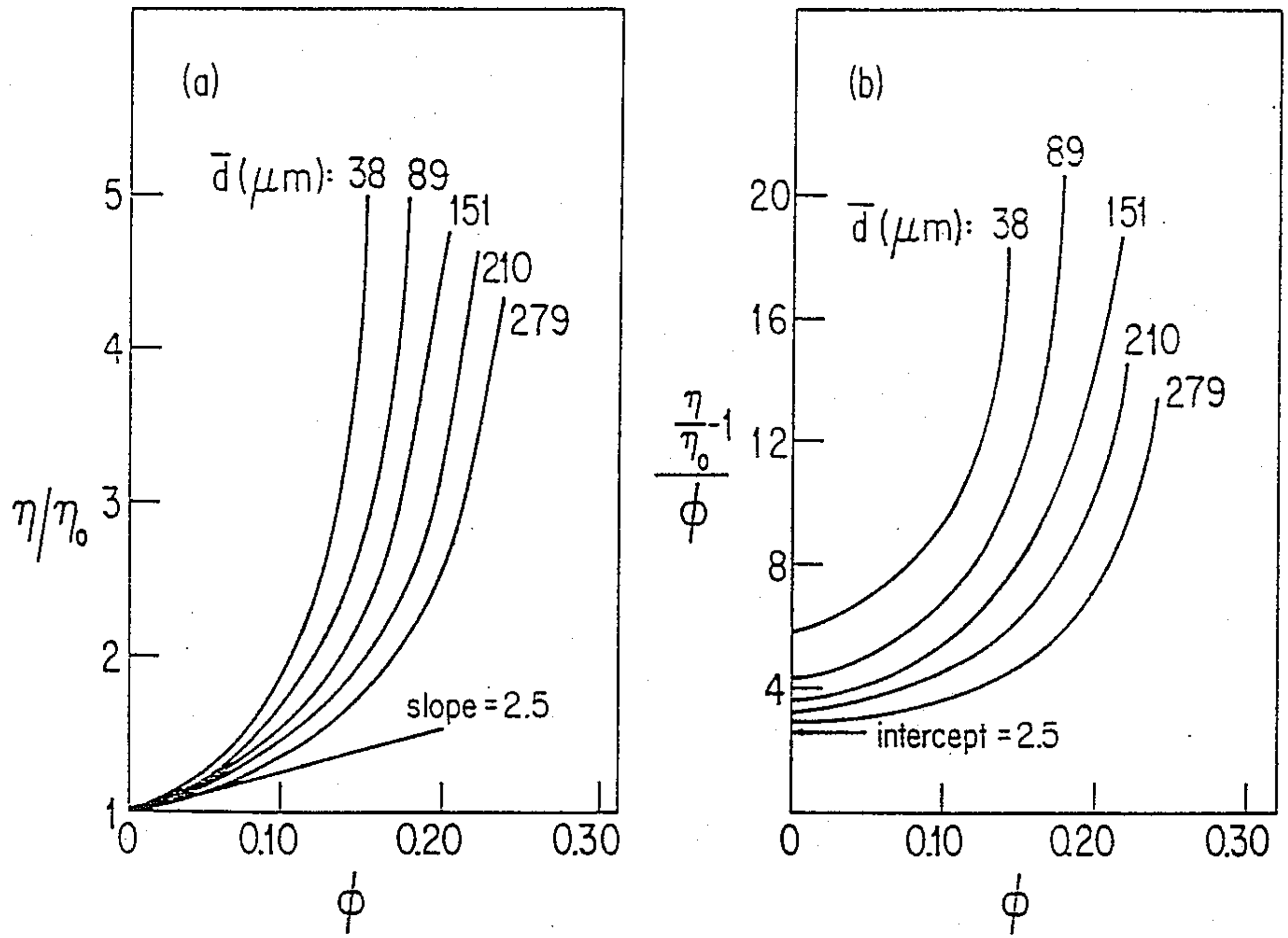

(a)
d̄ (μm): 38 89
151
210
279
5
4
η/η_0 3
2
1
slope = 2.5
0
0.10
0.20
0.30
ϕ

FIGURE 2.13 *(a) Relative viscosity and (b) specific viscosity versus volume fraction for polymethyl methacrylate particles having the average diameters shown. [Data from S. G. Ward and R. L. Whitmore,* Brit. J. Appl. Phys. **1**:325 (1950).]



