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海外出張報告

第9回PNC/KfK高レベル廃棄物管理会議発表資料集

(1989年10月9～11日 西独、カールスルーエ原子力センターで開催)

— 高放射性廃液固化研究報告 —

1990年7月

動力炉・核燃料開発事業団

東海事業所

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2001

海外出張報告

第9回PNC/KfK高レベル廃棄物管理会議発表資料集 (1989年10月9～11日 西独、カールスルーエ原子力センターで開催) － 高放射性廃液固化研究報告－



齋藤信一*，高橋武士**
吉岡正弘**，五十嵐寛**

要 旨

本資料集は1989年10月開催された「第9回PNC-KfK高レベル廃棄物管理会議において、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、高レベル廃棄物管理に係るR&D概要、白金族元素含有廃液処理時の溶融炉における白金族元素挙動、ガラス固化体特性およびオフガス処理系におけるRuとCsの挙動、WAK廃液のガラス固化計画、並びにWAK向けのガラス固化試験施設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号メルタの運転経験、白金族含有ガラスの特性、揮発性ルテニウムの水への吸収挙動および廃メルタ解体技術に関するものである。

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1. K f K 発表分

1.1 Overview about KfK Activities in HLLW Management

**OVERVIEW ABOUT KfK ACTIVITIES
IN HLLW MANAGEMENT**

for

**the 9th Annual PNC-KfK Meeting on High-Level
Waste Management held at KfK/INE**

October 1989

HAWC-WAK Project
Industrial Vitrification
PAMELA

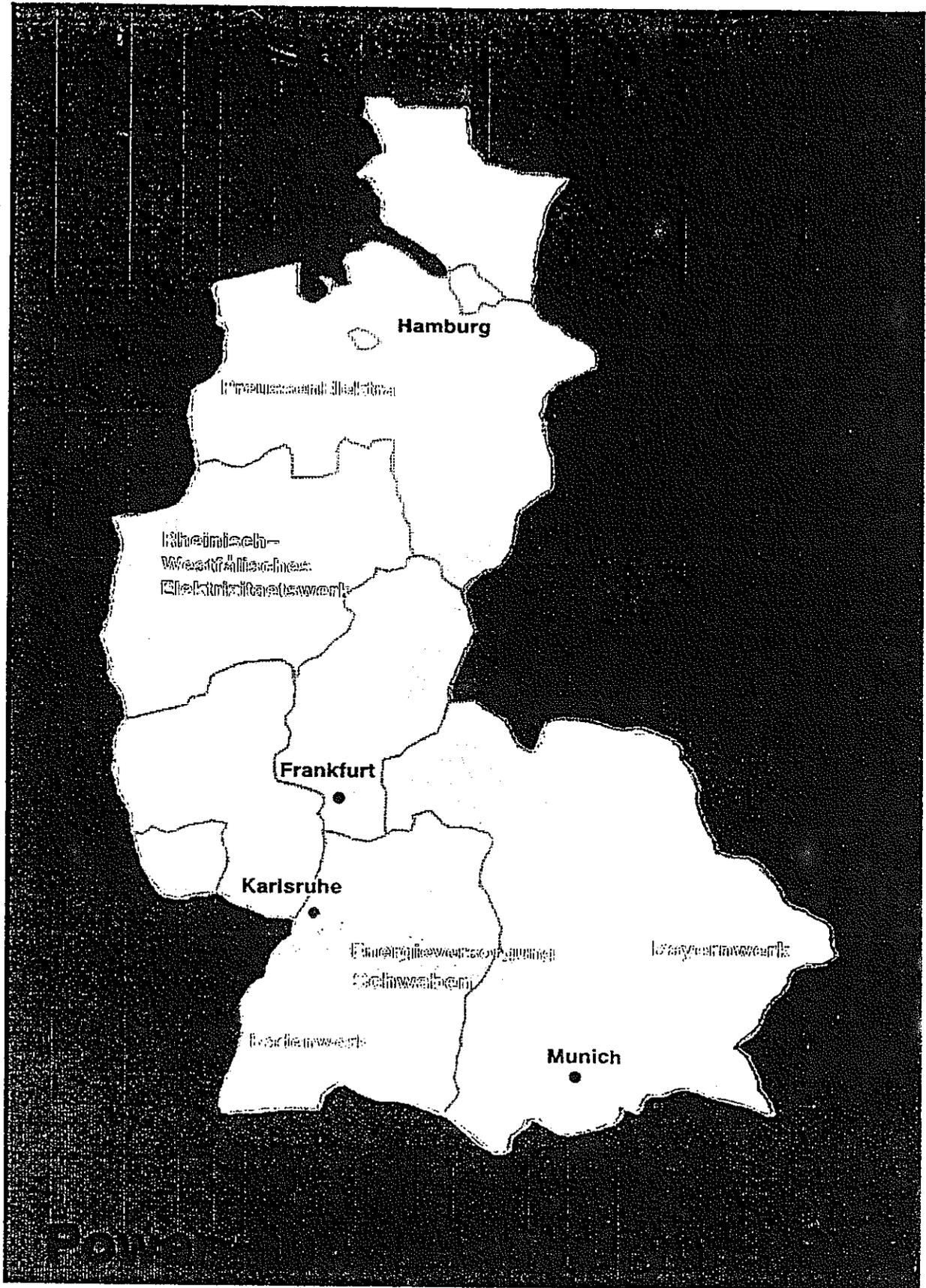
Melter Modelling

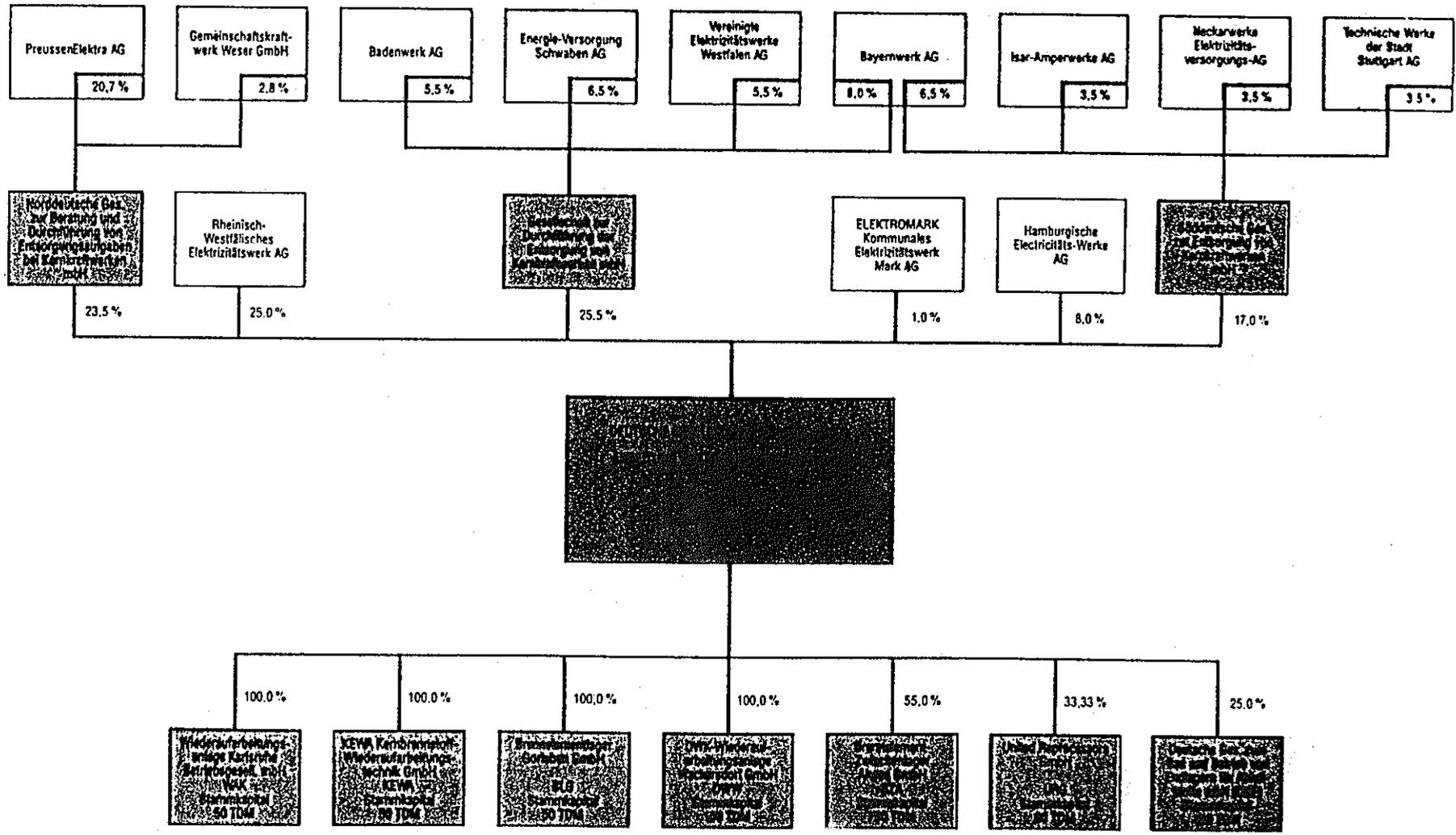
Noble Metal Structure

Process Chemistry

Melter Corrosion

Major HLLW Activities





 DWK	DWK - Anteilseigner und Beteiligungsgesellschaften Stand: 01.01.1989	D 23 17 70 1988
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Industrial Vitrification

Status:

Comprehensive experience available

63 d continuous operation

63 m³ HAWC simulate

275 kg noble metals

25 t glass product

Change of melter design required

Objective:

Completion of technology development

Promotion to technical maturity

**Construction of advanced K-W3 melter
(Modelling-aided design)**

Long-term testing of K-W3 melter

HLLW OF THE WAK PLANT

(HAWC-WAK)

Reprocessed Uranium	174 t
Average burn-up	16000 MWd/to
Existing volume	62 m³
Specific volume	356 l/t
Specific activity	528 Ci/l
Salt	155 g/l
Oxide residue	95 g/l
Free nitric acid	5.3 M/l
Sodium	16 g/l
Ruthenium	3 g/l
Palladium	1.8 g/l
Rhodium	0.8 g/l
Noble metal quantity	350 kg
Expected Volume	20 m³
Specific activity	1000 Ci/l appr.

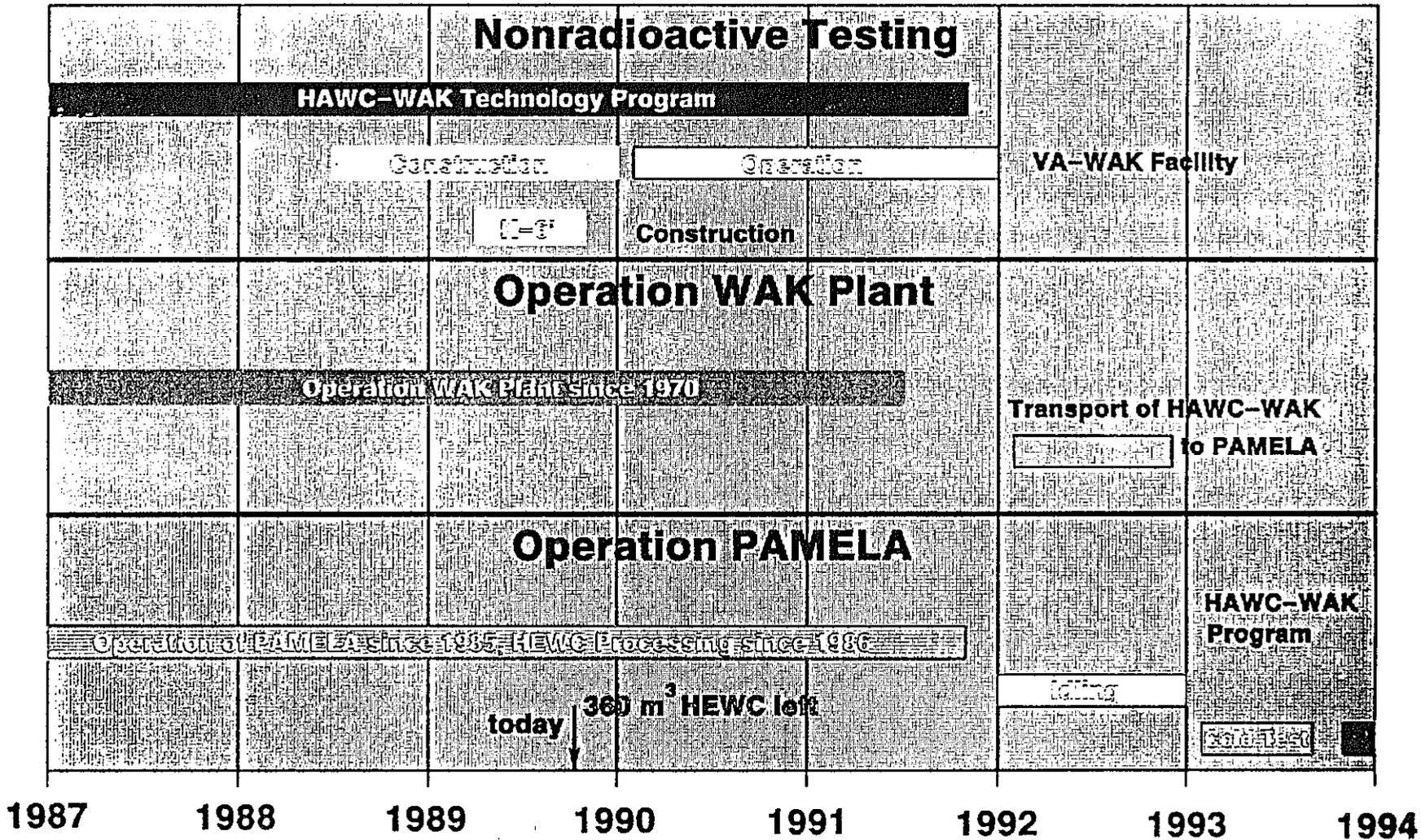
HAWC-WAK VITRIFICATION

Vitrification plant	PAMELA
Transport license	applied for
Start of processing	midnineties
Melter type	K-6 (KfK)

Nonradioactive Testing

Technol. program	1987-1991
Vitrification techn.	KfK-INE
Prototype melter	K-6'
Design throughput	22-25 l/h
Design melt rate	11-14 kg/h
Test facility	VA-WAK
Testing start-up	January '90

TIME TABLE FOR THE HAWC-WAK MANAGEMENT (PRELIMINARY)



MELTER MODELLING

Physical Modelling

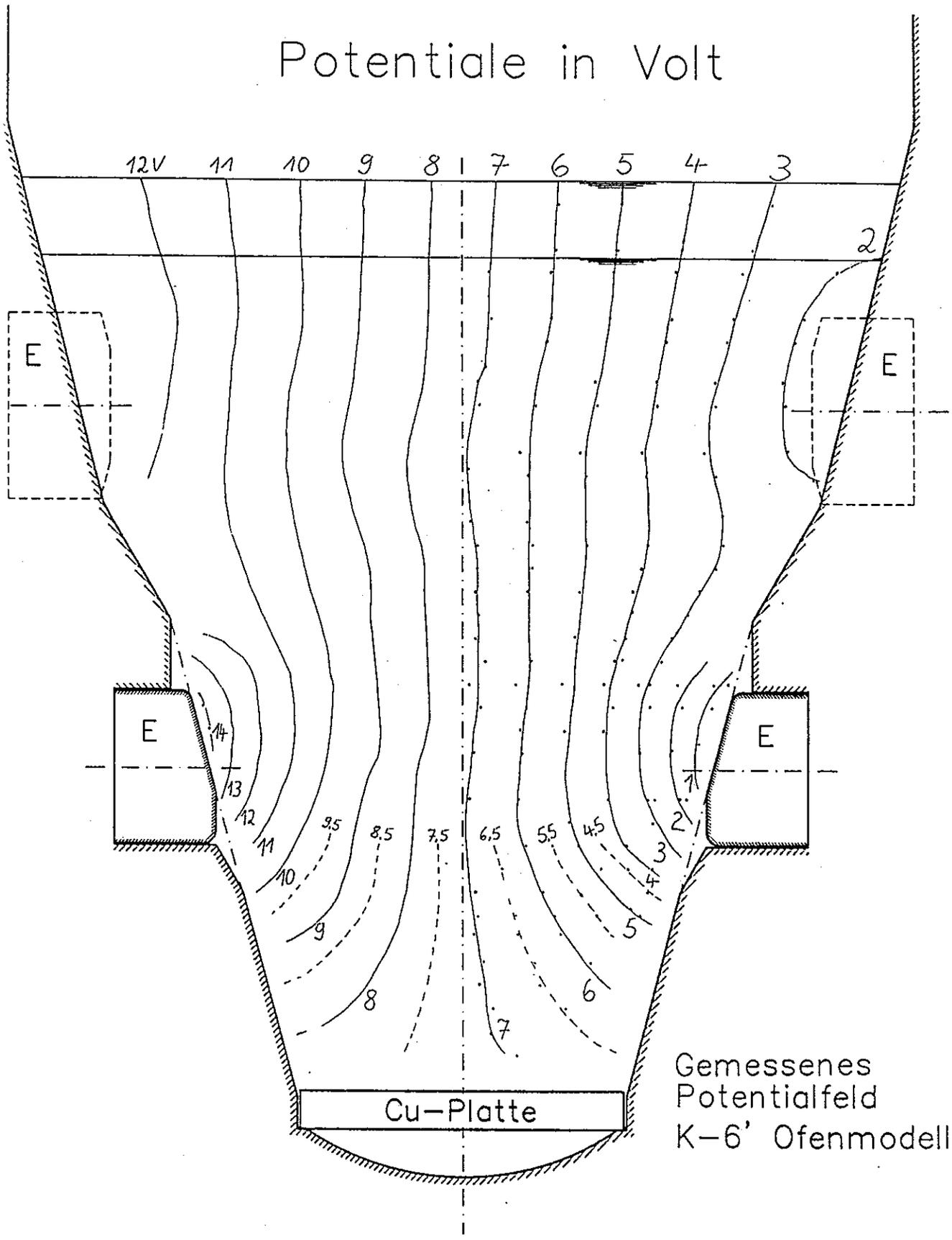
- **Electrical potential field**
- **Electrical field intensity**

Mathematical Modelling (3D)

- **Electrical potential field**
- **Power density field**
- **Temperature field**
- **Velocity field**

Flow Mechanic Studies

- **Flow behaviour of noble metal sludge layers along inclined melter walls**
- **Material properties**
- **Inclination**