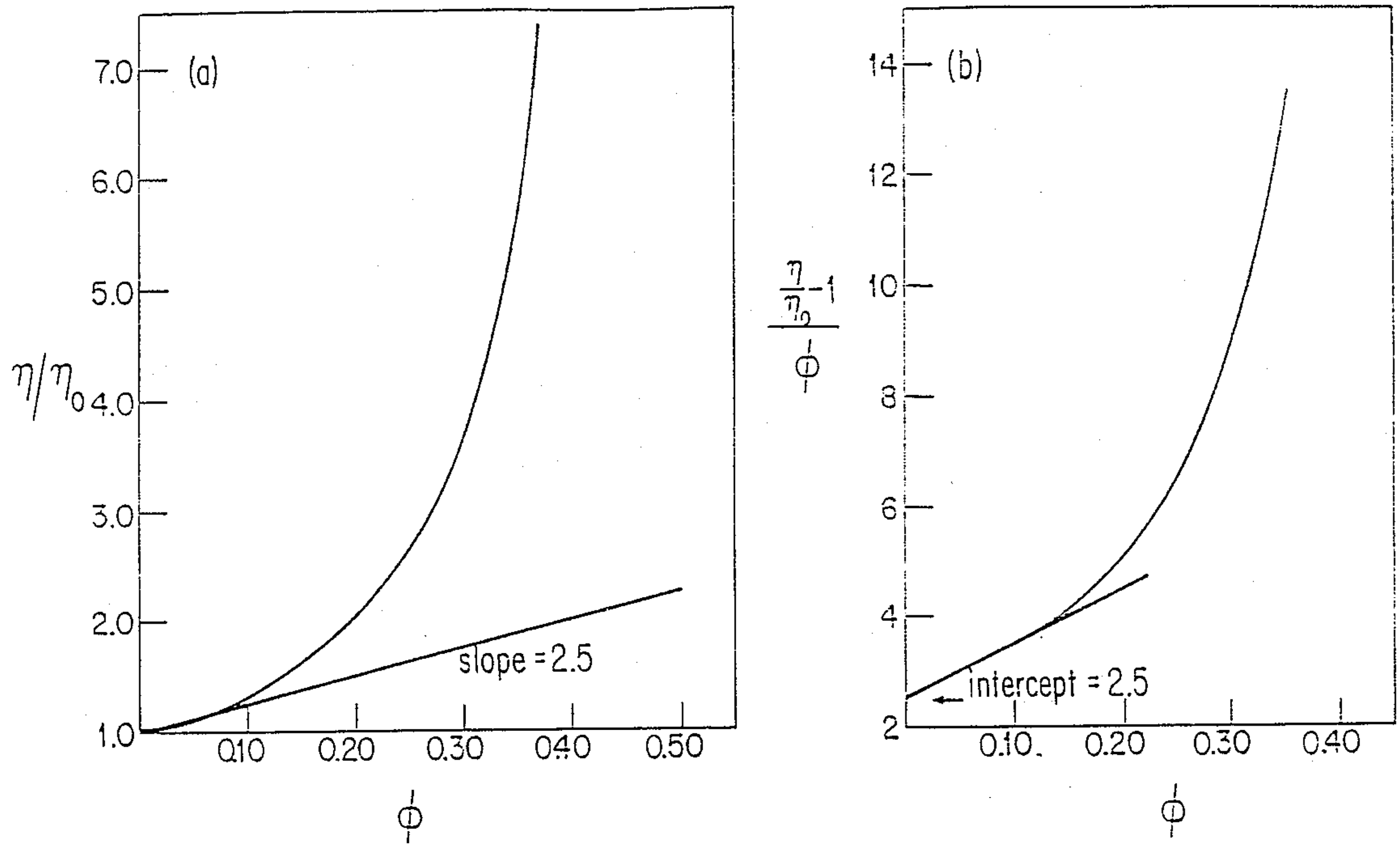
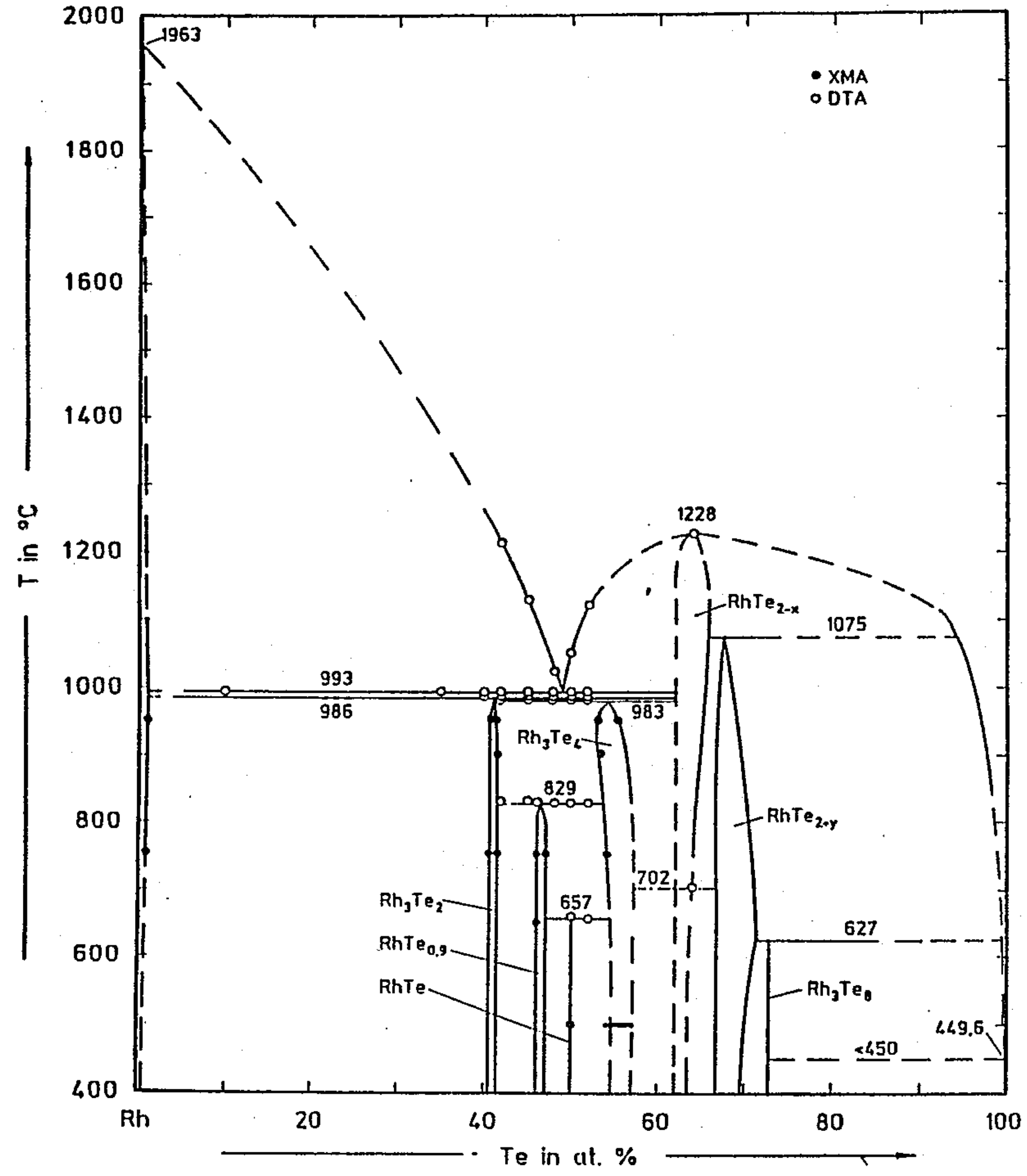
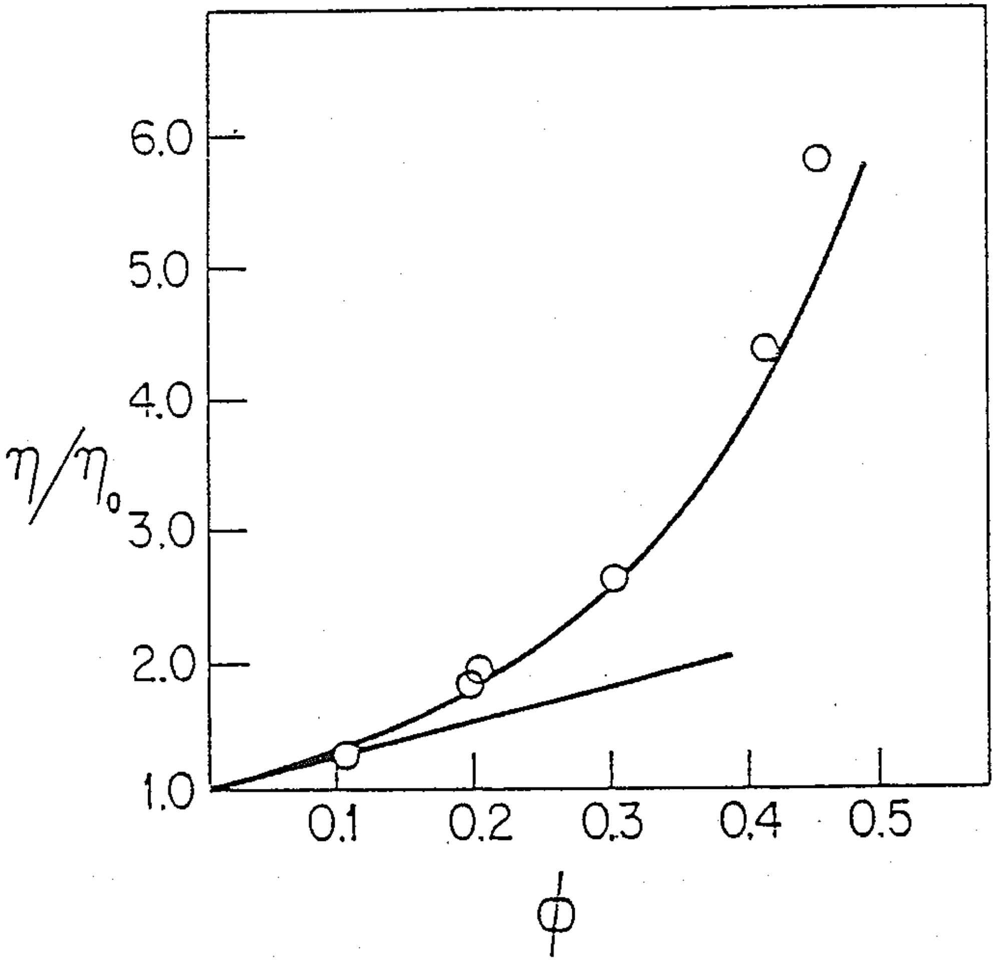


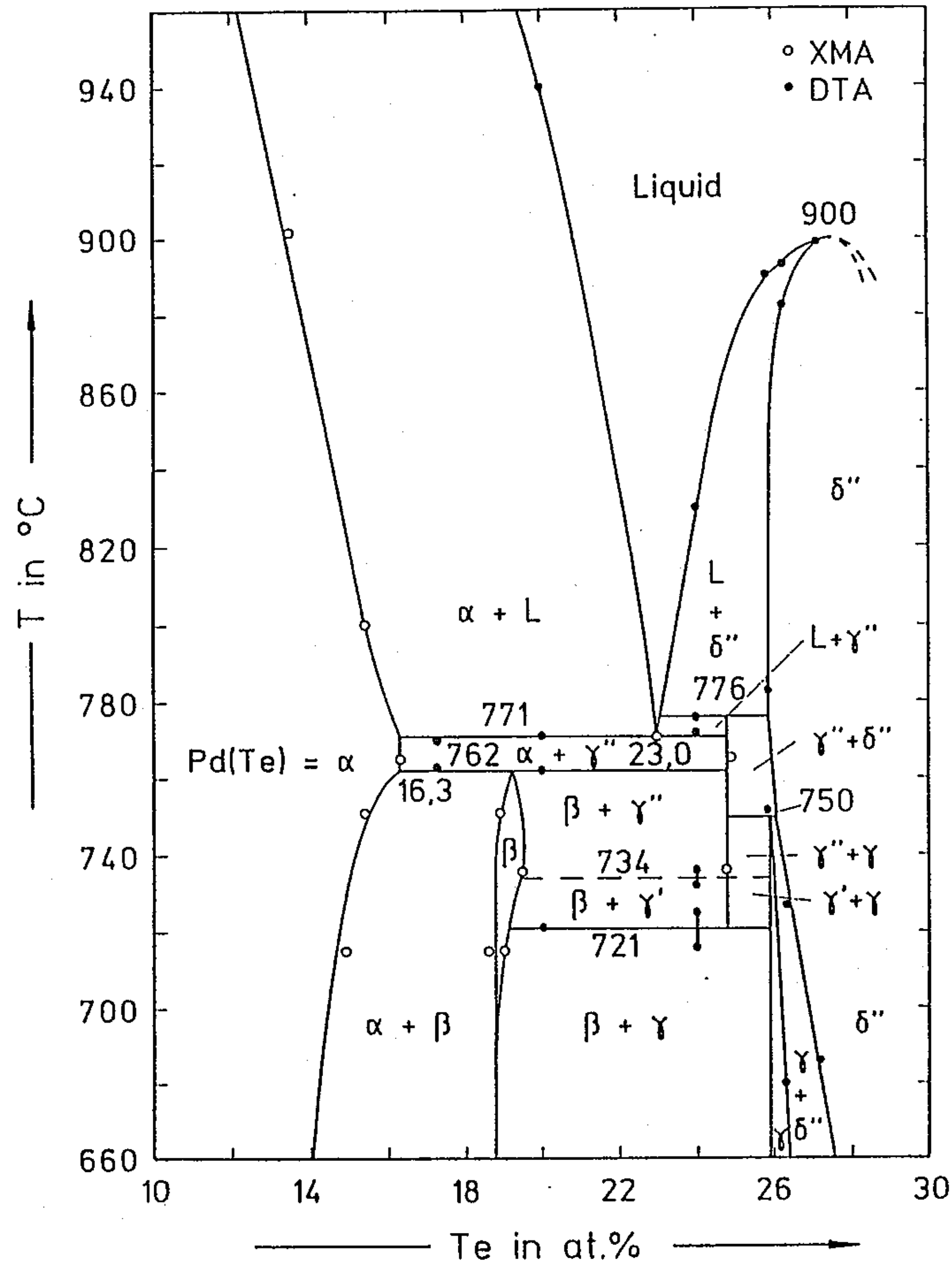
FIGURE 2.12 (*a*) *Relative viscosity versus volume fraction for a dispersion of glass spheres* ($R = 6.5 \times 10^{-3}$ *cm*) *up to* $\phi \simeq 0.40$. (*b*) *Same data plotted as reduced viscosity versus* ϕ. [*Data from V. Vand*, J. Phys. Colloid Chem. **52**:300 (1948).]