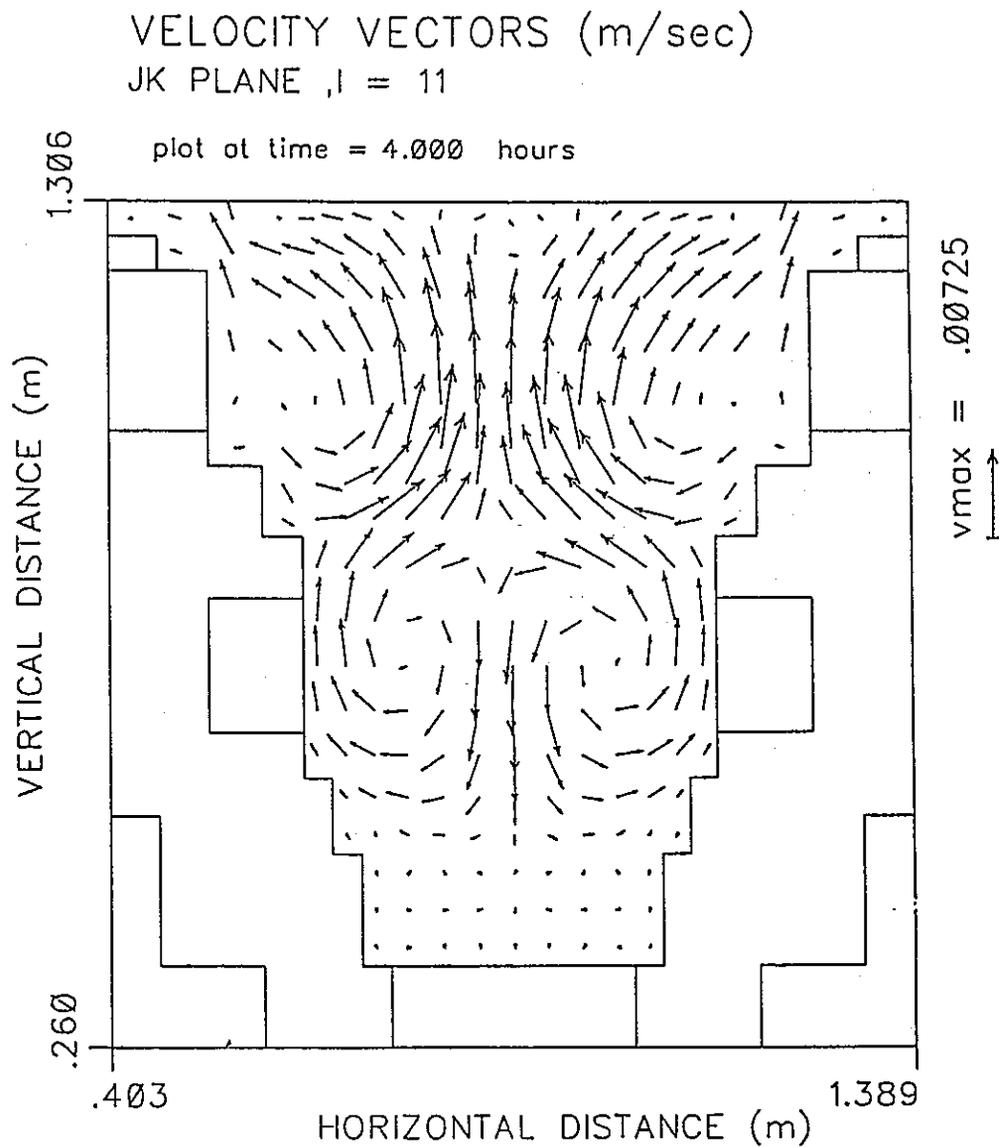


Figure 33.2: Computed velocity vectors in plane V_5 for case 1D

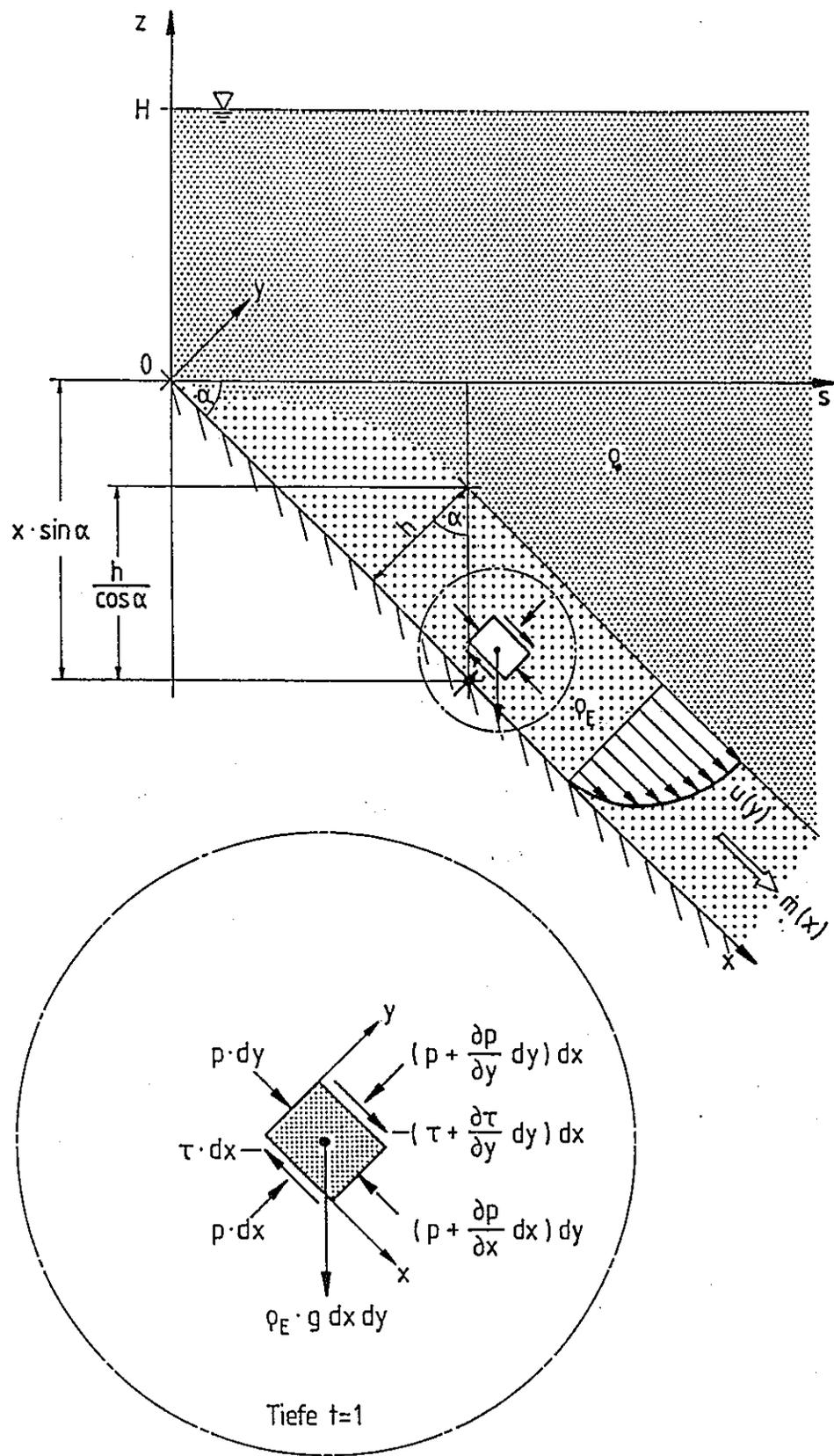


Bild 2: Filmströmung - Modellbetrachtung

1.2 Evaluation of the operational results of the noble
metal campaigns W4 and W5

EVALUATION OF THE OPERATIONAL RESULTS OF THE NOBLE METAL CAMPAIGNS W4 AND W5

for

the 9th Annual PNC-KfK Meeting on High-Level
Waste Management held at KfK/INE

October 1989

1. Objective and Results of the different Campaigns

2. Evaluation

Electrical

Noble Metal Sludge Residue in the Melter after the Campaigns

Forced Melt Convection

3. Investigations currently performed due to the Results of the Campaigns

Theoretical

Physical Modelling (electrical Field under the Influence of Noble Metals)

Mathematical Modelling (Fully coupled Solutions including noble metal influence, 2D and 3D Calculations)

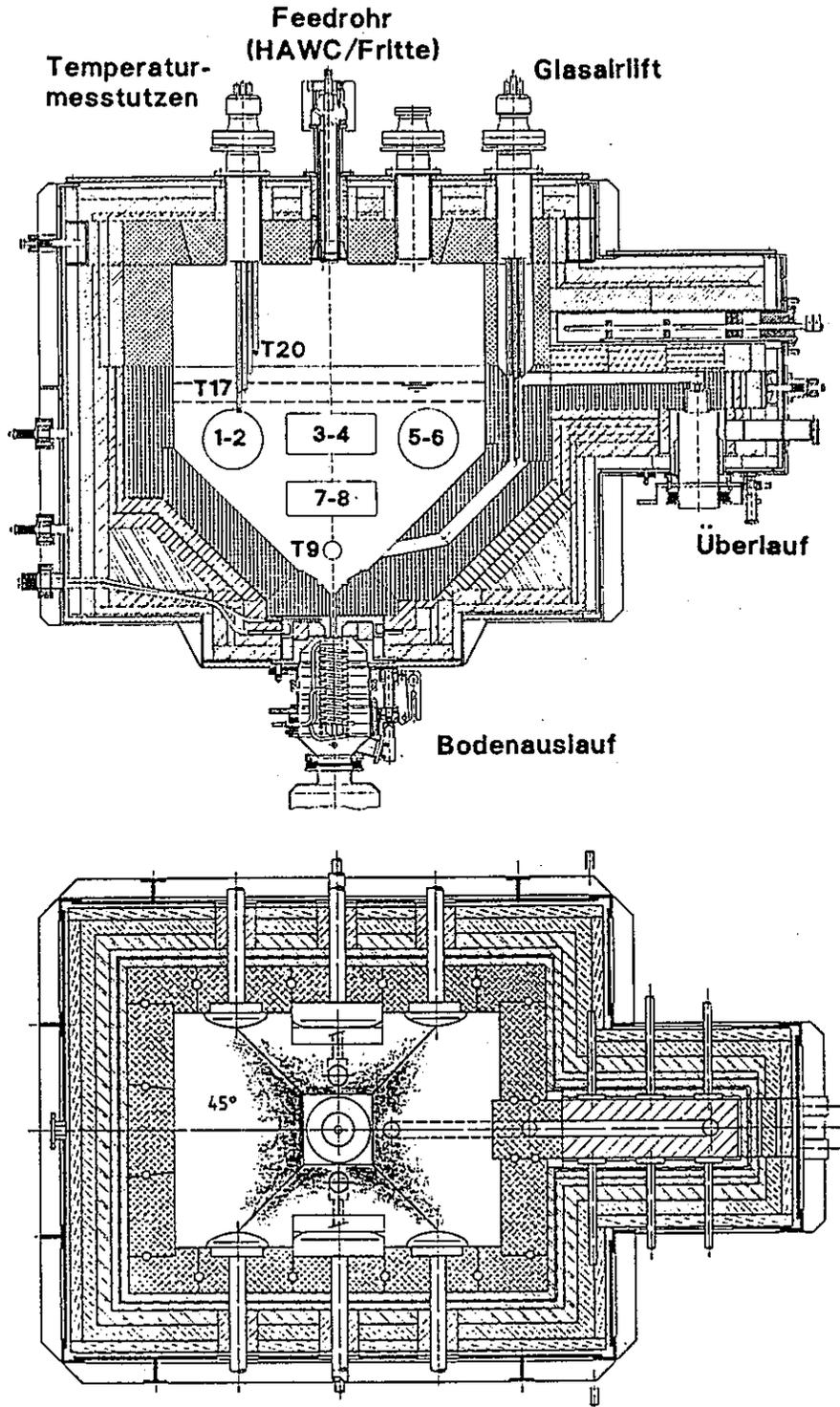


Abb. 2.1 : Längsschnitt und Horizontalschnitt durch den K-W2 Schmelzofen (vereinfachte Darstellung)

OBJECTIVES OF THE CAMPAIGNS W4 AND W5

W4 (April/May 1988)

Demonstrate

- 72 l/h Feed rate (design throughput)
- 30 kg/h Glass rate
- Problem free draining of noble metals
- Complete draining of glass from the melter after run termination
- Condition of melter after run termination
- 33 m³ HLLW-simulate
- 72.6 kg Ru
- 16.5 kg Rh
- 62.7 kg Pd

W5 (Jan./Febr./March 1989)

Demonstrate

- Influence of melter plenum heating on feed rate
- Influence of air bubbling on noble metal draining efficiency
- Feed rate under specified process condition at the glass pool surface
- Draining behaviour of noble metals during run
- Influence of a 10 day's process interruption
- Draining efficiency of noble metals via overflow system
- Draining of noble metals while emptying the melter
- Melter condition after run termination and emptying
- 30 m³ HLLW-simulate
- 66 kg Ru
- 57 kg Pd

MAJOR CAMPAIGN DATA AND RESULTS

W4 (April/May 1988)

95.5 % plant availability
2.2 g/l Ru
0.5 g/l Rh
1.9 g/l Pd
55-60 l/h maximum feed rate
45-50 l/h average feed rate
13.5 wt% waste glass loading
85-90 kW power release (average)
200-350 °C melter plenum temperature
13.9 to glass
35 cannisters (430φx1350)
800 glass samples

Major problems: bad process conditions on glass pool surface due to too high feed rates (crust thickness up to 10-40 cm), el. shortening due to extensive noble metal accumulations in the melter

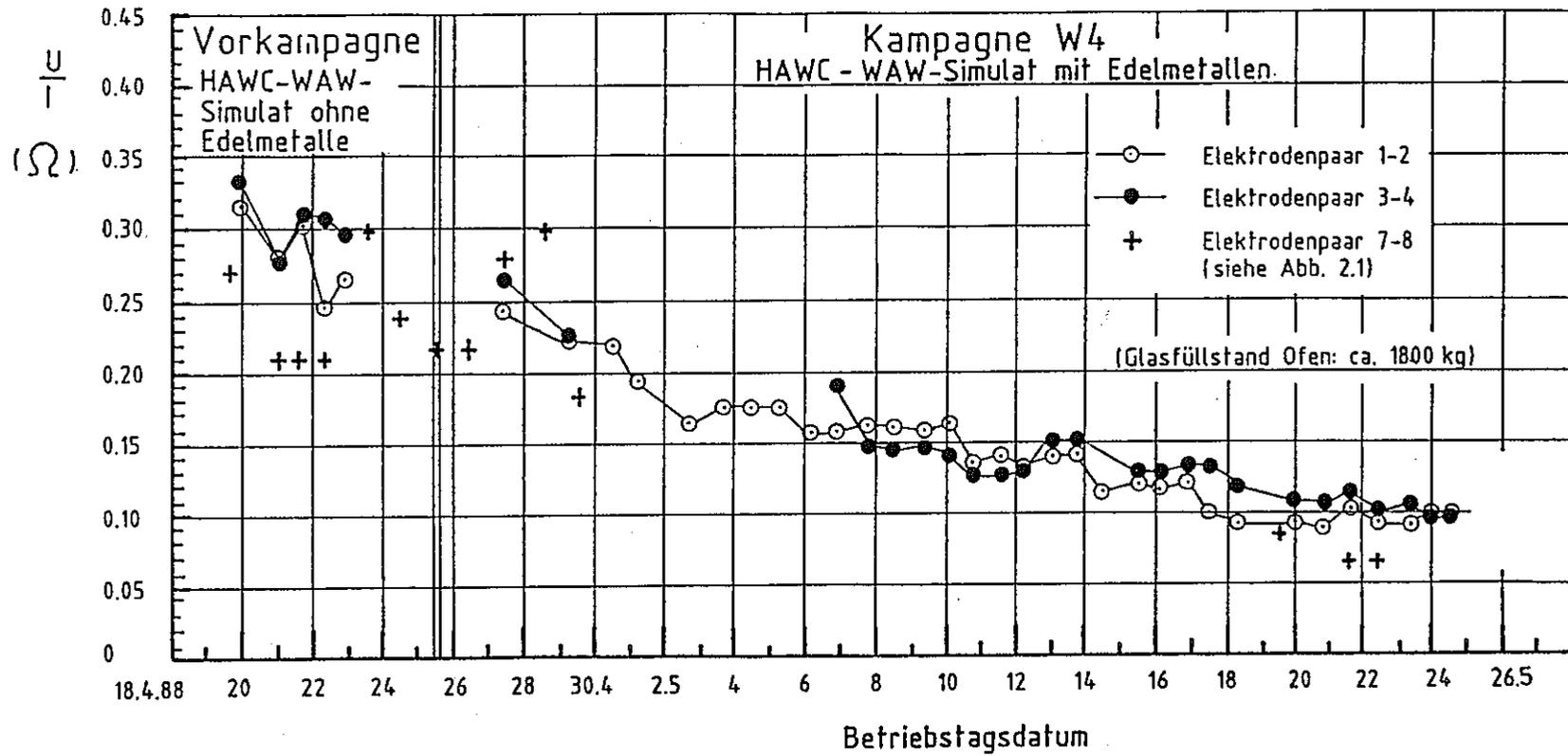
Draining efficiency: 70.5 % Ru
(during the run) 67.1 % Rh
63.0 % Pd

W5 (Jan/Feb/March 1989)

100 % plant availability
2.2 g/l Ru
1.9 g/l Rd
50 l/h maximum feed rate
47 l/h average feed rate
13.5 wt% waste glass loading
75-90 kW power release
450-600 °C melter plenum temperature
12.1 to glass
33 cannisters (430φx1350)
2050 glass samples