Phase diagram of the Rh - Te system

FIGURE 2.14 *The effect of polydispersity on viscosity. Solid line drawn according to Eq.* (70); *points are experimental results.* [*Data from R. Roscoe*, Brit. J. Appl. Phys. **3**:267 (





Detail of the phase diagram of the Pd - Te system

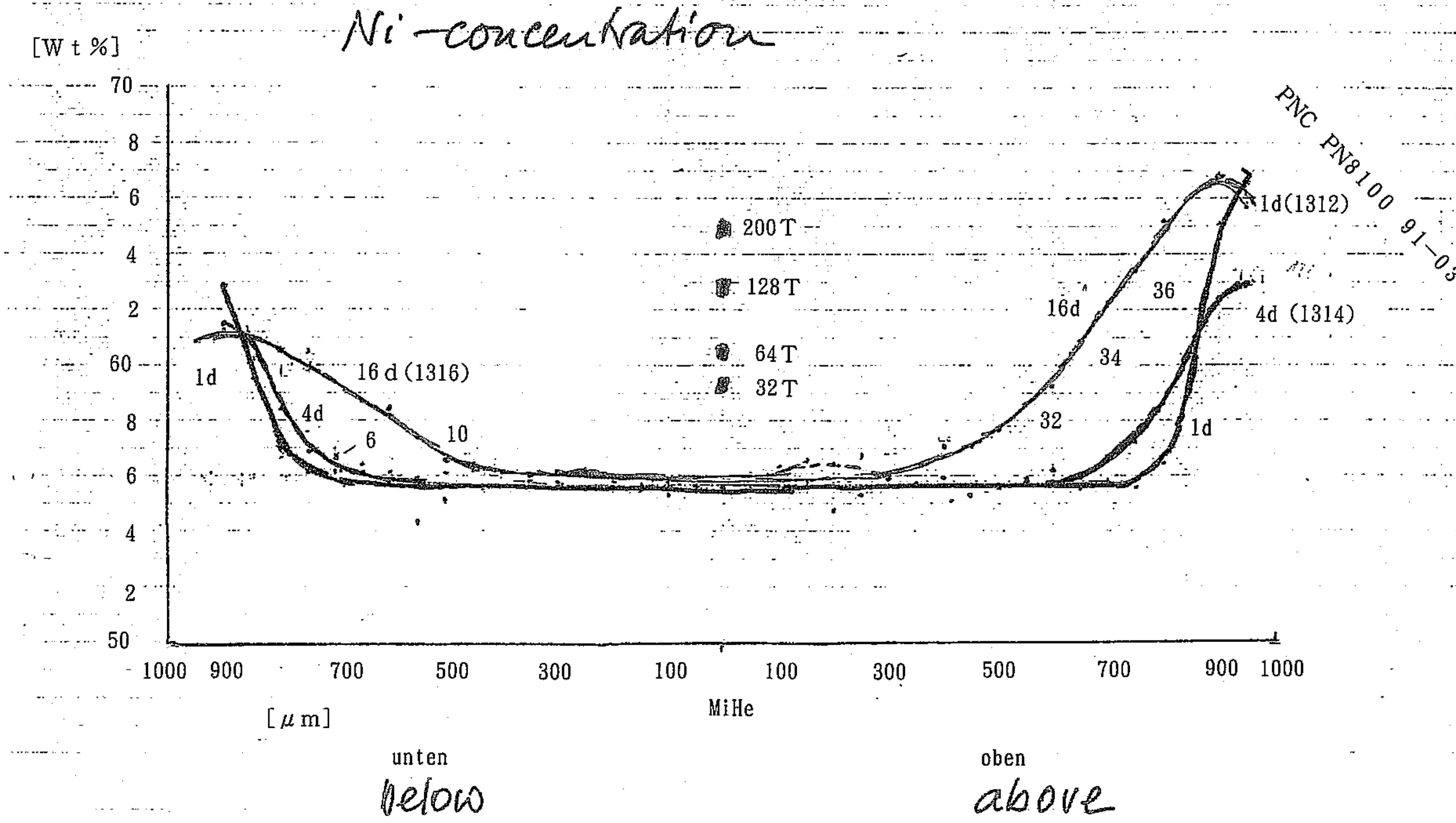
Conclusious

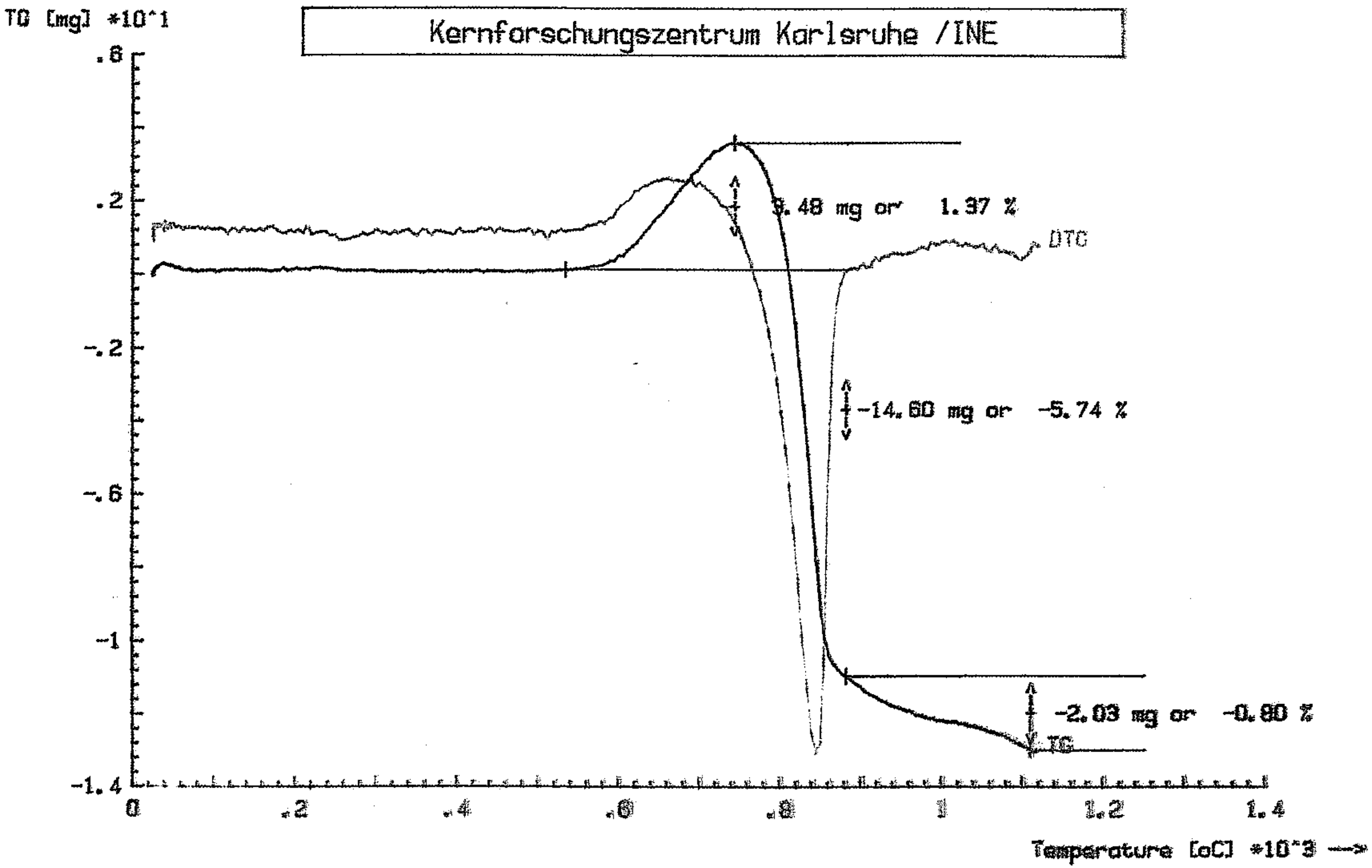
—INCONEL 690 alloy is being seleclively
Oxidiled in juch away that co and some 50% of the Fe(not shown here) leave the alloy appvethy as a function of oxygen availulilty

—Fe and Cr are being dinolvel in He glon poduct melt

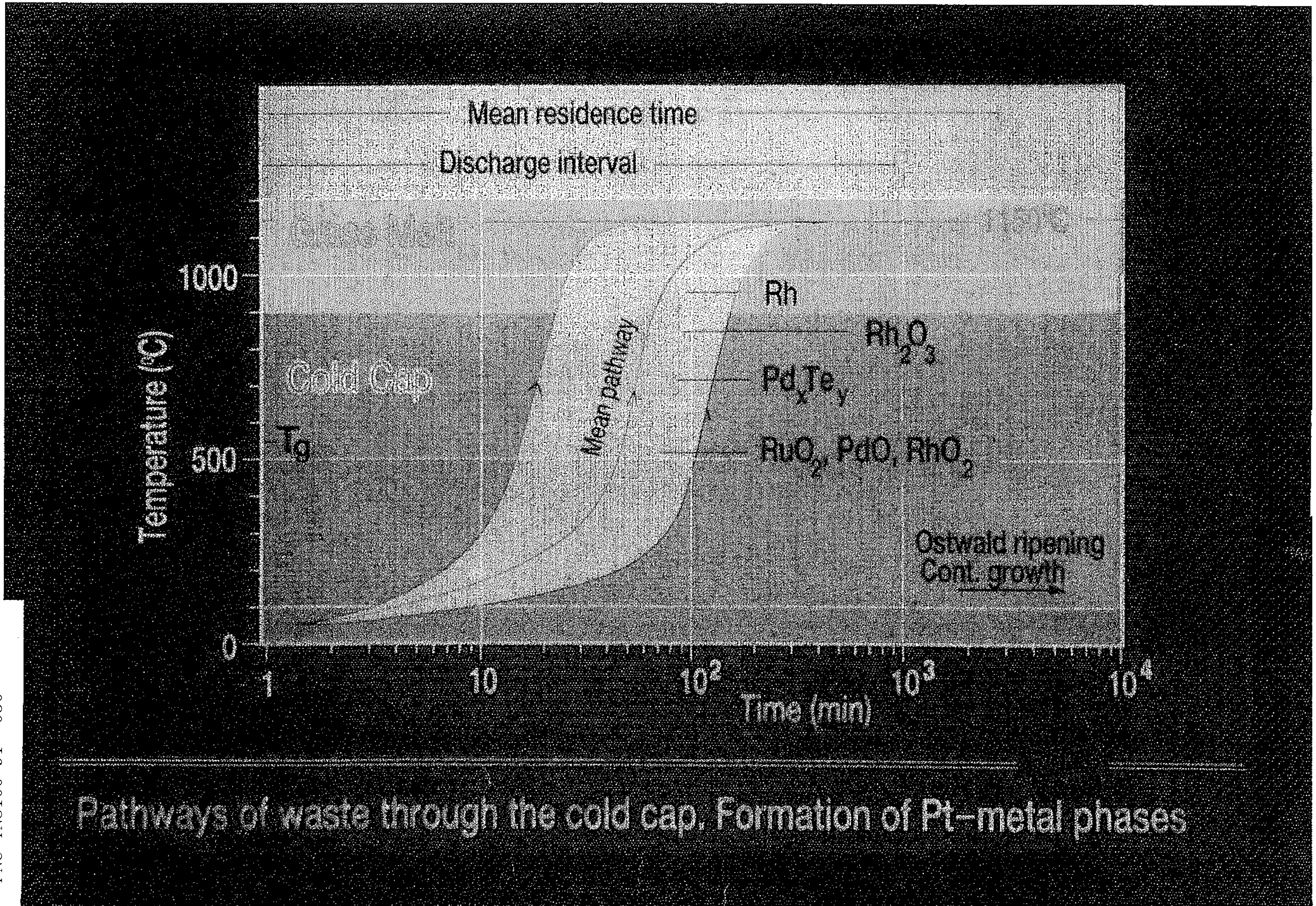
—Cr overatuation in the glon melt leads to a Hcordary fovmation of Cr_2O_3