Major problem: el. shortening due to noble metal accumulation

Draining efficiency: 85.5 % Ru
(during the run) 110.9 % Pd



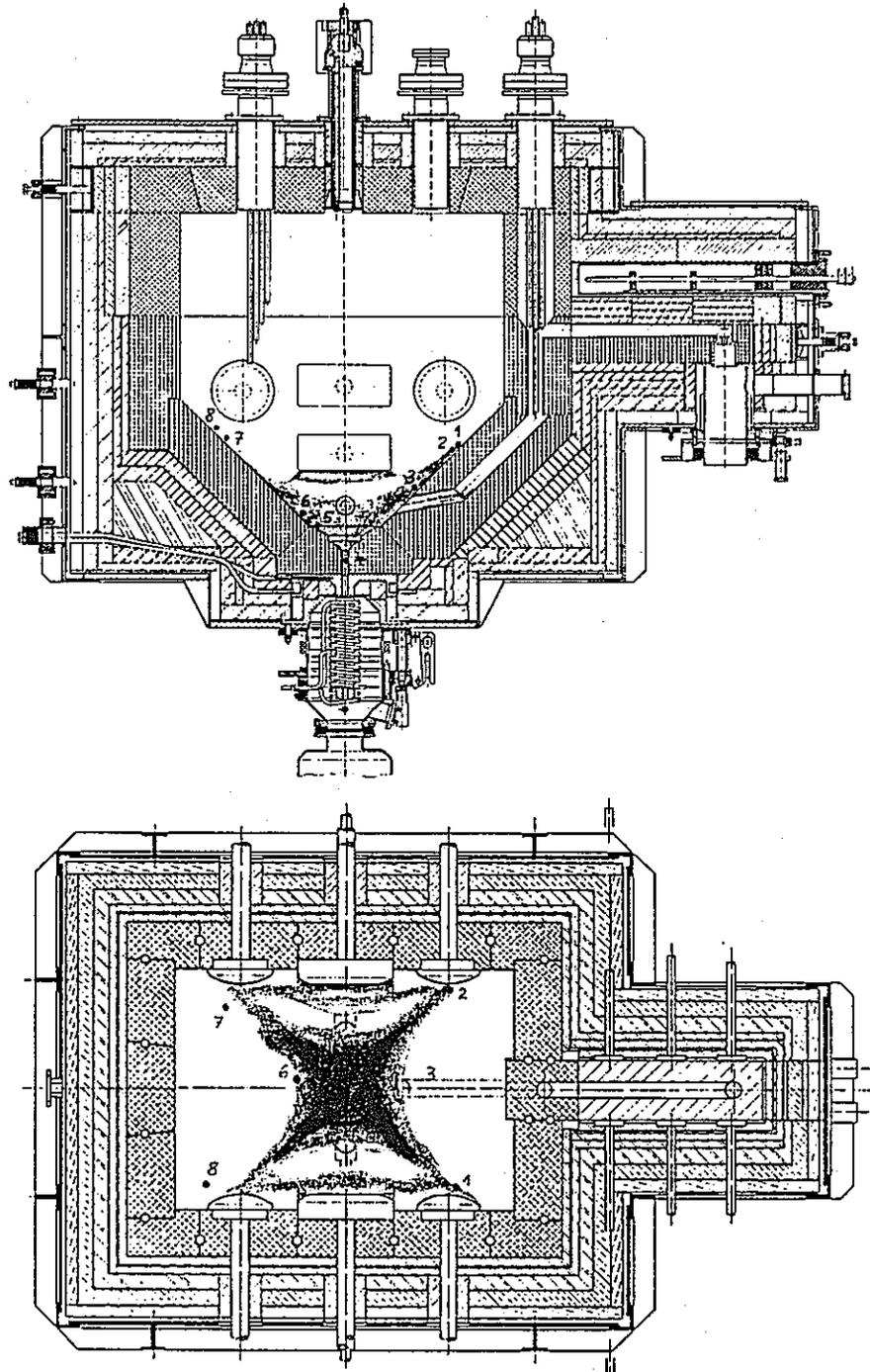
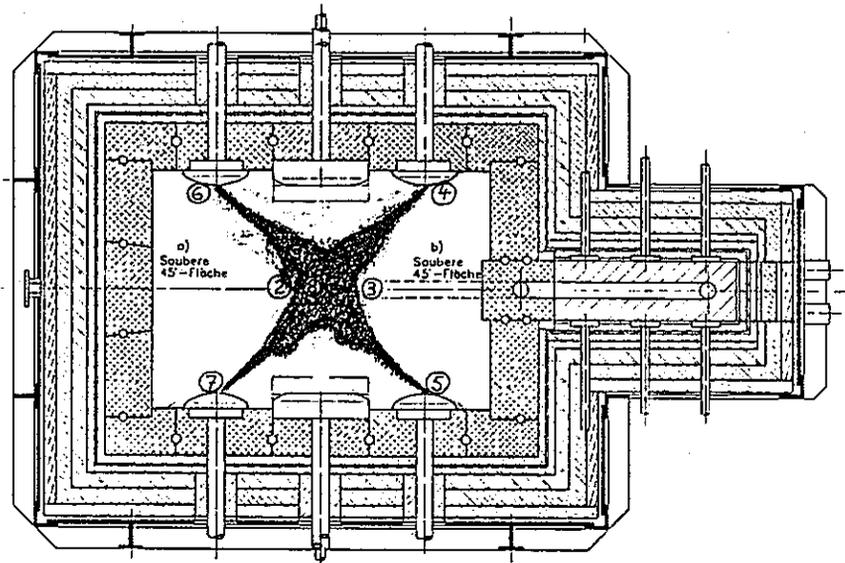
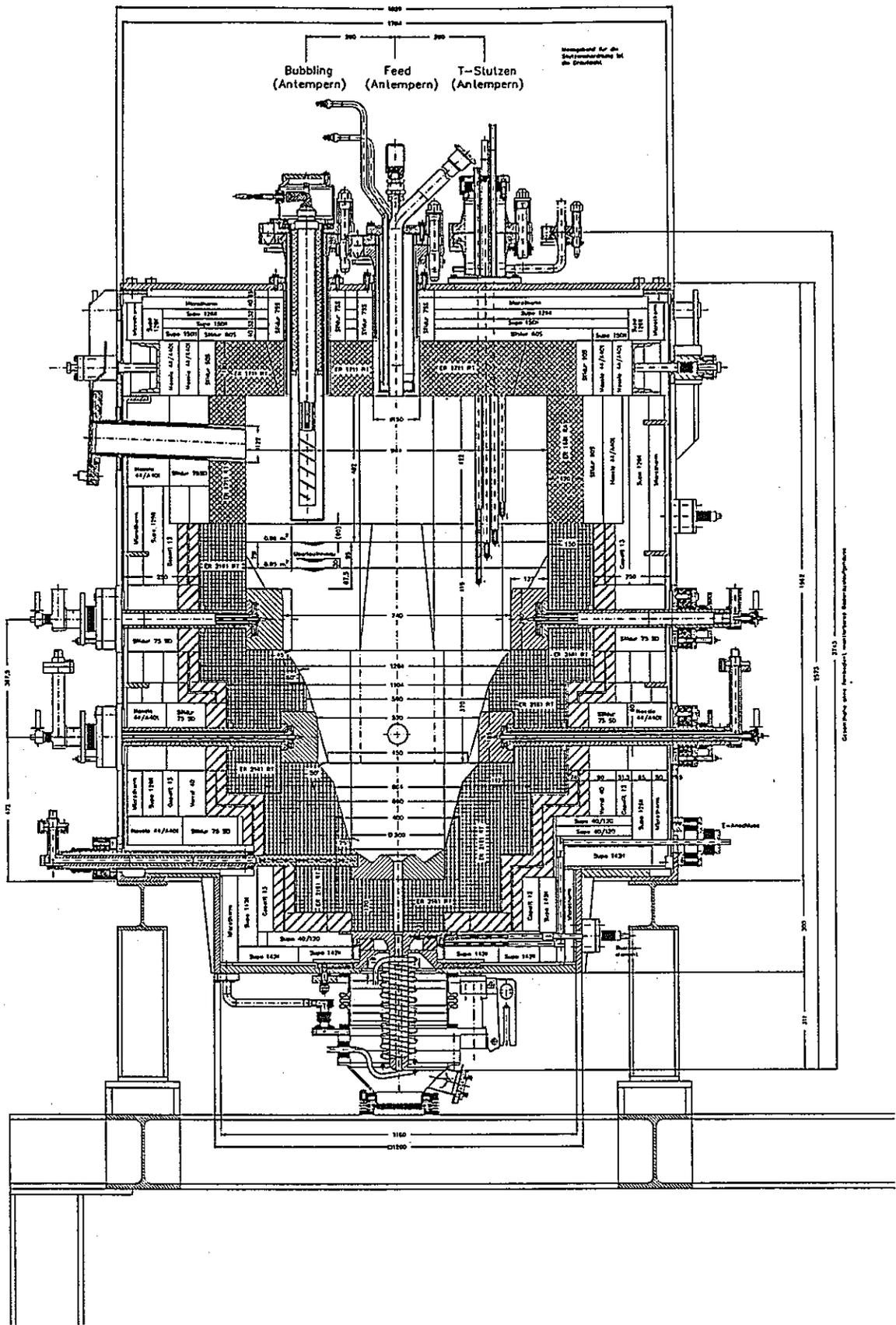


Abb. 3.6: Restglasschmelze im K-W2 Schmelzofen nach Entleerung des Ofens unmittelbar am Ende der Betriebskampagne W4. Die Zahlen bezeichnen Probenahmestellen. Von diesen Stellen wurden schmelzflüssige Glasproben genommen. (Analyseergebnisse siehe Tab. 4.3)



Probe Nr. *)	Platinmetallgehalt (Gew.%)			
	Ru		Pd	
	Ist	Soll	Ist	Soll
1	6.33	0.539	0.45	0.47
2	8.37	0.539	4.67	0.47
3	6.86	0.539	3.54	0.47
4	10.23	0.539	5.75	0.47
5	11.22	0.539	4.42	0.47
6	11.23	0.539	4.44	0.47
7	11.28	0.539	5.85	0.47
8	5.08	0.539	3.0	0.47
9	8.23	0.539	3.66	0.47
Mittelwert	8.75	-	3.98	-

*) Probenahmestellen siehe Abb. 3.34
 1) Restglasschmelze 150 kg, Mittelwert Ru 8.75 Gew.%, Mittelwert Pd 3.98 Gew.%



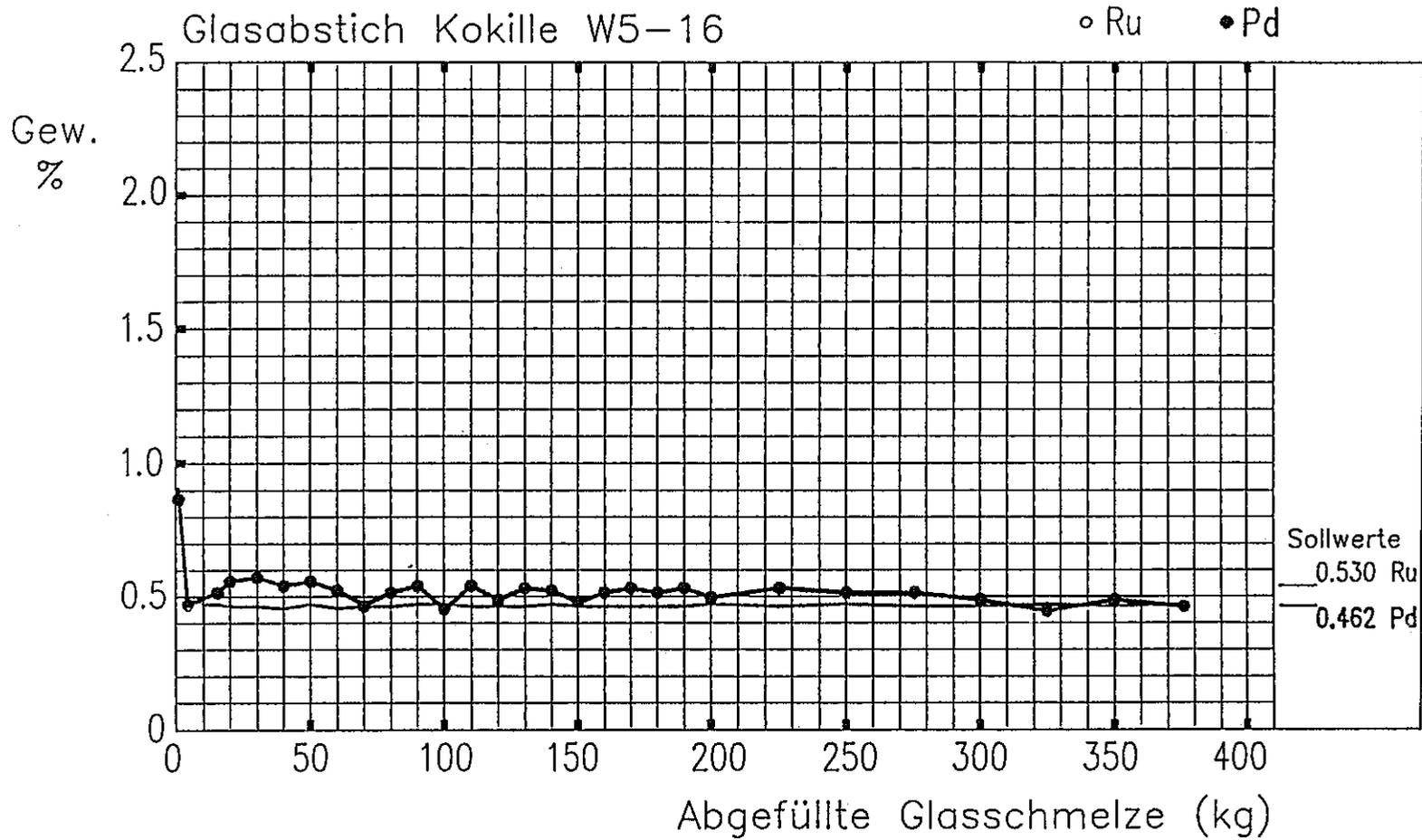


Abb 3.16 : Ruthenium- und Palladiumkonzentration im Glas-
produkt bei der Abfüllung von Kokille W5-16

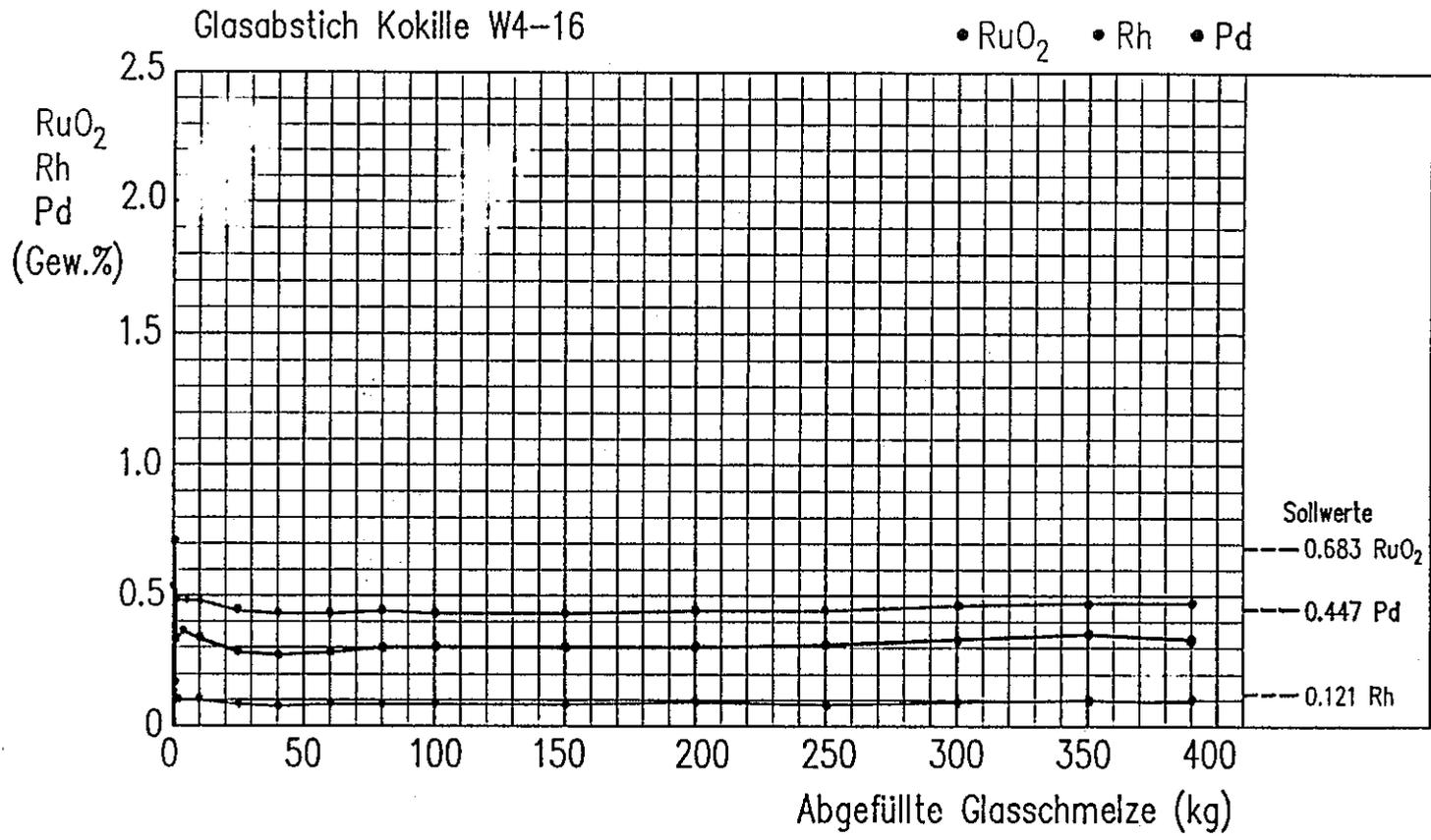


Abb. 4.22 : Edelmetallkonzentrationen im Glasprodukt bei der Abfüllung von Kokille Nr. W4-16

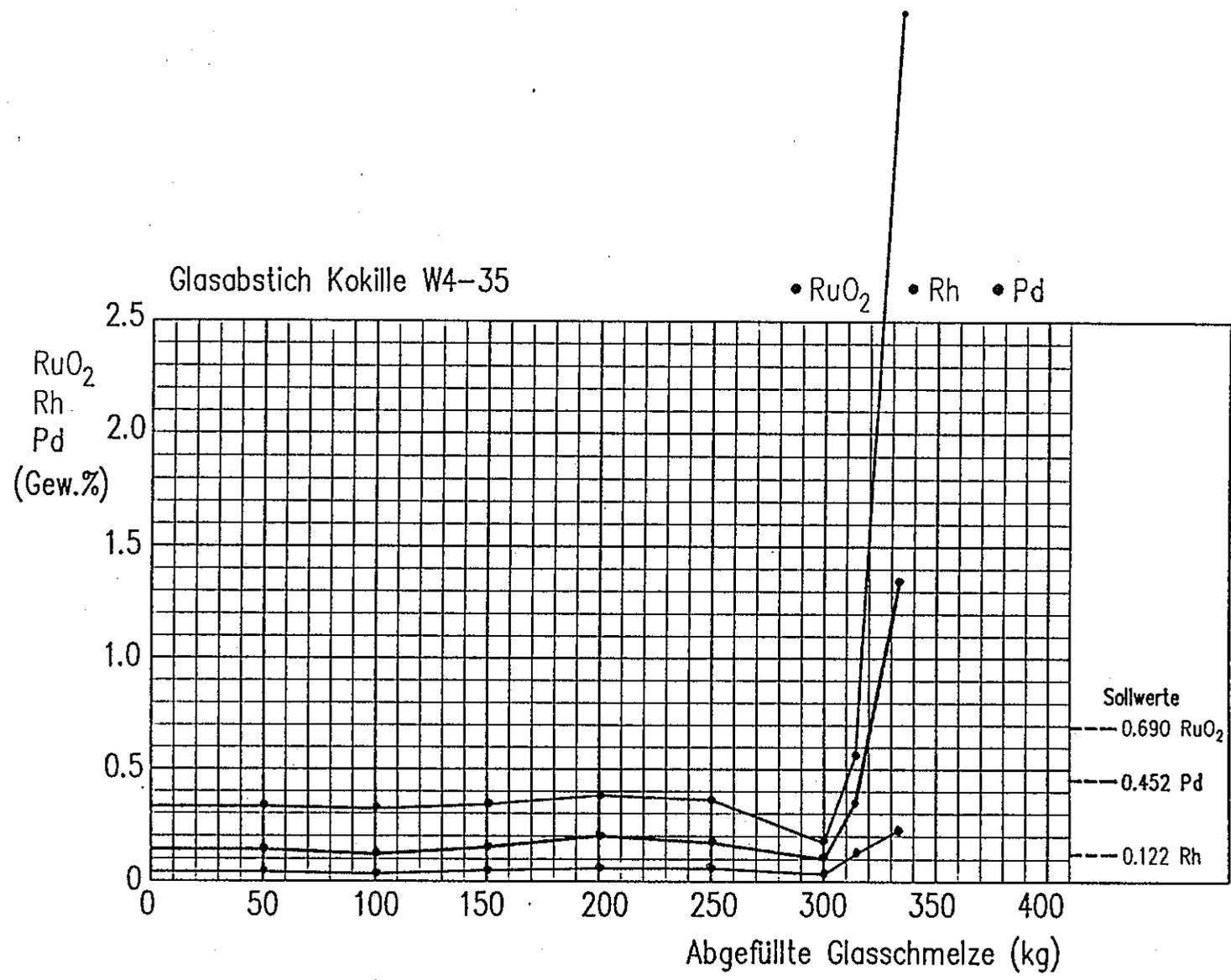


Abb. 4.41 : Edelmetallkonzentrationen im Glasprodukt bei der Abfüllung von Kokille Nr. W4-35