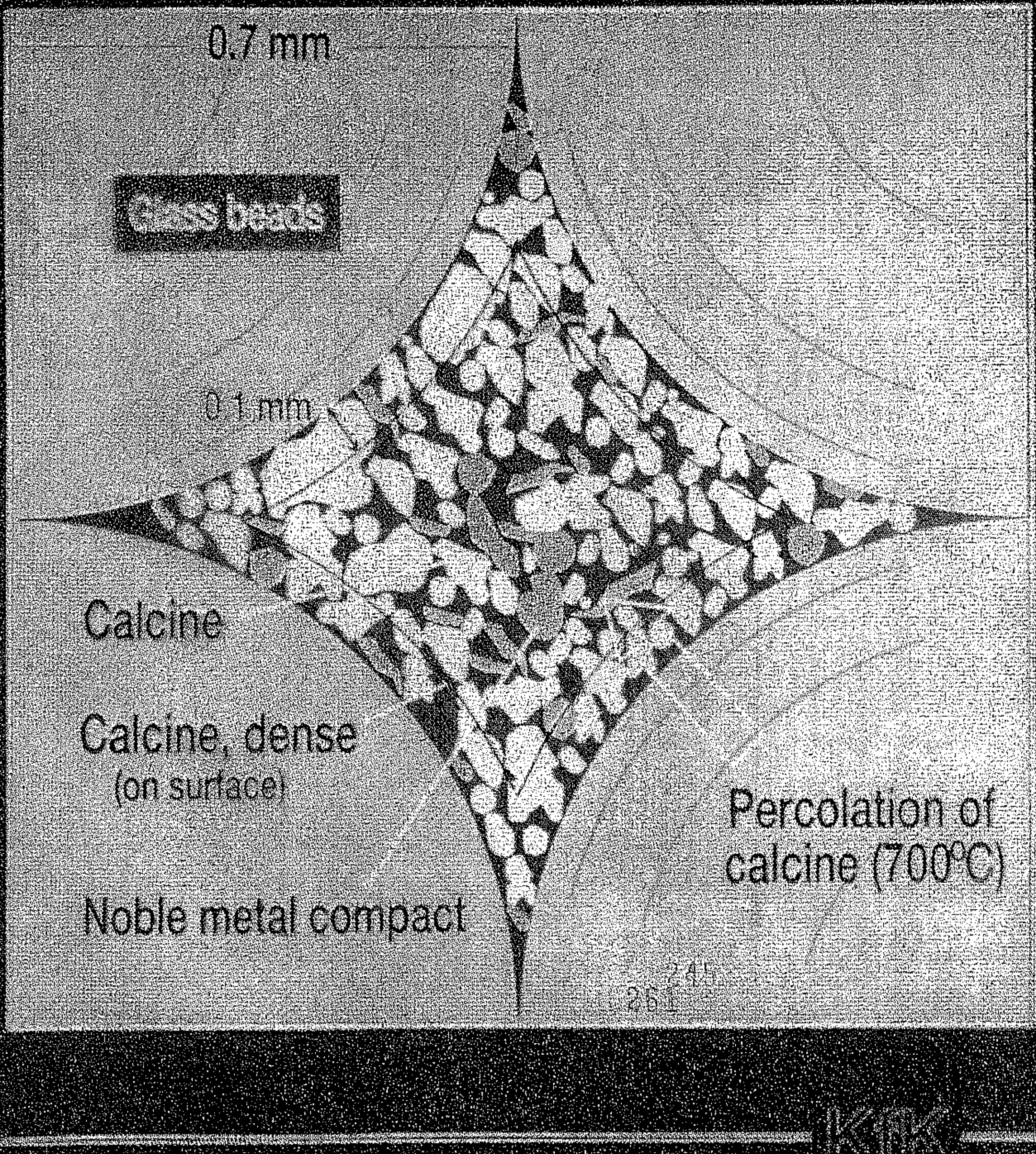
—Salecfive oxidafion Kiuefies are influenced by Cr-CowcenHafion in Hre melt


Ni-concentration
[Wt %]
70
8
6
4
2
60
8
6
4
2
50
1d
16 d (1316)
4d
6
10
200 T
128 T
64 T
32 T
16d
36
34
32
1d(1312)
4d (1314)
1d
PNC PN8100 91—030
1000 900 700 500 300 100 100 300 500 700 900 1000
[μm]
MiHe
unten
below
oben
above

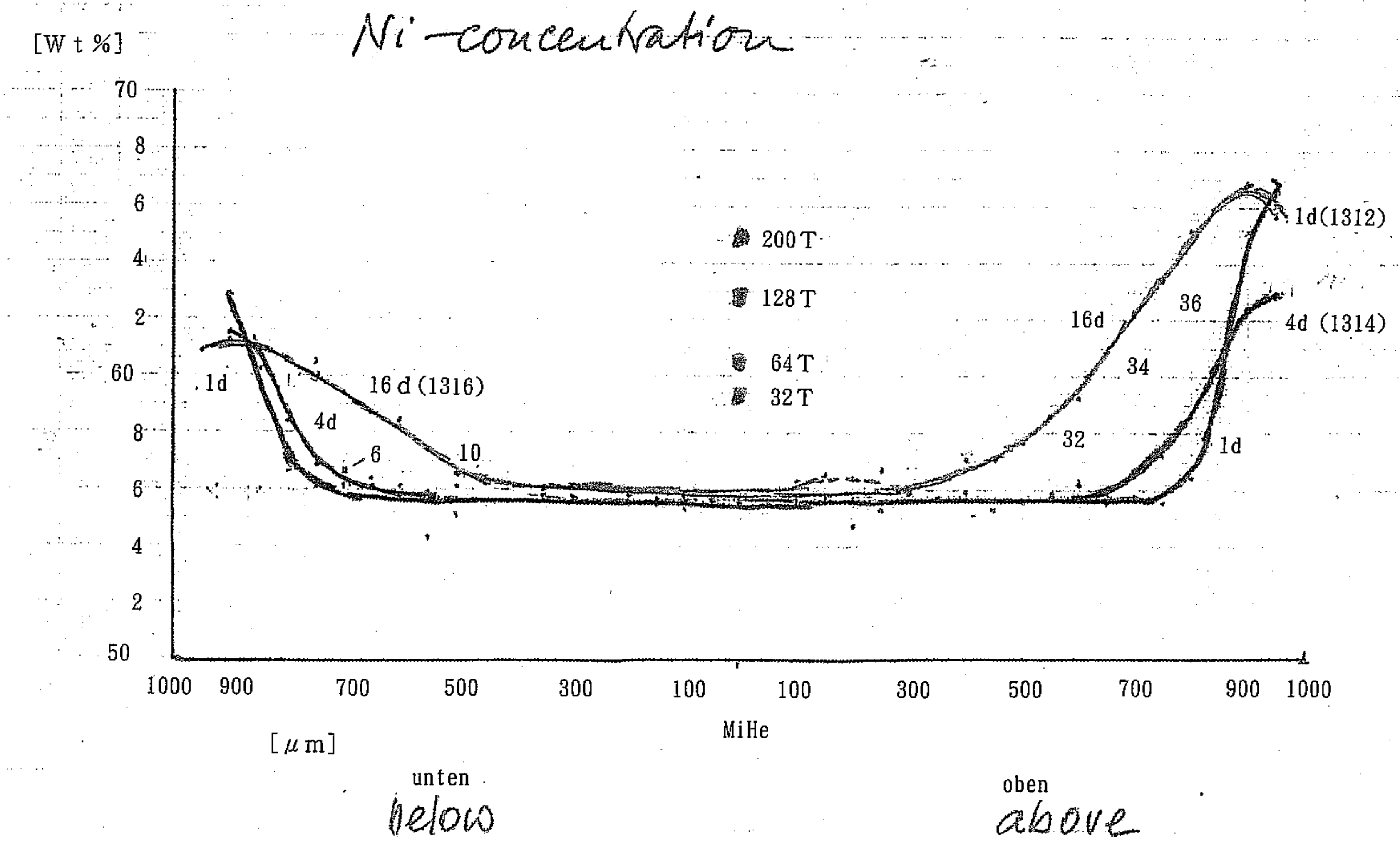


LABORATORY:	INE	SAMPLE:	PdO/TeO2	254.4 mg	RANGE: DTA	100.0 uV
OPERATOR:	NM	REFERENCE:	AL203	245.8 mg	RANGE: TG	250.0 mg
IDENTITY No.:	464	ATMOSPHERE:	LUFT	0.0 ccm/min	RANGE: DTG	200.0 uV (3)
DATE:	15.3.89	CRUCIBLE:	AL203	---> SEGMENT 1 /	(10.0 K/min	Corr. K3)

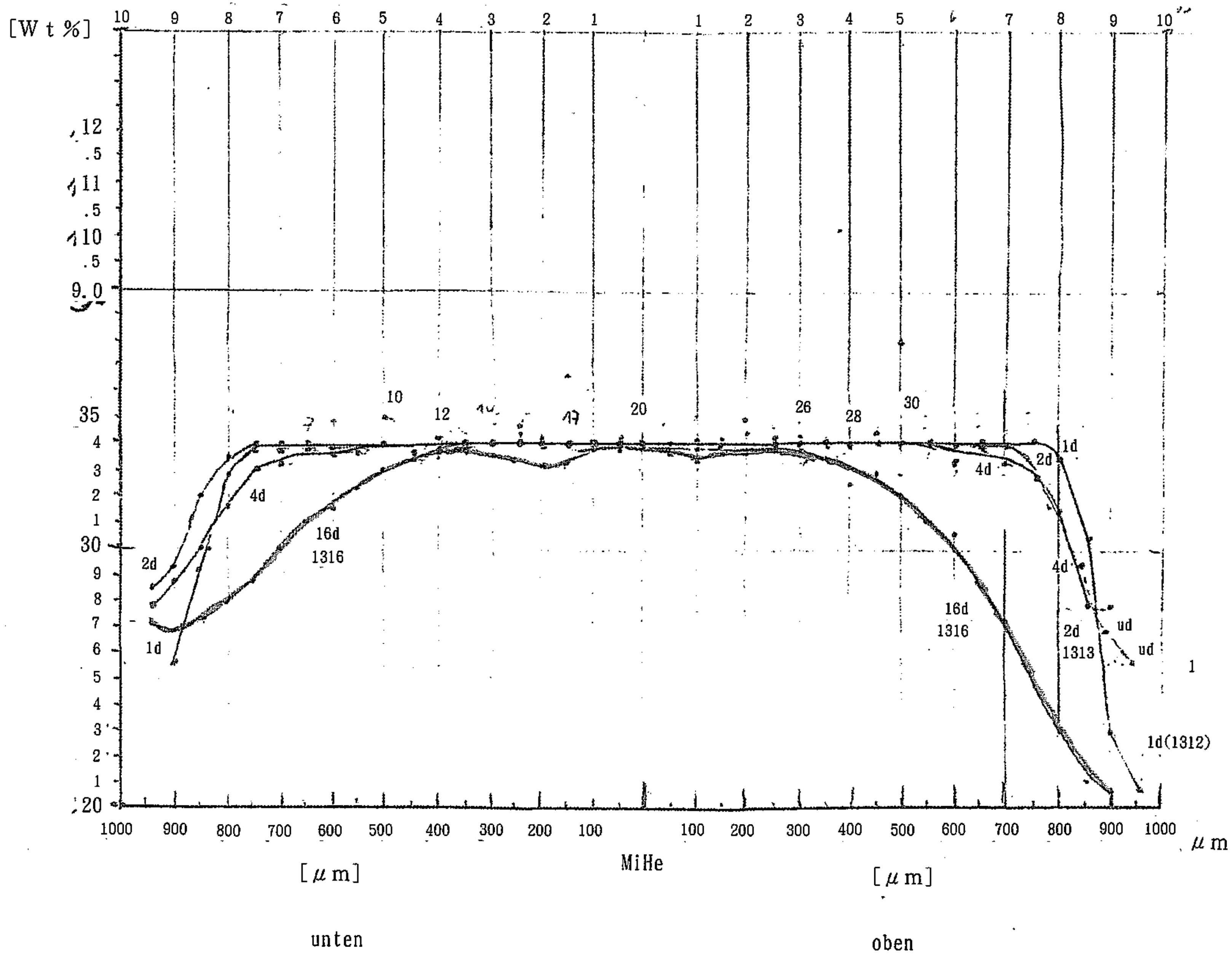

Mean residence time
Discharge interval
Rh
Rh_2O_3
Pd_xTe_y
RuO_2, PdO, RhO_2
Cold Cap
T_g
Mean pathway
Ostwald ripening
Cont. growth
Temperature (°C)
1000
500
0
1
10
10^2
10^3
10^4
Time (min)
Pathways of waste through the cold cap. Formation of Pt–metal phases