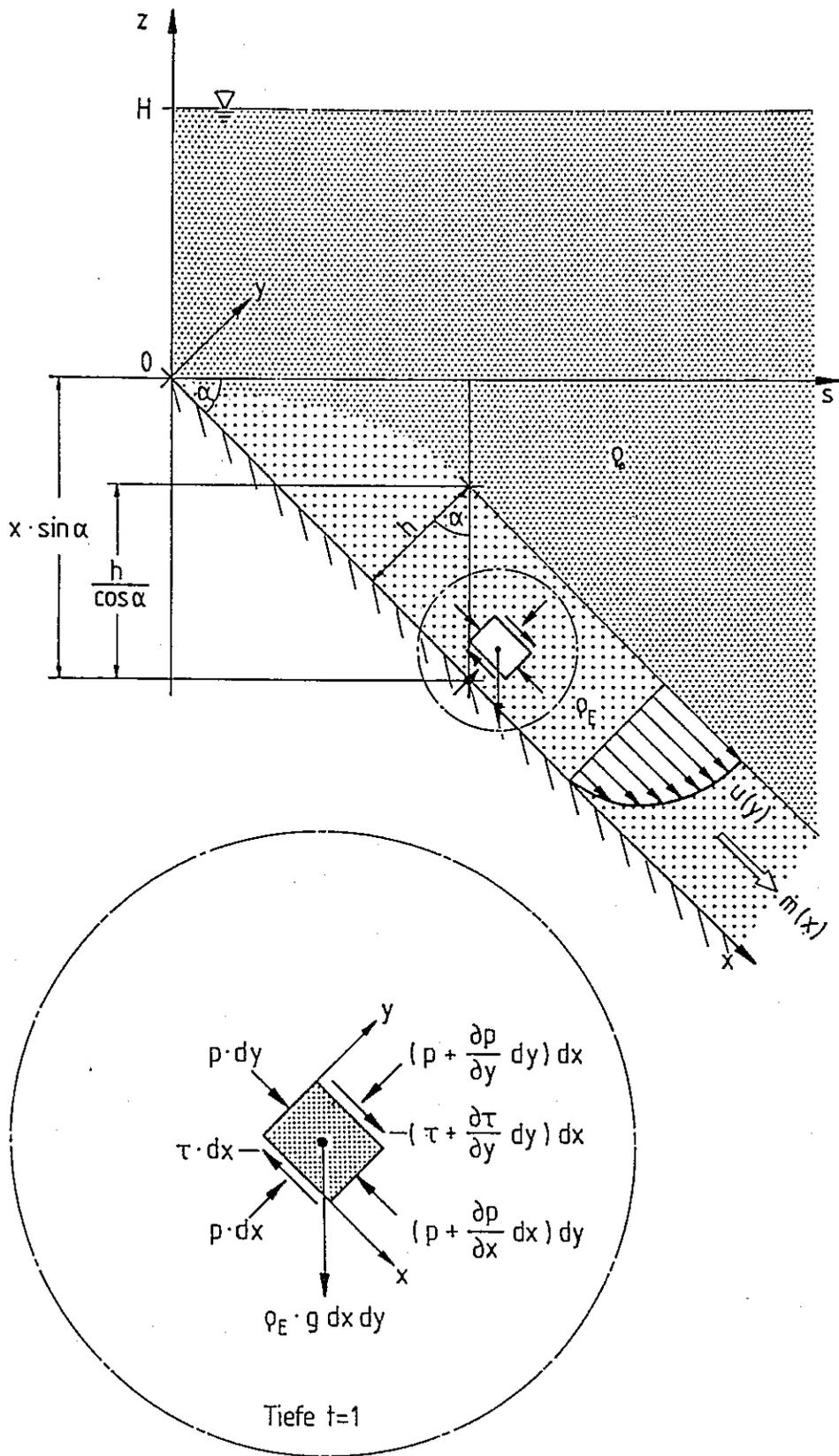


Bild 2: Filmströmung - Modellbetrachtung

2D CASE 2E: TEMPERATURE (deg. C)
 Layer = 1.0cm, bottom OFF

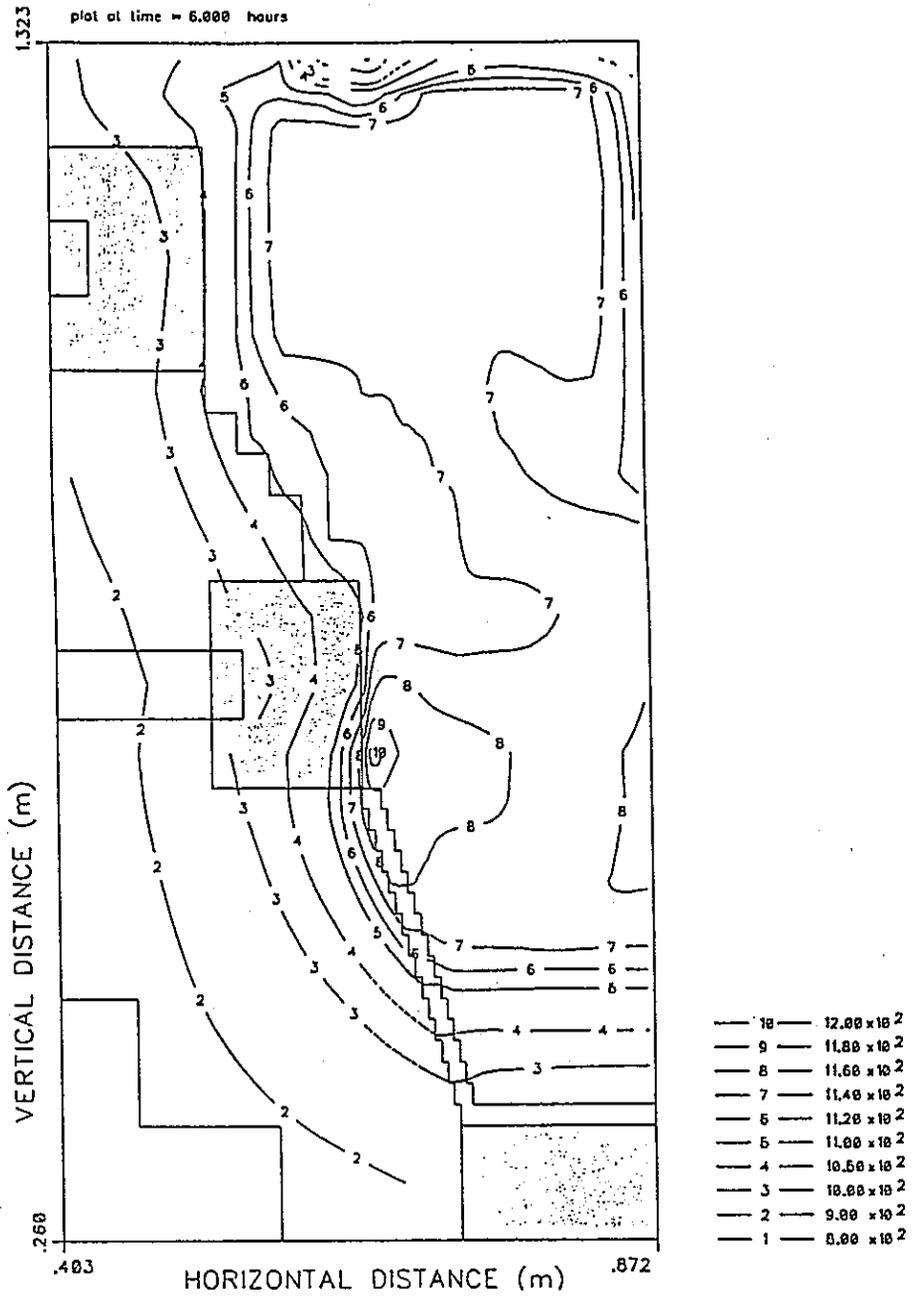


Fig. 14: Temperature contours in melt region for case 2 E

2D CASE 2E: POWER DENSITY (w/m**3)
 Layer = 1.0cm, bottom OFF

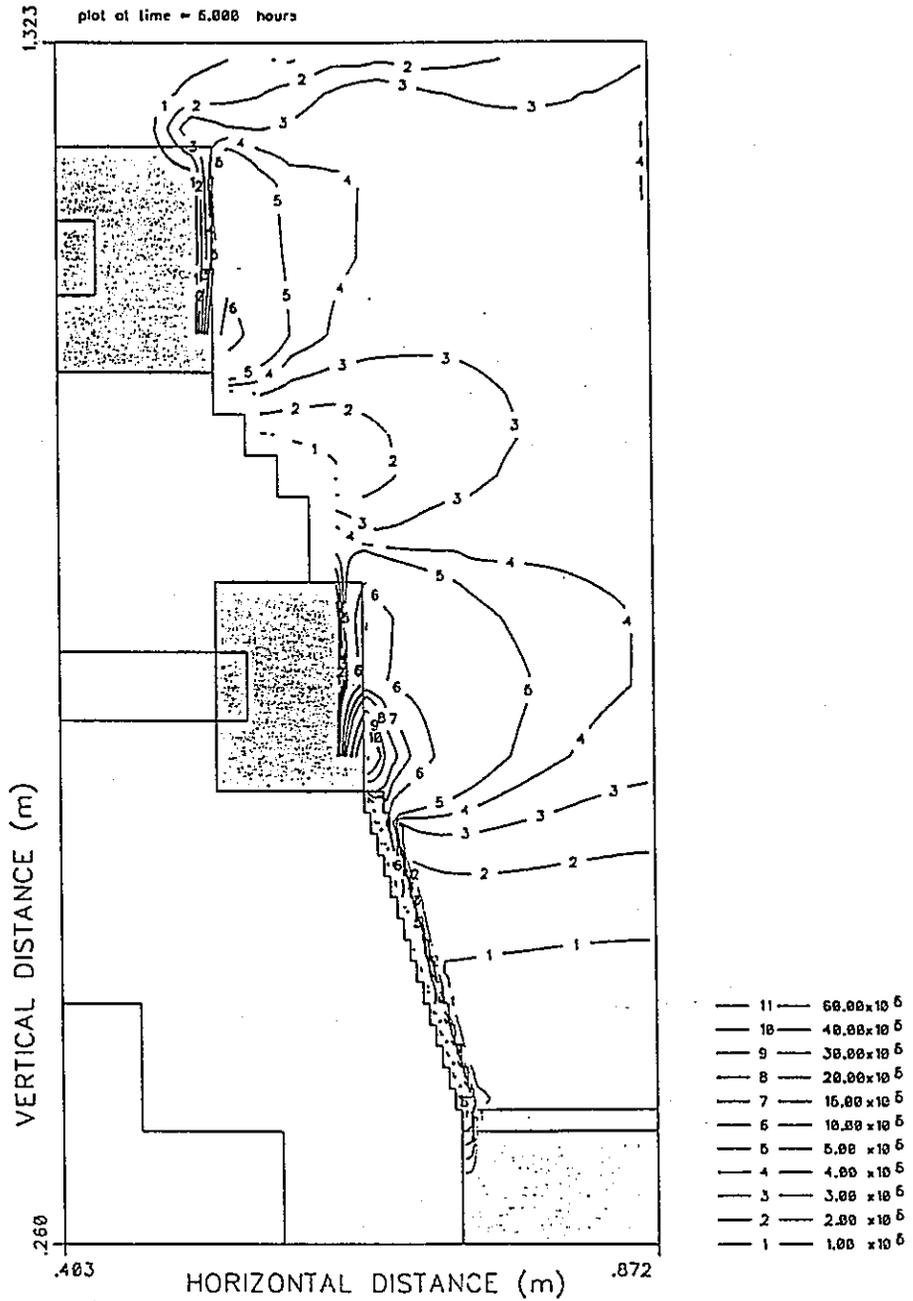


Fig. 16: Computed Joule heat power density for case 2 E

2D CASE 2E: VELOCITY (m/s)
Layer = 1.0cm, bottom OFF

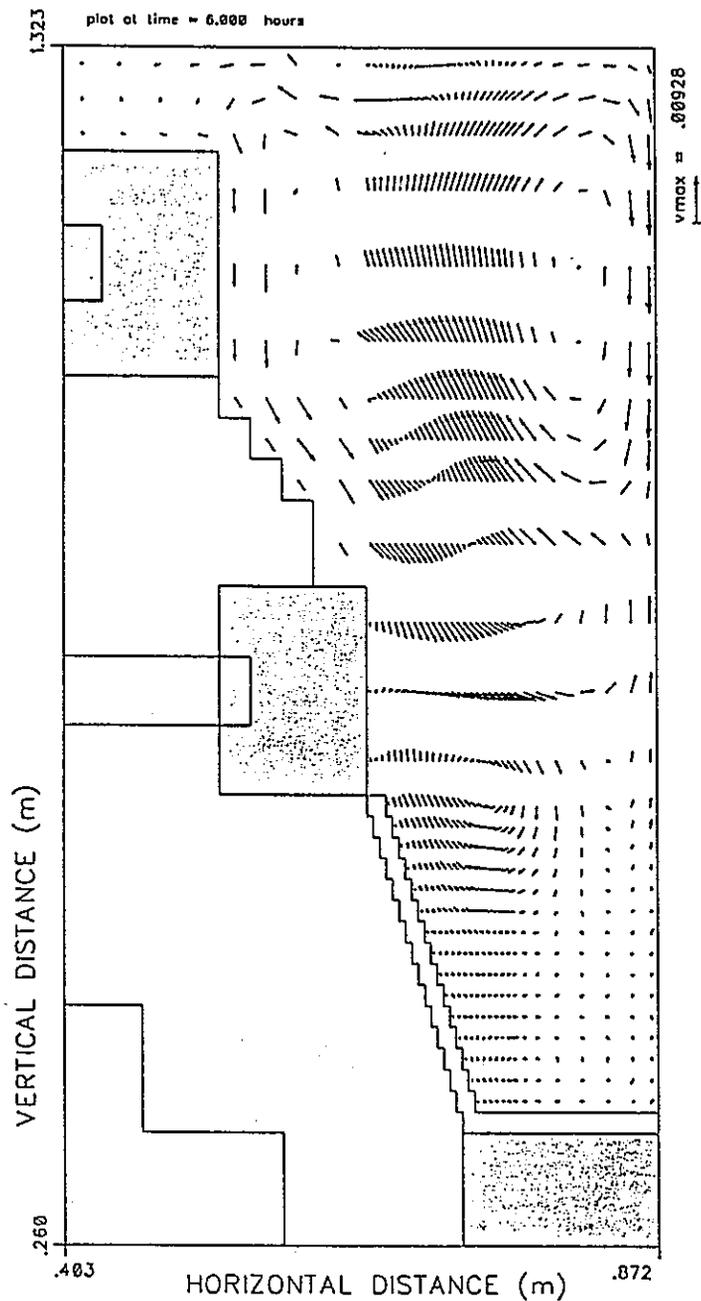


Fig. 15: Velocity vectors in glass melt for case 2 E

2D CASE 1C: TEMPERATURE (deg. C)
 NO layer, bottom OFF

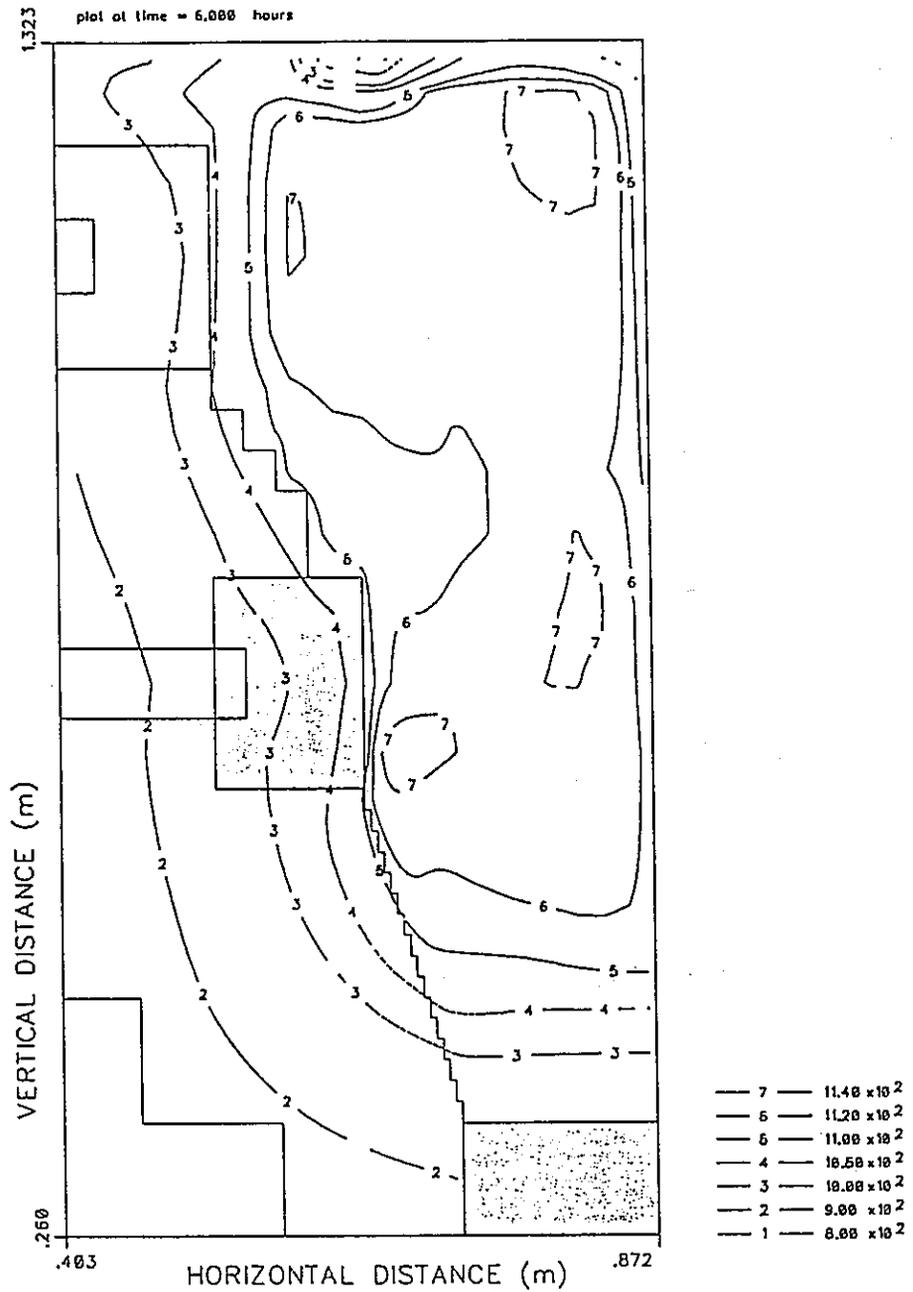


Fig. 7: Temperature contours in the melt region

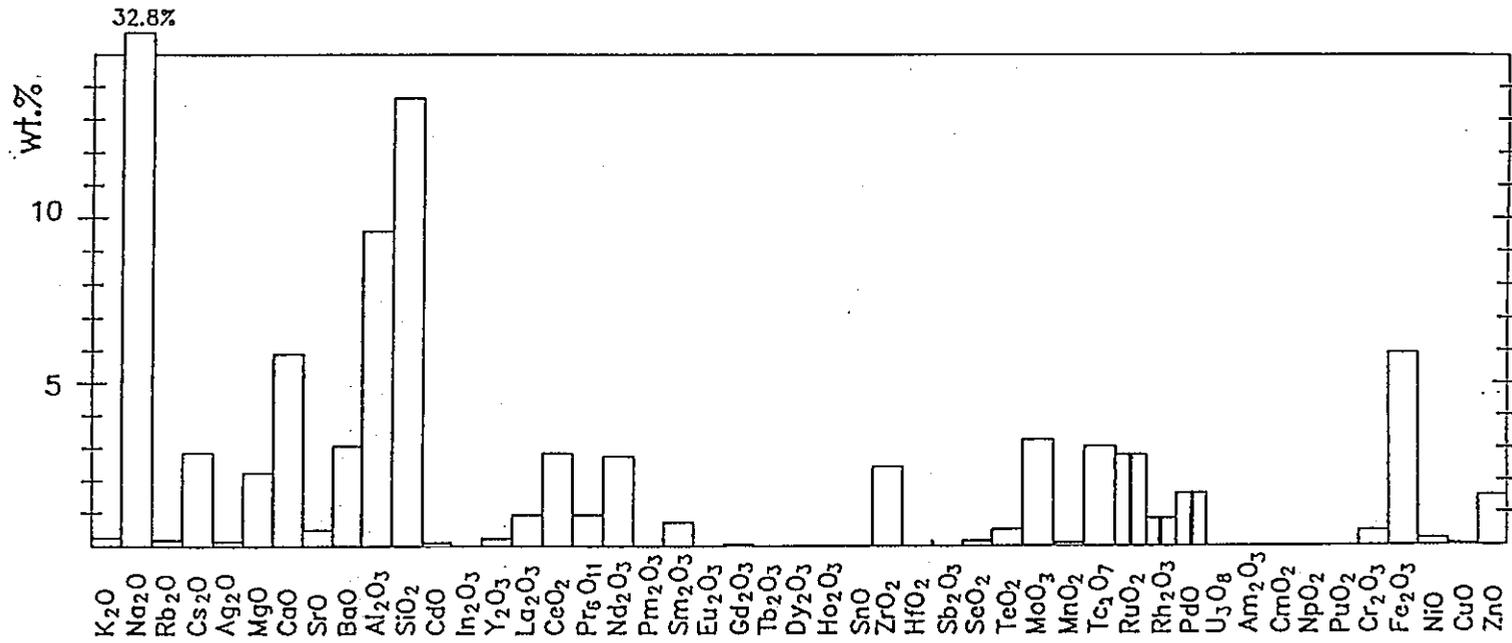
1.3 Formation and characteristics of noble metal sediments
in the melter

Conclusions

- Sufficient residence time in the lab scale melter gives a homogeneous vitreous phase in the glass product
- Aggregative sedimentation of noble metal particles occurs in the melter from the very beginning
- The particle size of the noble metal particles is increasing with the melter operation time ending at mean values of 20 μ m
- The composition of the Pd–Rh–Te particles ranges from low Rh and Te concentrations in fine particles up to 80% Rh or 23% Te in coarse particles
- Coarse Pd–Rh–Te particles are separated into two phases, one high in Te and low in Rh, the other high in Rh and low in Te
- The fine RuO₂–particles contain only minor amounts of other elements. The coarser particles contain an increasing percentage of Cr and Rh (max. approx. 6 mol% each)

Table 8: Composition of the glass product prepared by melting a mixture of NaNO_3 , MoO_3 , RuO_2 and glass frit

Oxide	Content (wt%)
Na_2O	5,2
MoO_3	1,3
RuO_2	0,7
Glass Frit	93,0



Mixture of Waste Oxides from the Nuclear Fuel Cycle

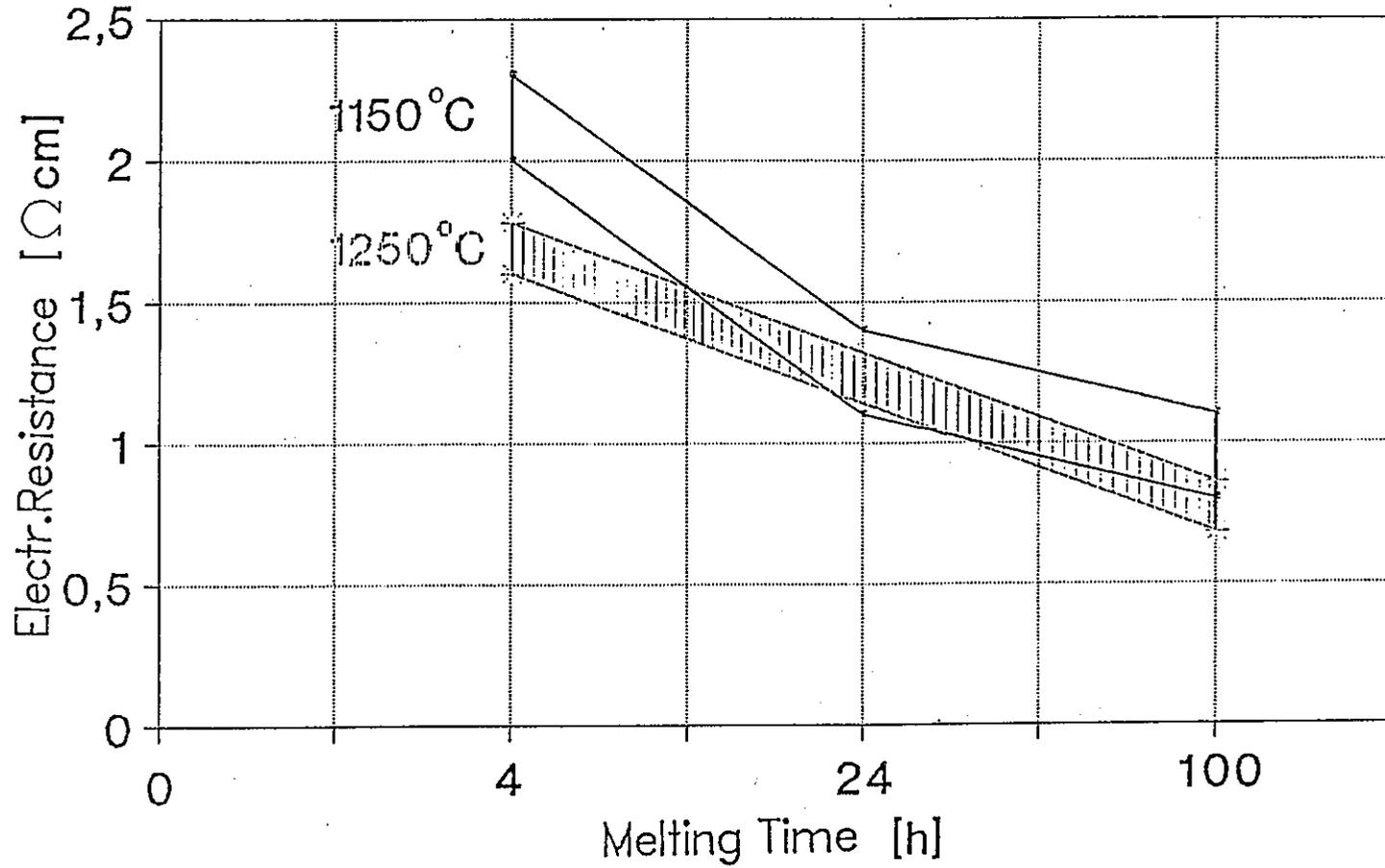
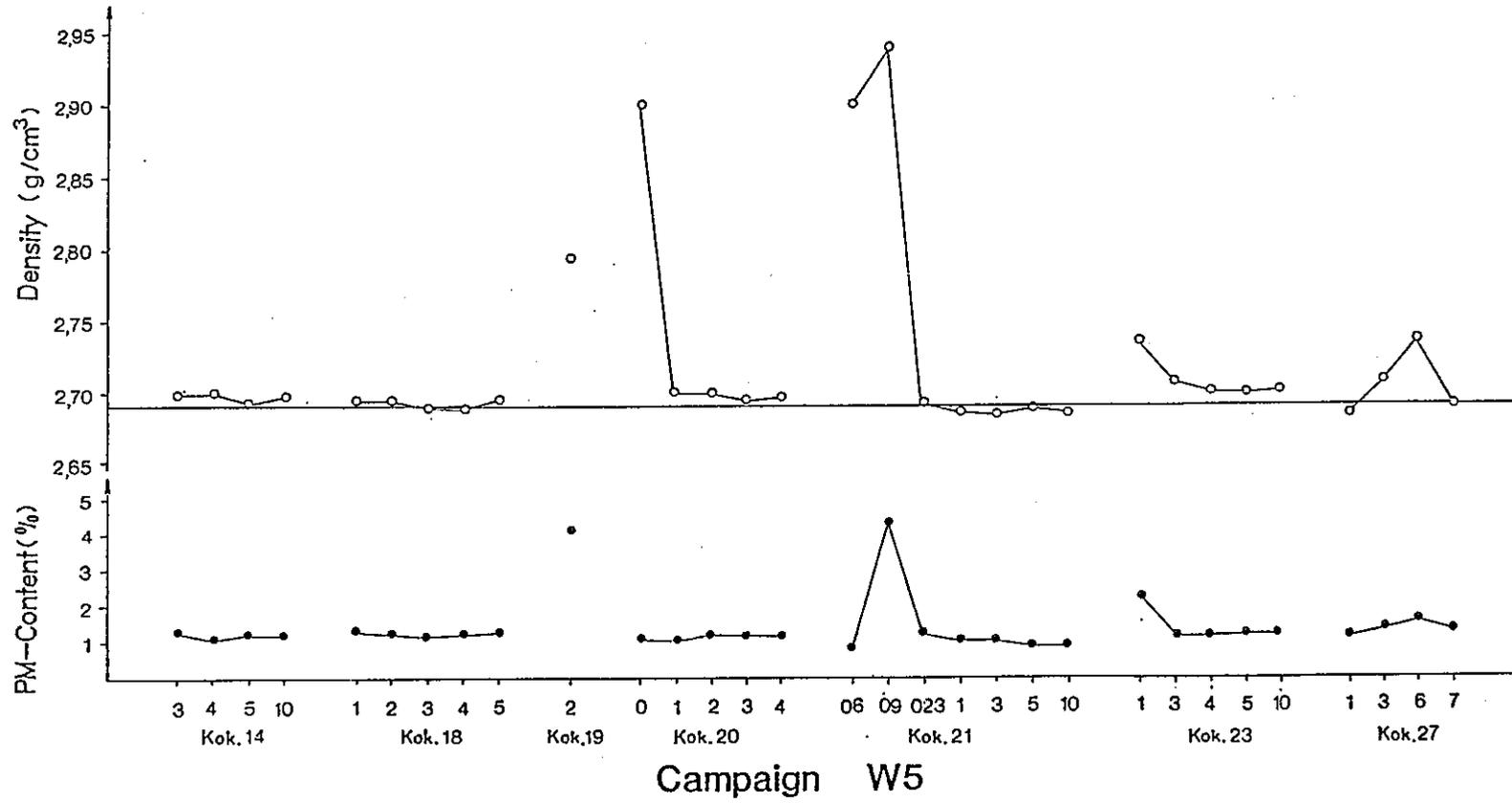


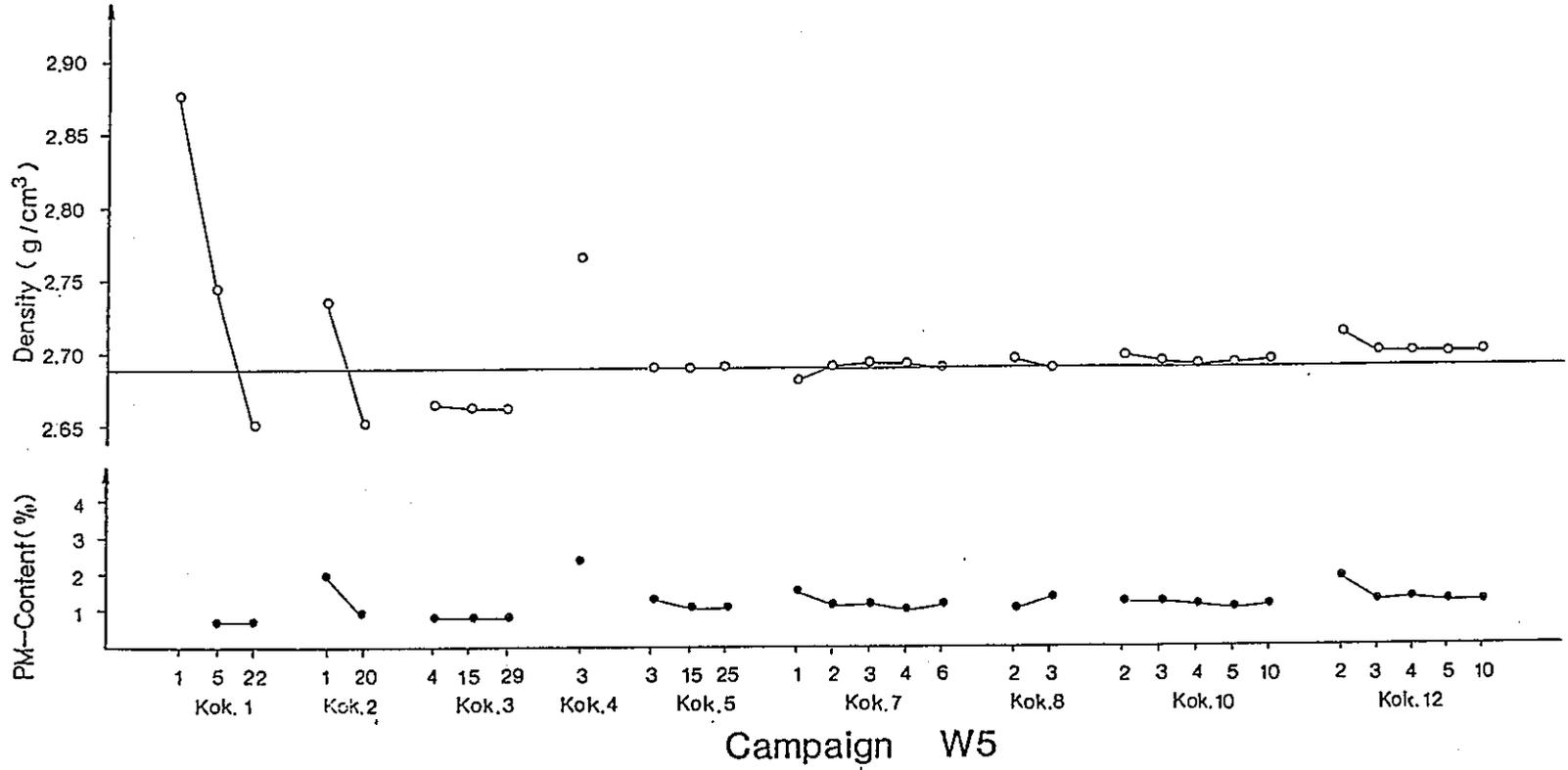
Abb.13: Elektrischer Widerstand von EM-Sedimenten in Laborgläsern bei 1150°C nach Schmelzzeiten von 4,24 und 100 h und Schmelztemperaturen von 1150°C und 1250°C

Table : Density and electrical resistance of fine-grained Pt-metal sediments in laboratory glass products after melting times of 4,24 and 100 h and melting temperatures at 1150°C and 1250°C

Glass Sample	PM-Content wt.%	Melting Time h	Density of PM-Sediment (at RT)		Electr. Resistance of PM-Sediment (at 1150°C)	
			1150	Melting Temperature (°C) 1250	1150	1250
1. Glass Frit + Ru,Rh,Pd	6.6	4	2.75	2.79	2.05	1.60
		100	2.90	2.90	0.85	0.85
2. Glass Prod.W4a + Ru,Rh,Pd	6.6	4	2.83	2.83	2.15	1.75
		24	2.84	2.85	1.05	1.35
		100	2.94	3.10	0.90	0.70
3. Glass Prod.W4 + Ru,Rh,Pd	0.5	4	-	2.97	-	0.80
	6.6	24	3.0	-	1.1	-
		100	2.95	3.10	1.0	0.85
4. Glass Prod.W4a + Ru	3.2	4	2.77	2.82	2.3	1.60
		24	2.77	-	1.4	-
		100	2.79	2.75	1.2	1.40
5. Glass Prod.W4 + Ru	0.5	4	2.80	2.82	2.0	1.60
	3.2	24	2.89	-	1.3	-
		100	2.90	2.91	0.8	0.75
6. Glass Prod.W4 + Ru,Pd	0.5	4	2.85	2.84	1.60	1.20
	5.9	24	-	-	-	-
		100	2.94	2.94	1.15	1.15



- 40 -



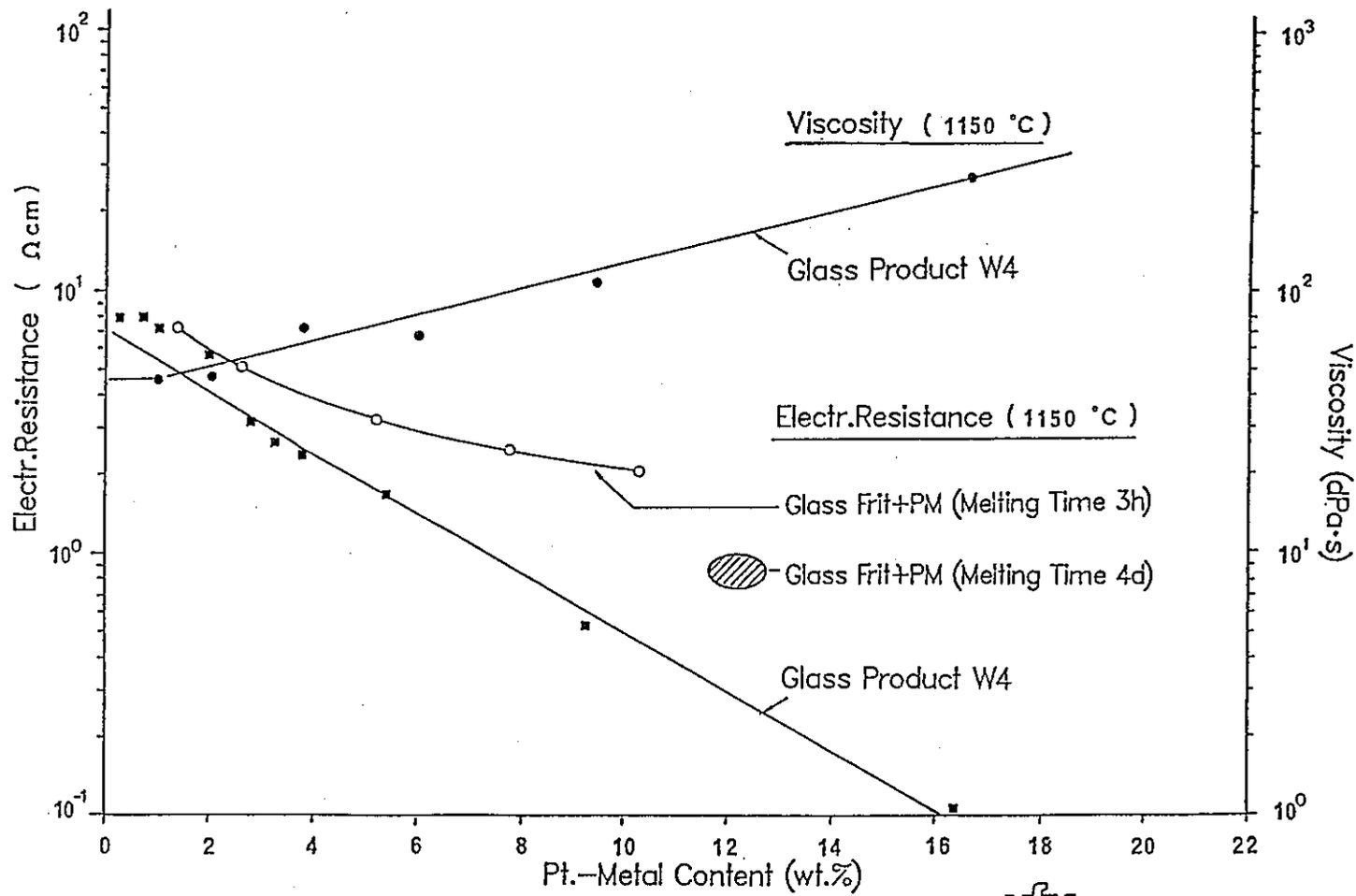
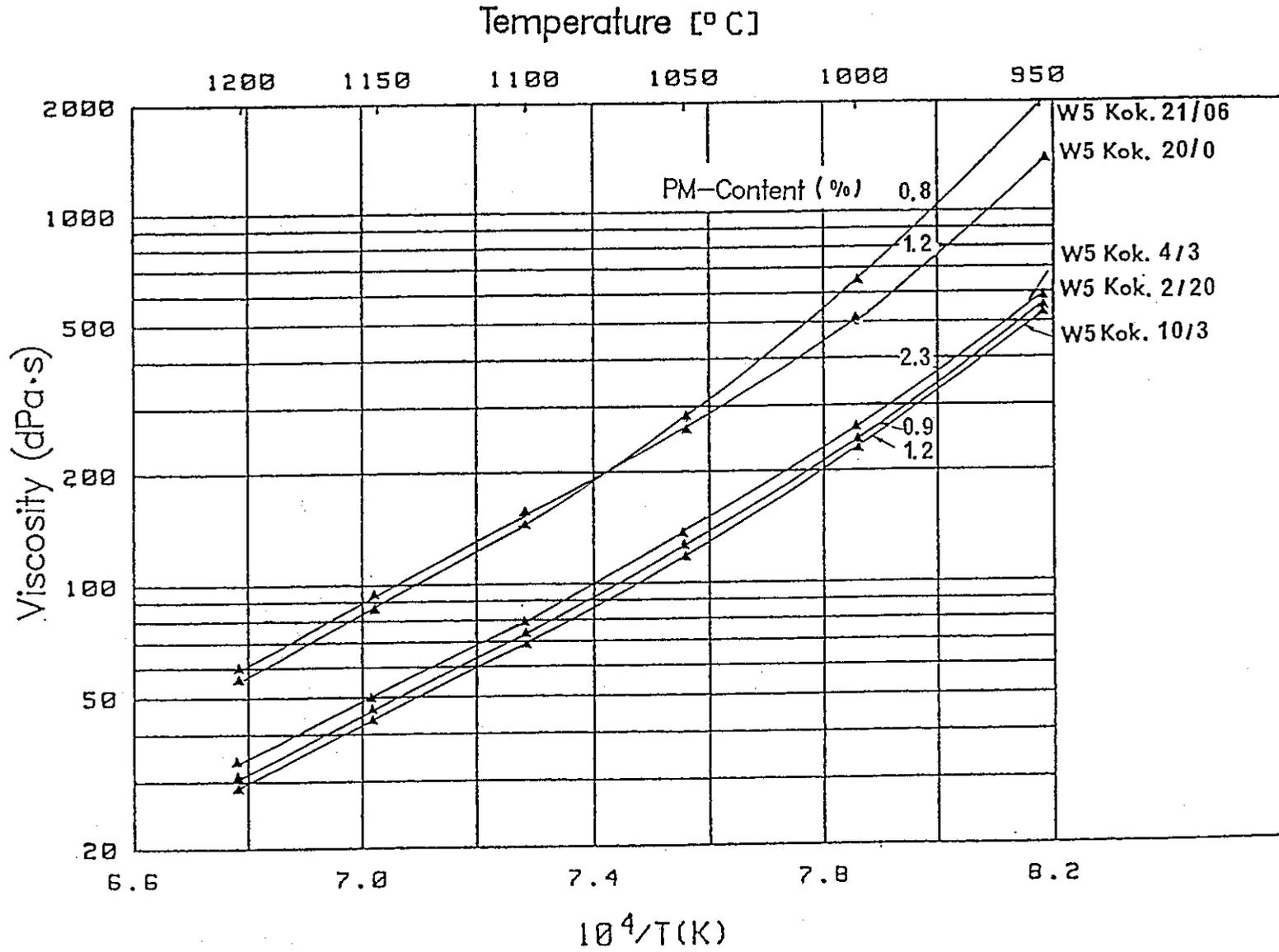