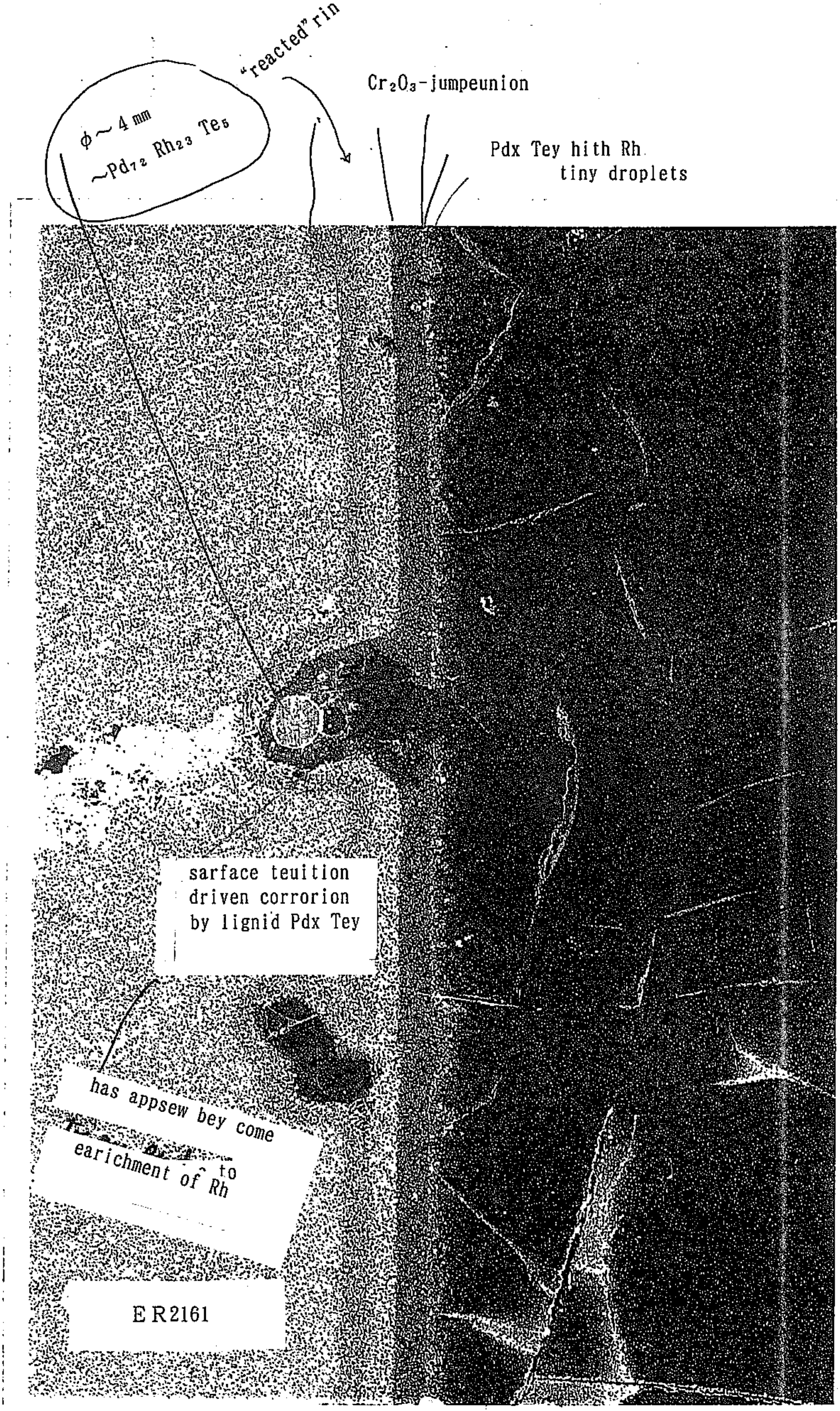


Calcination of HLLW on the surface of glass beads in the cold cap (idealized)

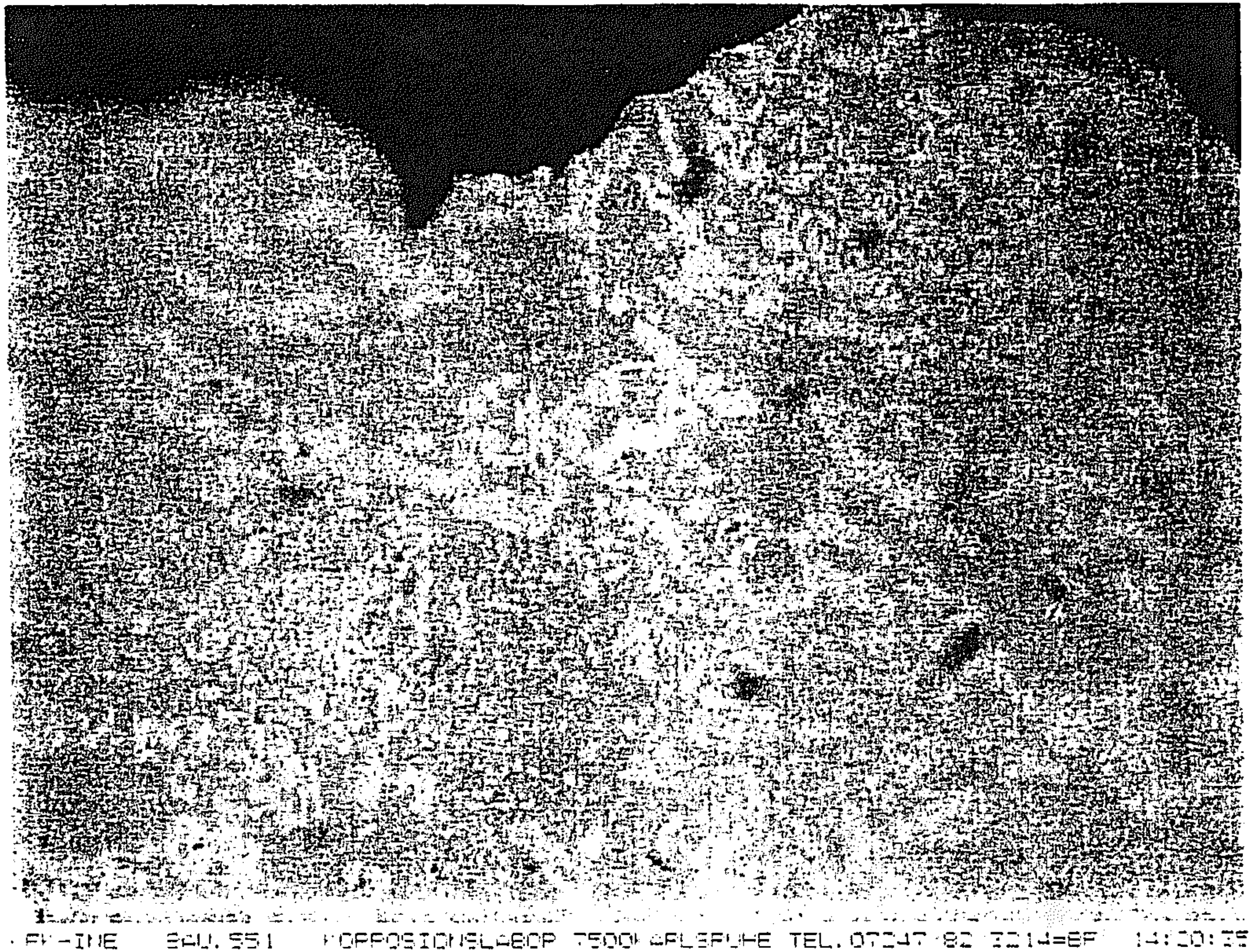

Ni-concentration
[W t %]
70
8
6
4
2
60
8
6
4
2
50
200 T
128 T
64 T
32 T
1d
16 d (1316)
4d
6
10
16d
36
34
32
1d
1d(1312)
4d (1314)
1000
900
700
500
300
100
100
300
500
700
900
1000
MiHe
[μ m]
unten
below
oben
above

Cr concentrahan




"reacted" rin
φ ~ 4 mm
~Pd72 Rh23 Te5
Cr2O3-jumpeunion
Pdx Tey hith Rh
tiny droplets
sarface teuition
driven corrorion
by lignid Pdx Tey
has appsew bey come
earichment of Rh to
ER2161

1i177190




TEL.07247
14:43:58

Conclusions

- INCONEL 690 alloy is being selectively oxidized in such a way that Cr and some 50% of the Fe (not shown here) leave the alloy apparently as a function of oxygen availability
- Fe and Cr are being dissolved in the glass product melt
- Cr oversaturation in the glass melt leads to a secondary formation of Cr_2O_3
- Selective oxidation kinetics are influenced by Cr-Concentration in the melt