Abb. 7: Viskosität und elektrischer Widerstand bei 1150°C der Glasproben aus Verglasungskampagne W4 in Abhängigkeit vom EM-Gehalt.
 Zum Vergleich der Verlauf des elektrischen Widerstands bei EM-haltigen Laborgläsern





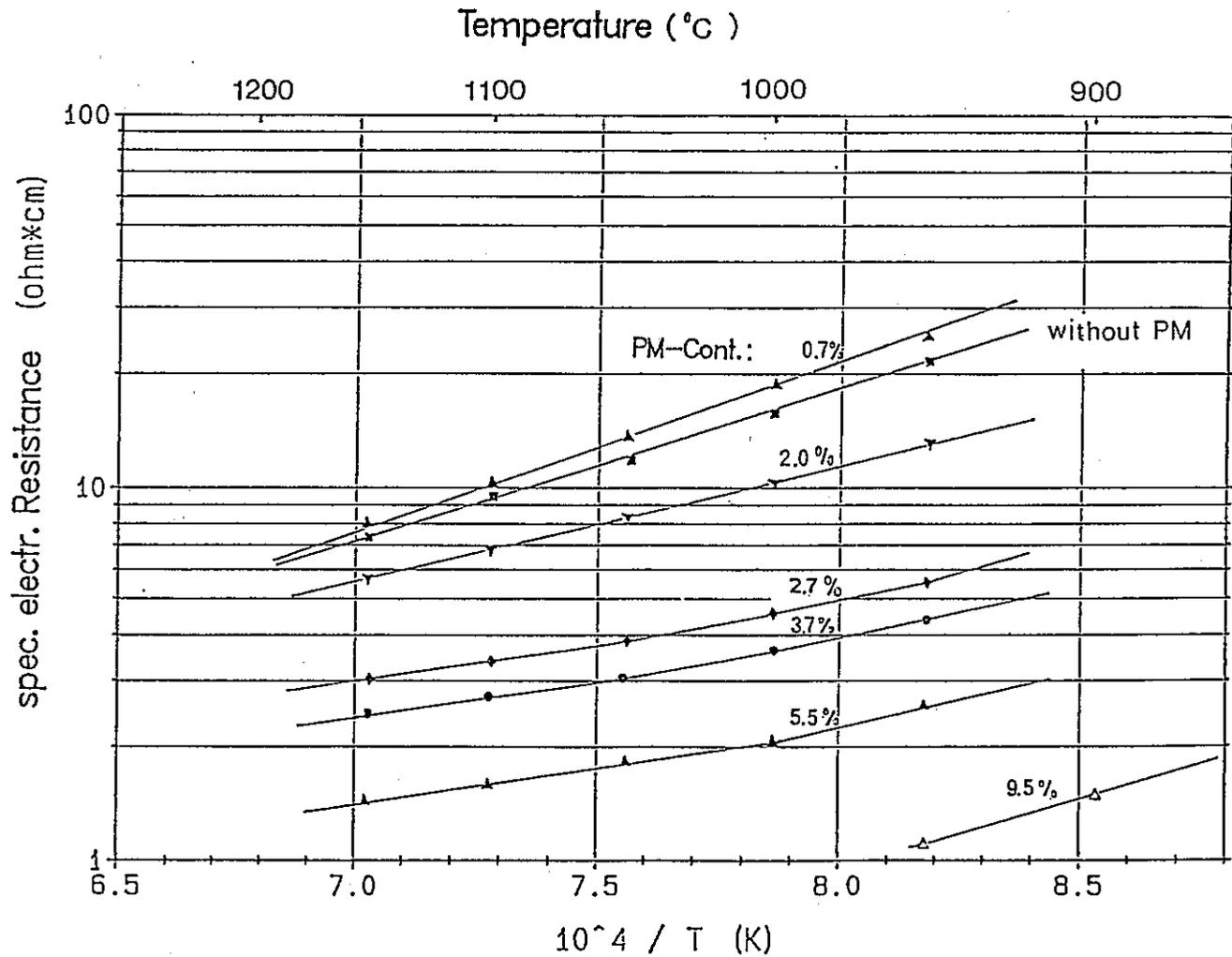


Abb. 6: Spezifischer elektrischer Widerstand von Glasproben mit unterschiedlichem EM-Gehalt aus der Verglasungskampagne W4

3.44-2

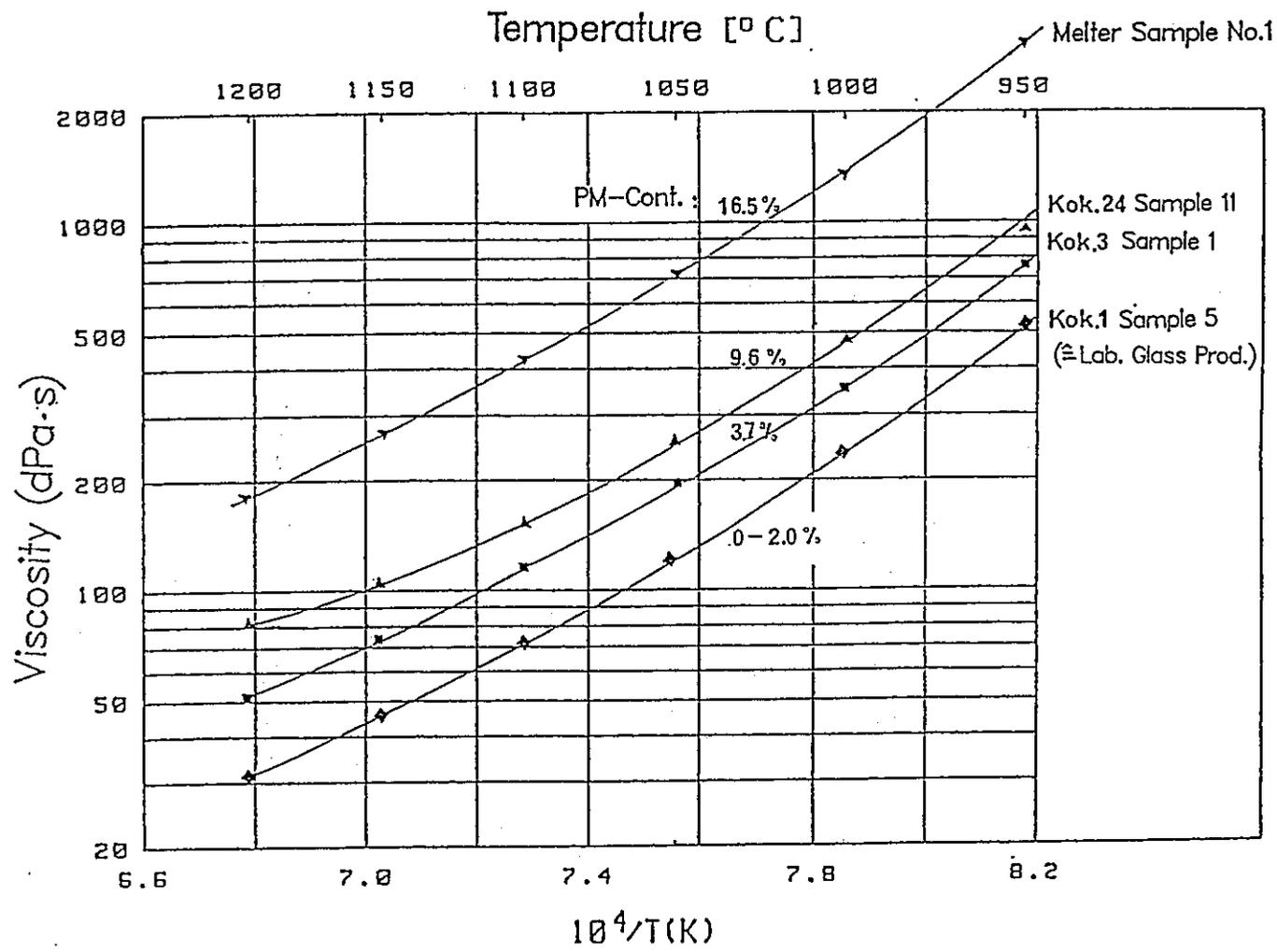
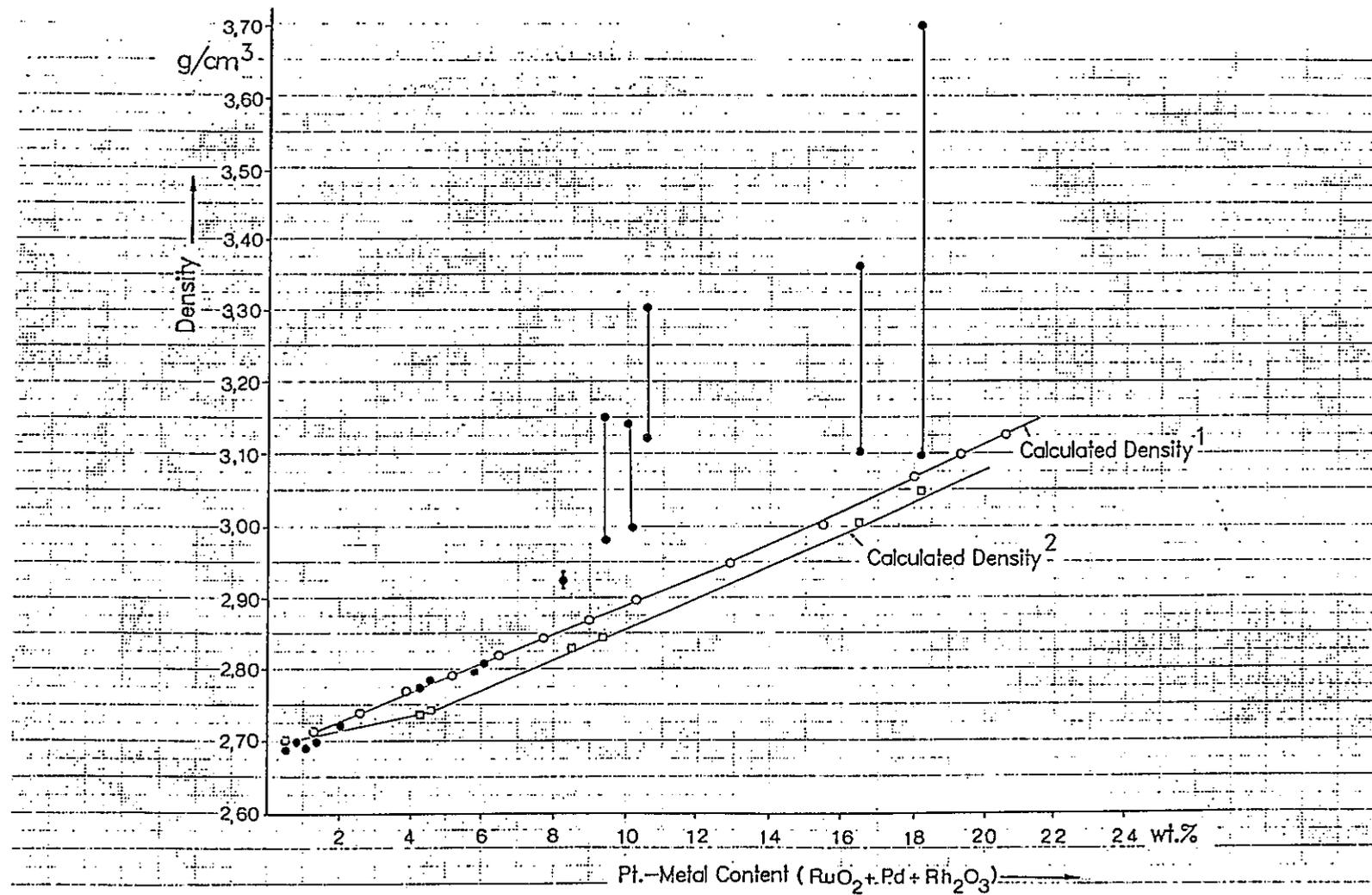


Abb. 5: Viskoositätskurven von Glasproben mit unterschiedlichem EM-Gehalt aus der Verglasungskampagne W4

3.4.4-1



3.4.3-3

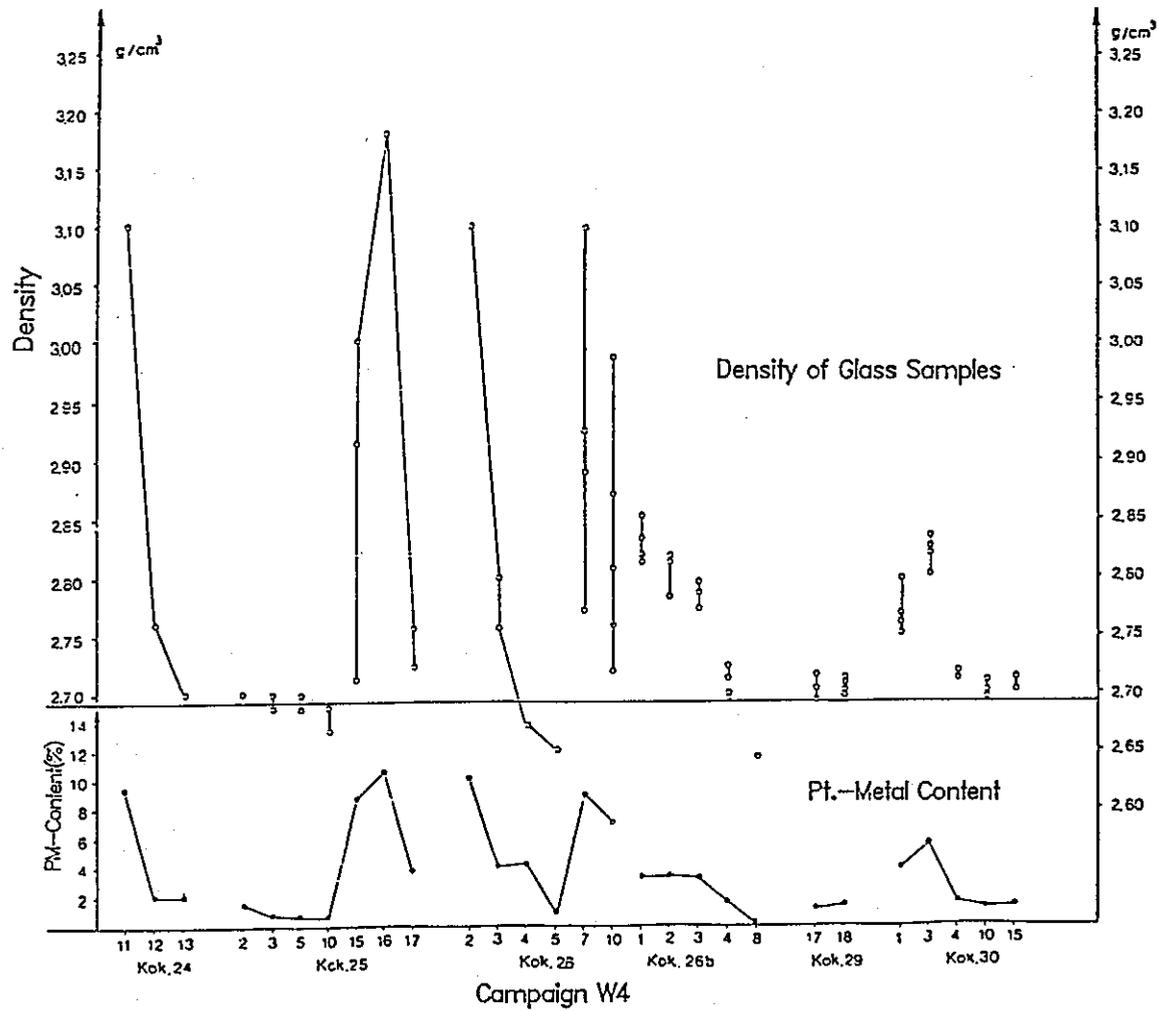


Abb. 3: Dichte bei RT von Glasproben aus der Verglasungskampagne W4 und deren analysierten EM-Gehalt

Nobel Metal Vitrification Campaigns

1987 Campaign W4 with Ru, Rh and Pd

1988 Campaign W5 with Ru and Pd
(air bubbling in the glass melt)

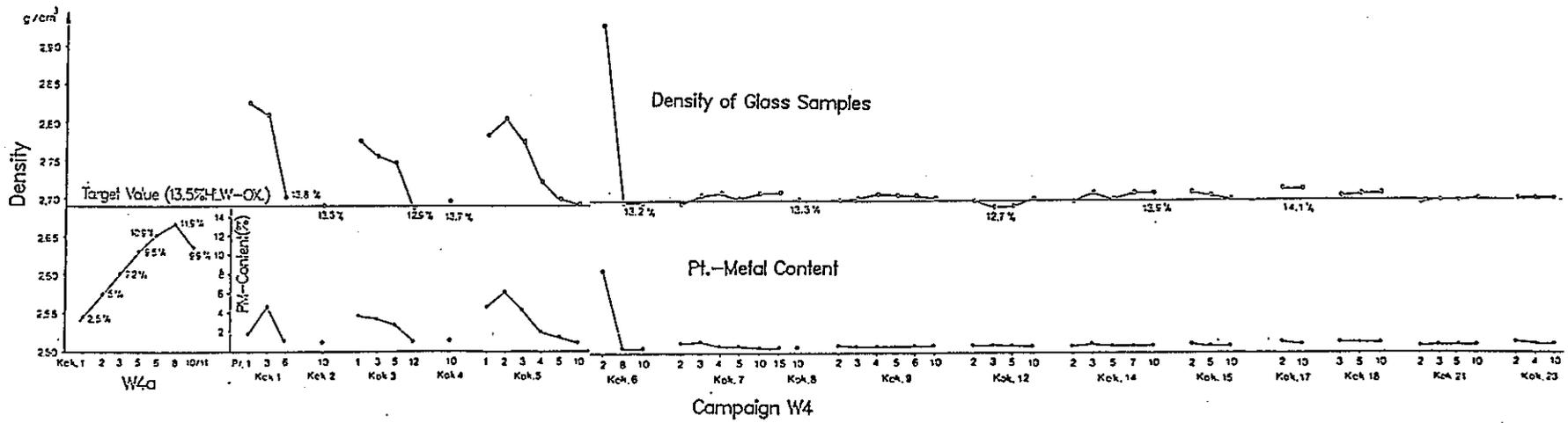


Abb. 2: Dichte bei RT von Glasproben aus der Verglasungskampagne W4 und deren analysierten EM-Gehalt

1.4 Behaviour of ruthenium and cesium in the off-gas system during the noble metal campaigns W4 and W5

BEHAVIOUR OF RUTHENIUM AND CESIUM IN
THE OFF-GAS SYSTEM DURING THE NOBLE
METAL CAMPAIGNS W4 and W5

for

the 9th Annual PNC-KfK Meeting on High-Level
Waste Management held at KfK/INE

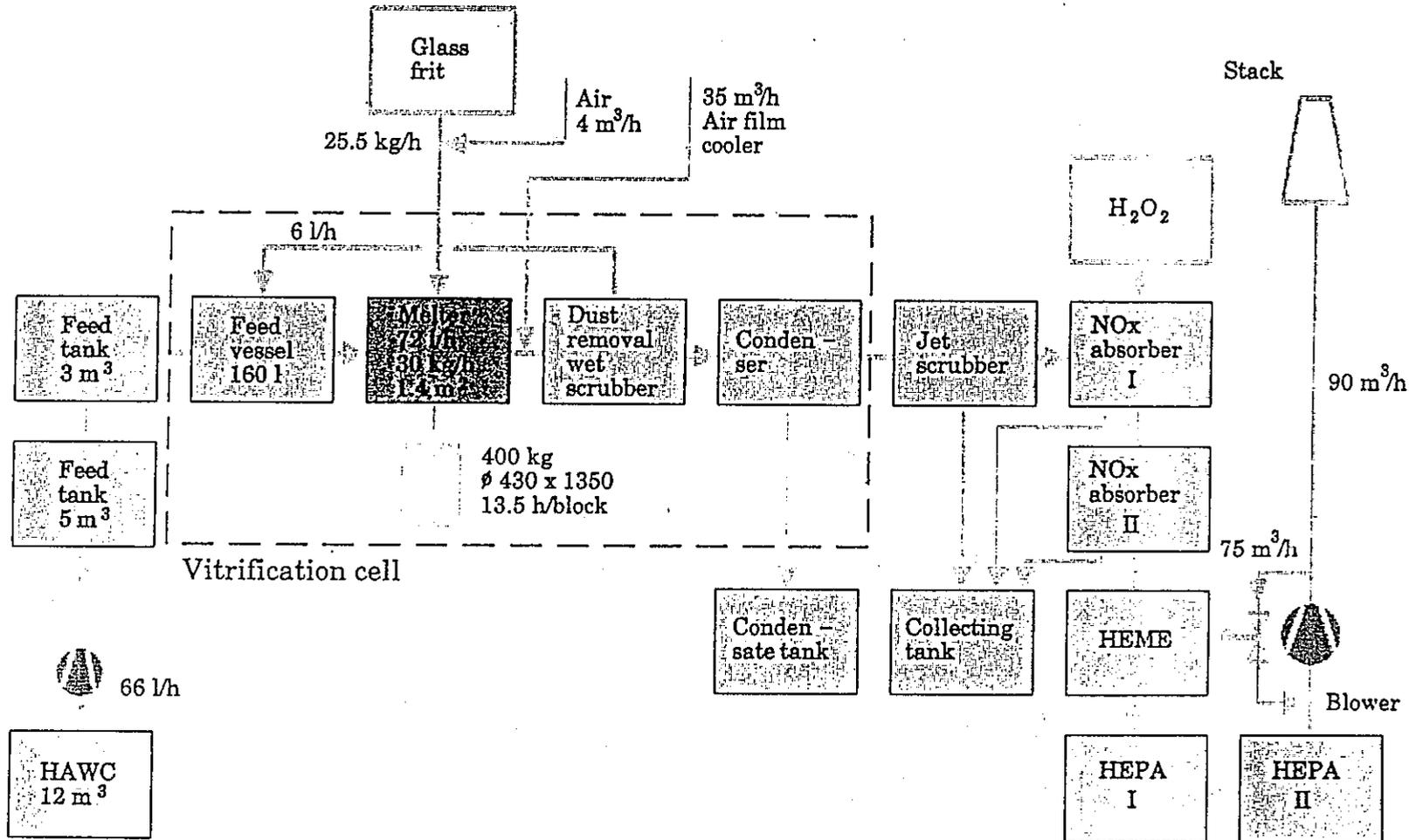
October 1989

- * Description V-W1
off-gas system

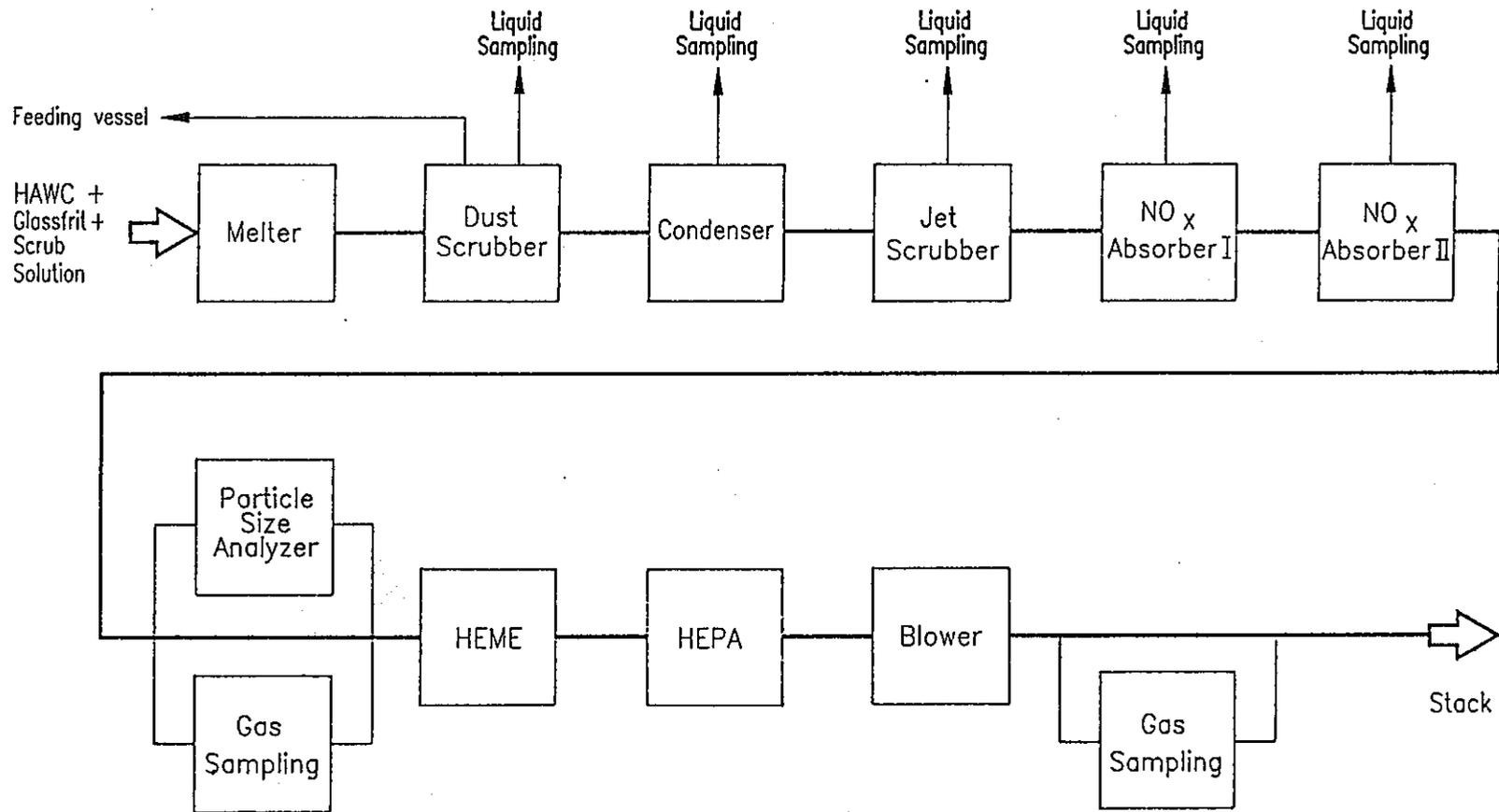
- * Noble metal campaigns
W4 and W5
 - * Basic data
 - * Operation conditions

- * Balances of Ruthenium
and Cesium

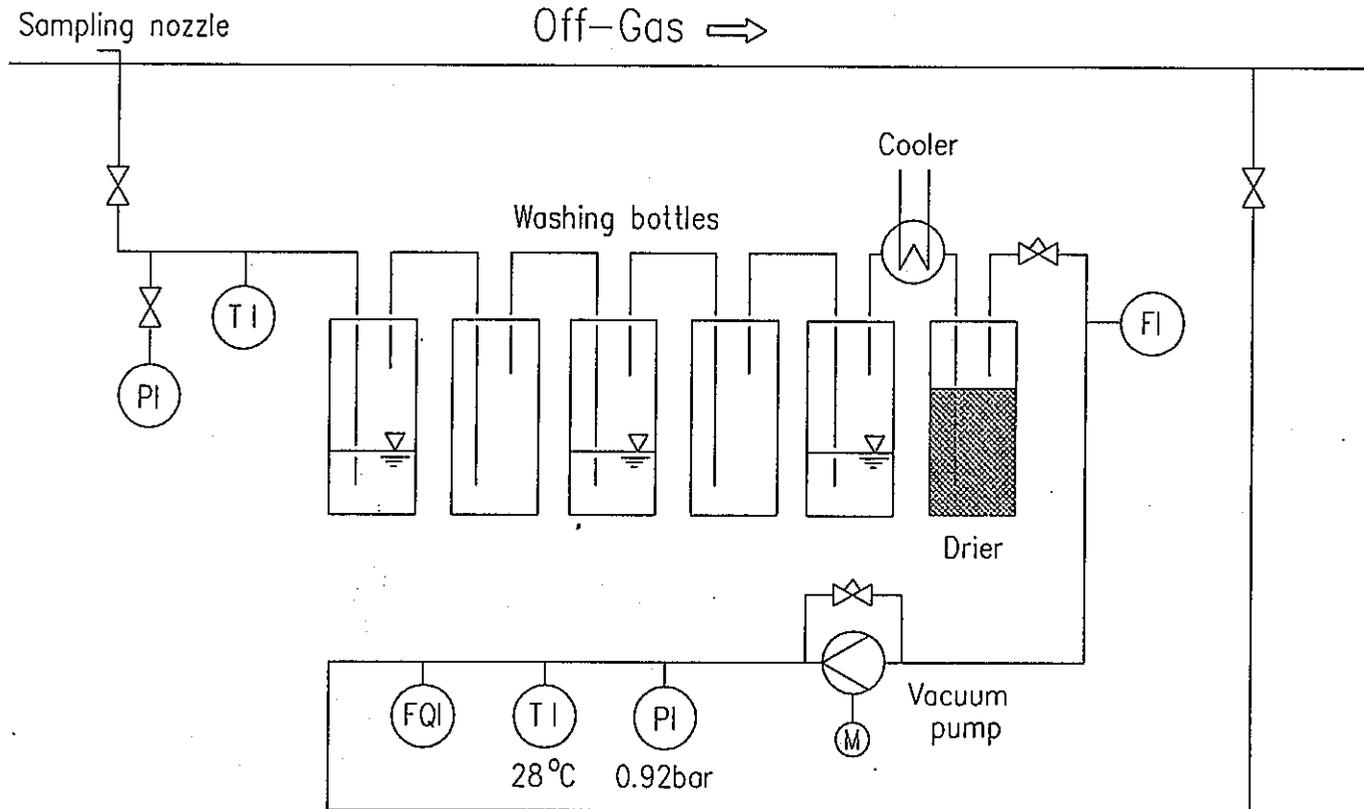
- * Decontamination factors of
melter and off-gas
components for Ru and Ce



Basic flowsheet mock-up plant V-W1 for WAW-Wackersdorf



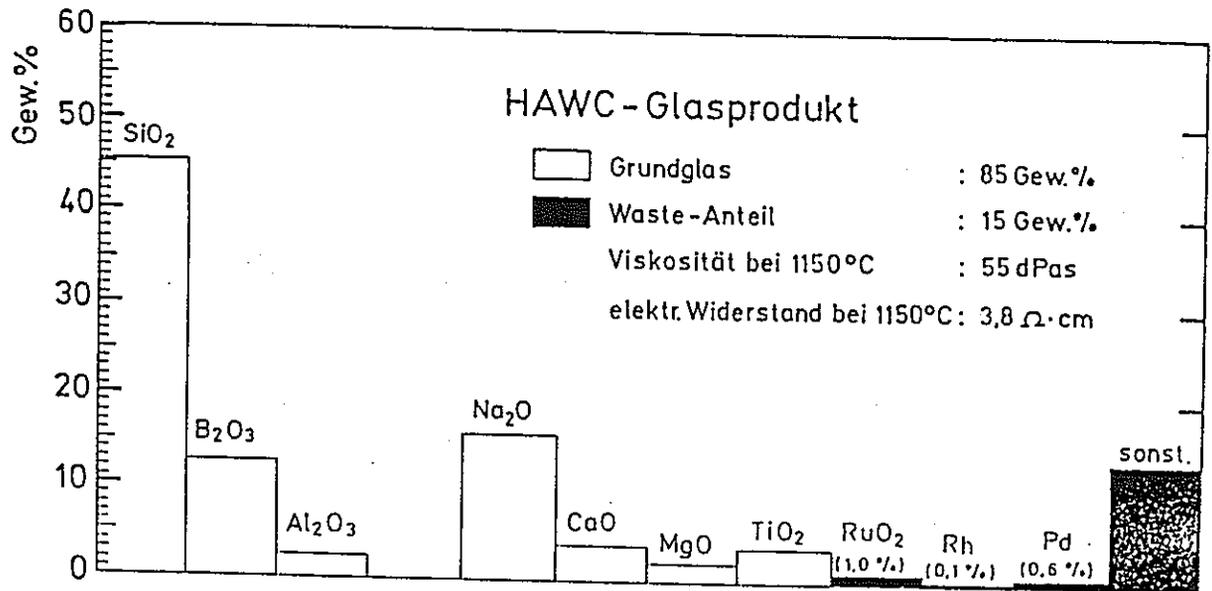
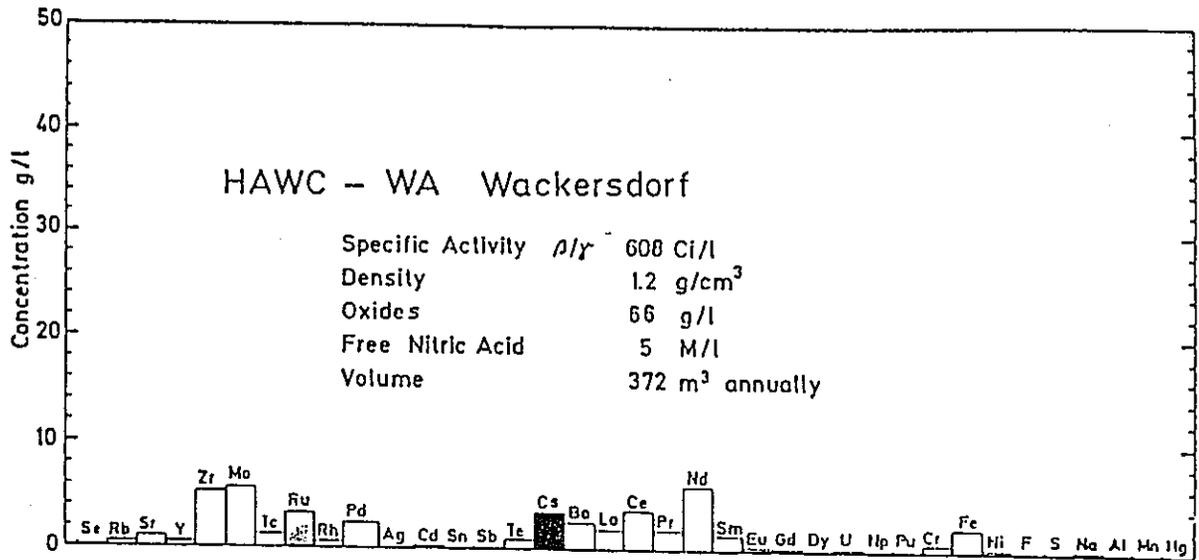
V-W1 OFF-GAS SAMPLING SYSTEM

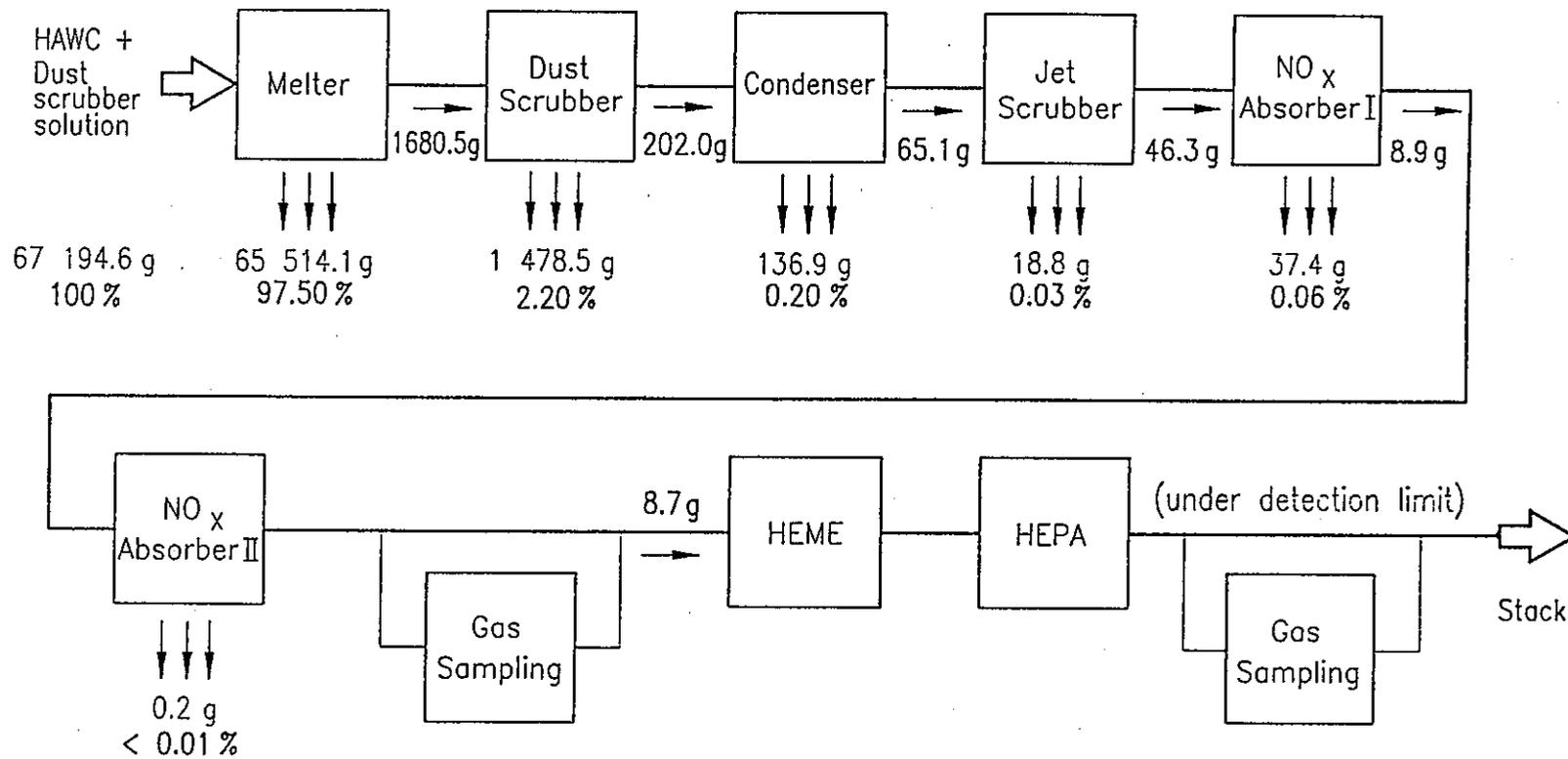


GAS SAMPLING ARRANGEMENT

PARAMETER		W4	W5
Feed rate	l/h	45-60	40-50
Power input	kW	75-90	75-85
Plenum heating	kW	-	15
Bubbling	m ³ /h	-	2 × 0.8
Process conditions		complete pool coverage	optimal coverage
Central temperature	°C	1150	1150
Plenum temperature	°C	300-450	450-600
Off-gas temperature (scrubber entrance)	°C	115-150	120-160
Quantities of			
Ruthenium	kg	73	66
Cesium	kg	122	111

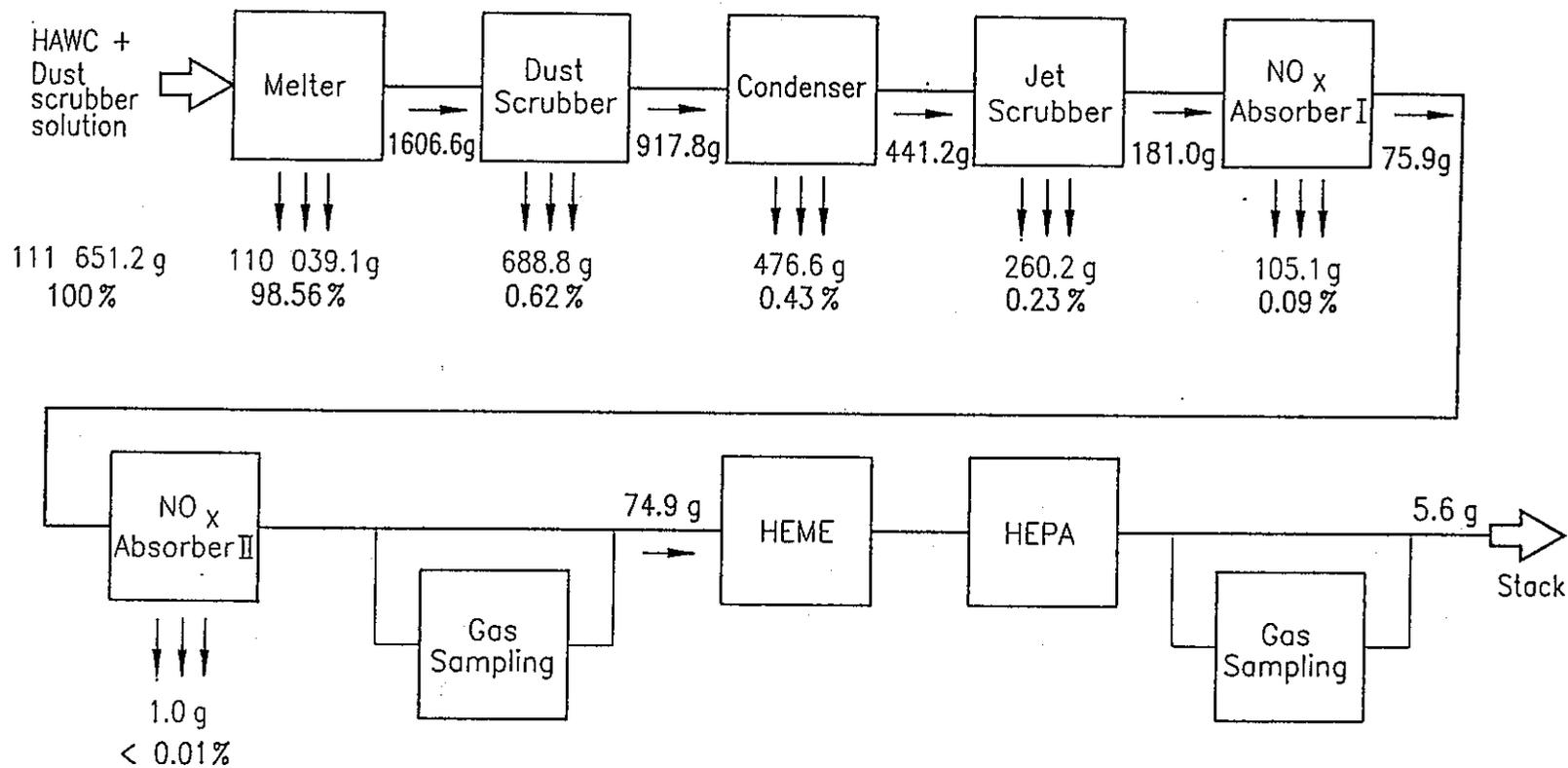
OFF-GAS RELEVANT PARAMETERS OF THE NOBLE METAL CAMPAIGNS



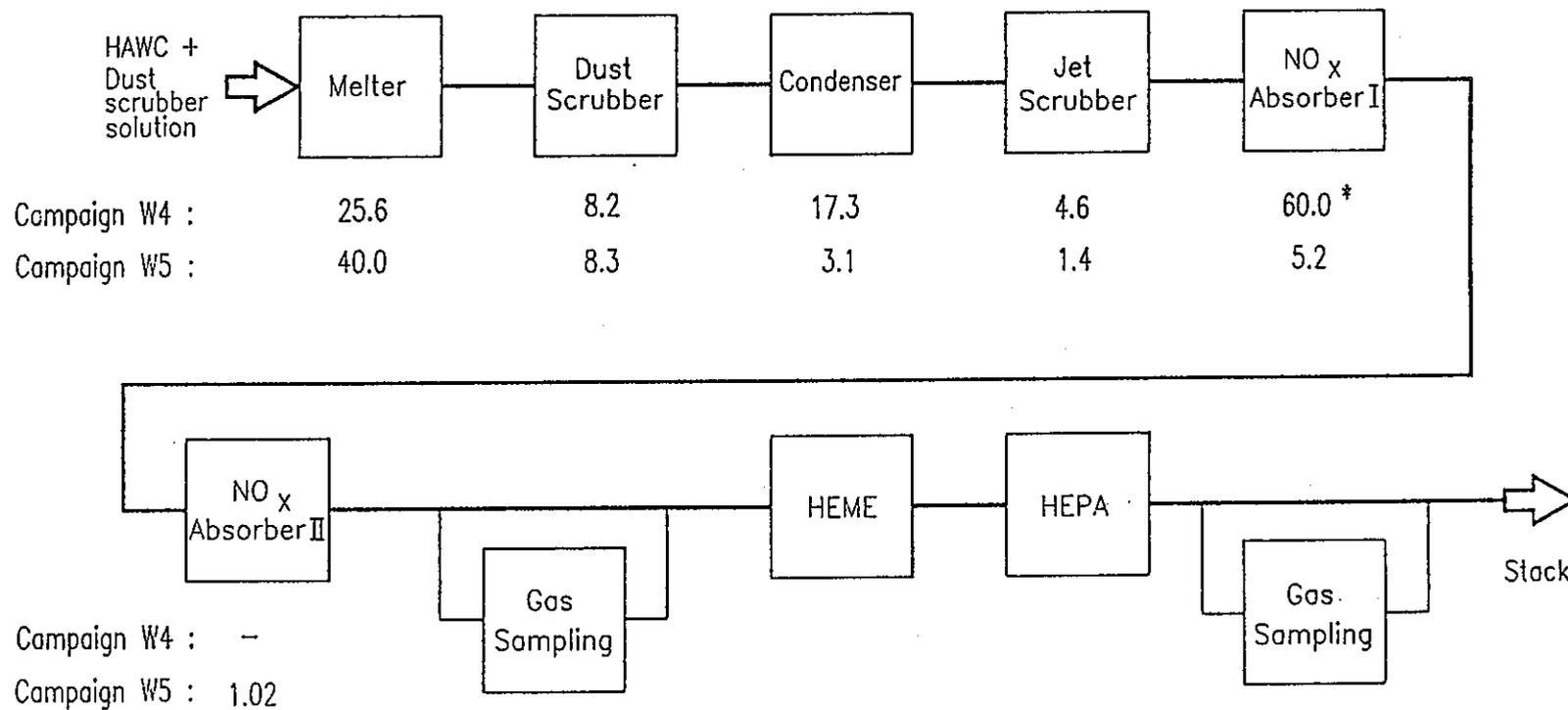


- 57 -

RUTHENIUM BALANCE DURING THE NOBLE METAL CAMPAIGN W5 (January-March 1989)

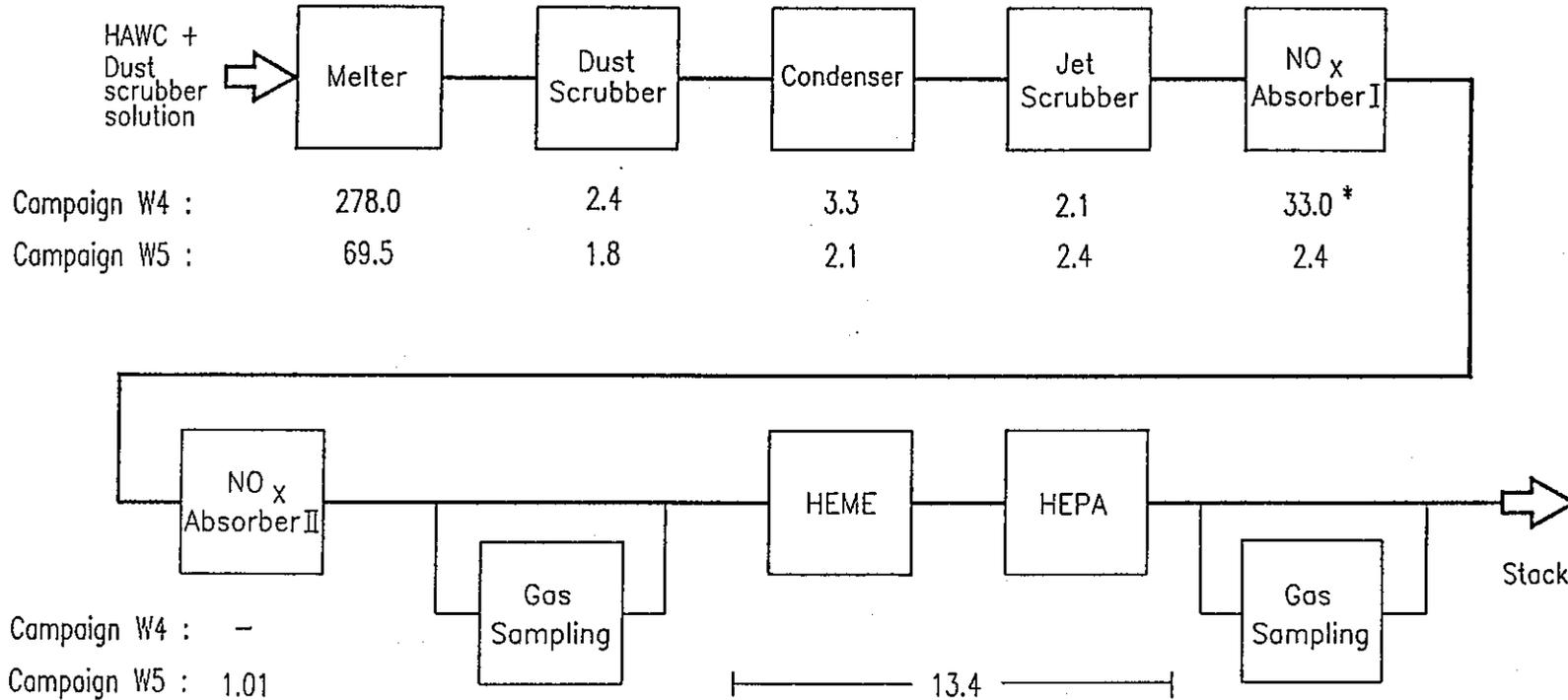


CESIUM BALANCE DURING THE NOBLE METAL CAMPAIGN W5 (January-March 1989)



* high DF, because no analytical data available after NO_x-Absorber II

DECONTAMINATION FACTORS OF MELTER AND OFF-GAS COMPONENTS FOR RUTHENIUM INVESTIGATED DURING THE NOBLE METAL CAMPAIGNS



* high DF, because no analytical data available after NO_x-Absorber II

DECONTAMINATION FACTORS OF MELTER AND OFF-GAS-COMPONENTS FOR CESIUM INVESTIGATED DURING THE NOBLE METAL CAMPAIGNS

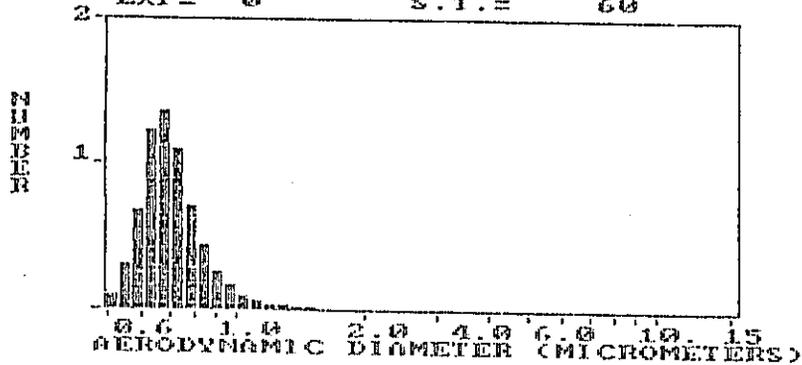
			Ruthenium	Cesium
W4	Total DF	Melter to NO _x - Absorber I	1.0×10^6	1.5×10^5
W5	Total DF	Melter to NO _x - Absorber I	3.9×10^5	1.2×10^5
		Melter to NO _x - Absorber II	7.6×10^3	1.5×10^3

TOTAL DF'S FOR THE CAMPAIGNS W4 AND W5

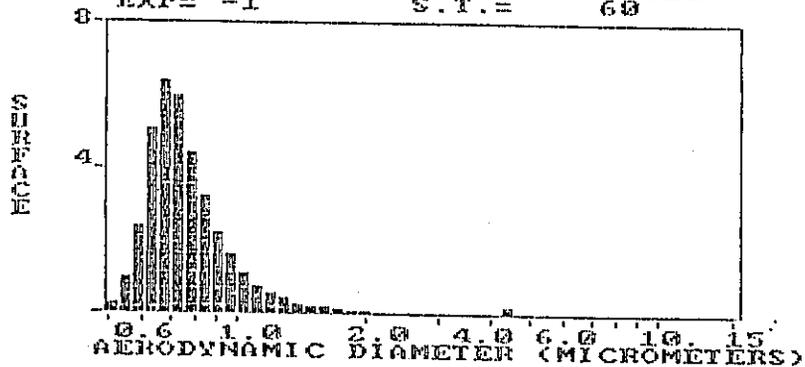
TSI AERODYNAMIC PARTICLE SIZER
W-5 12.26

08-10-1989 SAMPLE # 1
RESPIRABLE MASS 8.120491E-04 DENSITY: 3
DIL. RATIO: 1 : 1 EFFIC. CORRECT.: D1

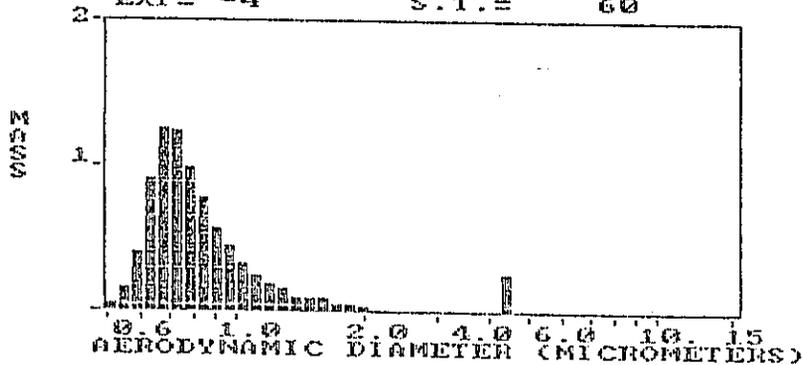
NUMBER CONC VS PARTICLE SIZE
EXP= 0 S.T.= 60



SURFACE CONC VS PARTICLE SIZE
EXP= -1 S.T.= 60



MASS CONC VS PARTICLE SIZE
EXP= -4 S.T.= 60



PARTICAL SIZE DISTRIBUTION
after NO_x -Absorber II, Campaign W5

CONCLUSIONS

- * Satisfying melter DF's for Ruthenium and Cesium
- * Unsatisfying retention of Cesium in the wet cleaning system, especially the Jet scrubber
- * No more Ruthenium found behind the filters
- * Some amount of Cesium also detected after the dry cleaning system
- * Backcalculation of the balances without concentration measurements in the filter area unreliable

- 1.5 Vitrification of HAWC of the WAK plant, The new inactive test facility VA-WAK with the melter K-6

Vitrification of HAWC of the WAK plant

The new indicative test facility VA-WAK with the melter K-6'

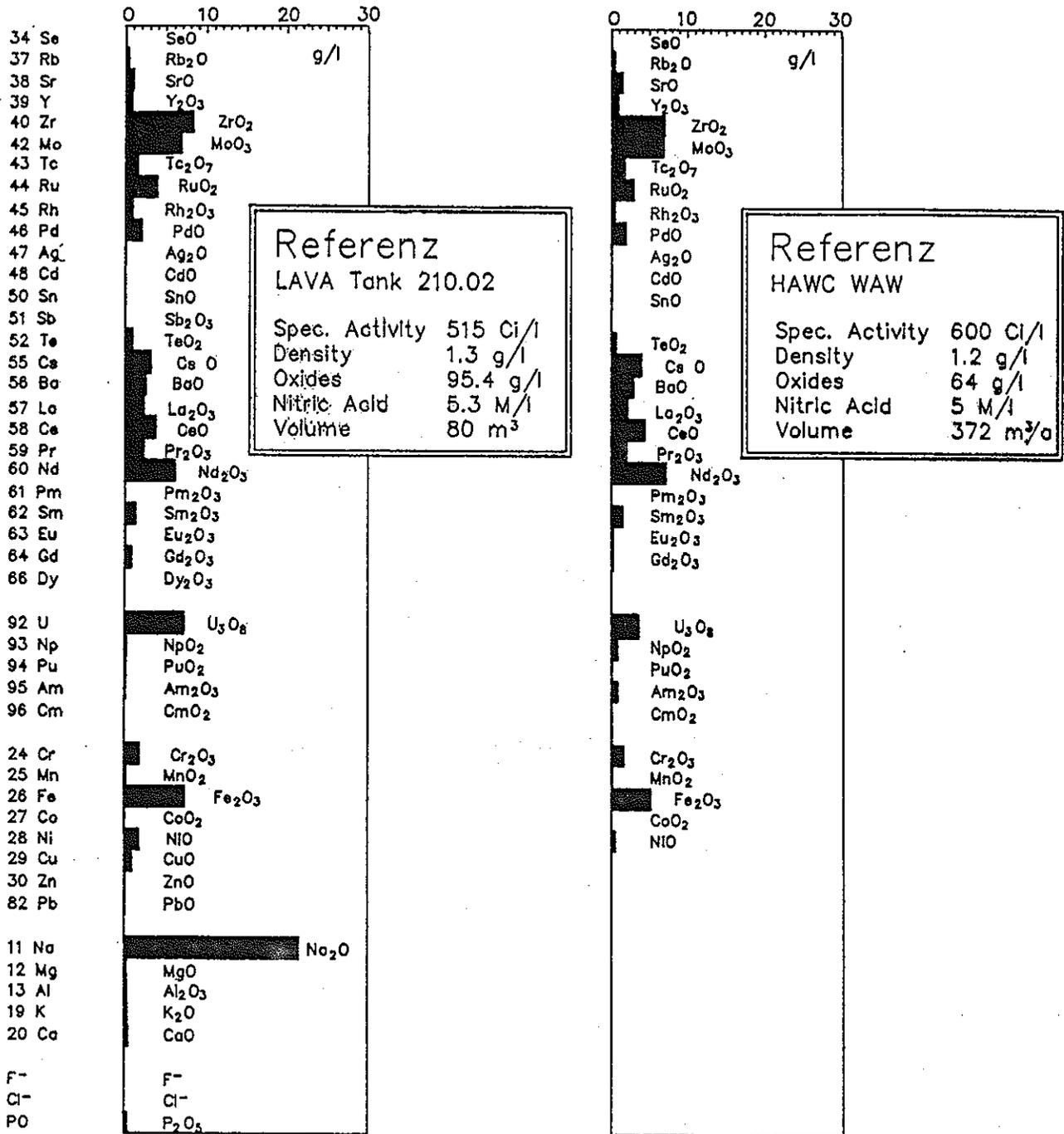
for

the 9th Annual PNC-KfK Meeting on High-Level
Waste Management held at KfK/INE

October 1989

Plant operation	Since September 1971
Shut-down	End of 1990
Reprocessed Uranium	204 to (expected)
HAWC-Volume	62 m ³ (Tank 210.02) 20 m ³ exp. (Tank 210.03)
Waste management concept	<ul style="list-style-type: none"> - Transportation of the HAWC-WAK to Mol/Belg. Vitrification by PAMELA (Start in 1993) - In case no Transportation licence will be given Vitrification in a new plant at KfK-site might be possible

Status of the reprocessing plant
Karlsruhe - WAK -



Comparison of HAWC-WAK to waste produced by industrial reprocessing

Objective

- Development of a melter type suitable for the vitrification of noble metal containing waste in the PAMELA plant at KfK-INE/Karlsruhe
- Development of a glass frit with possibly high waste loading at HMI/Berlin

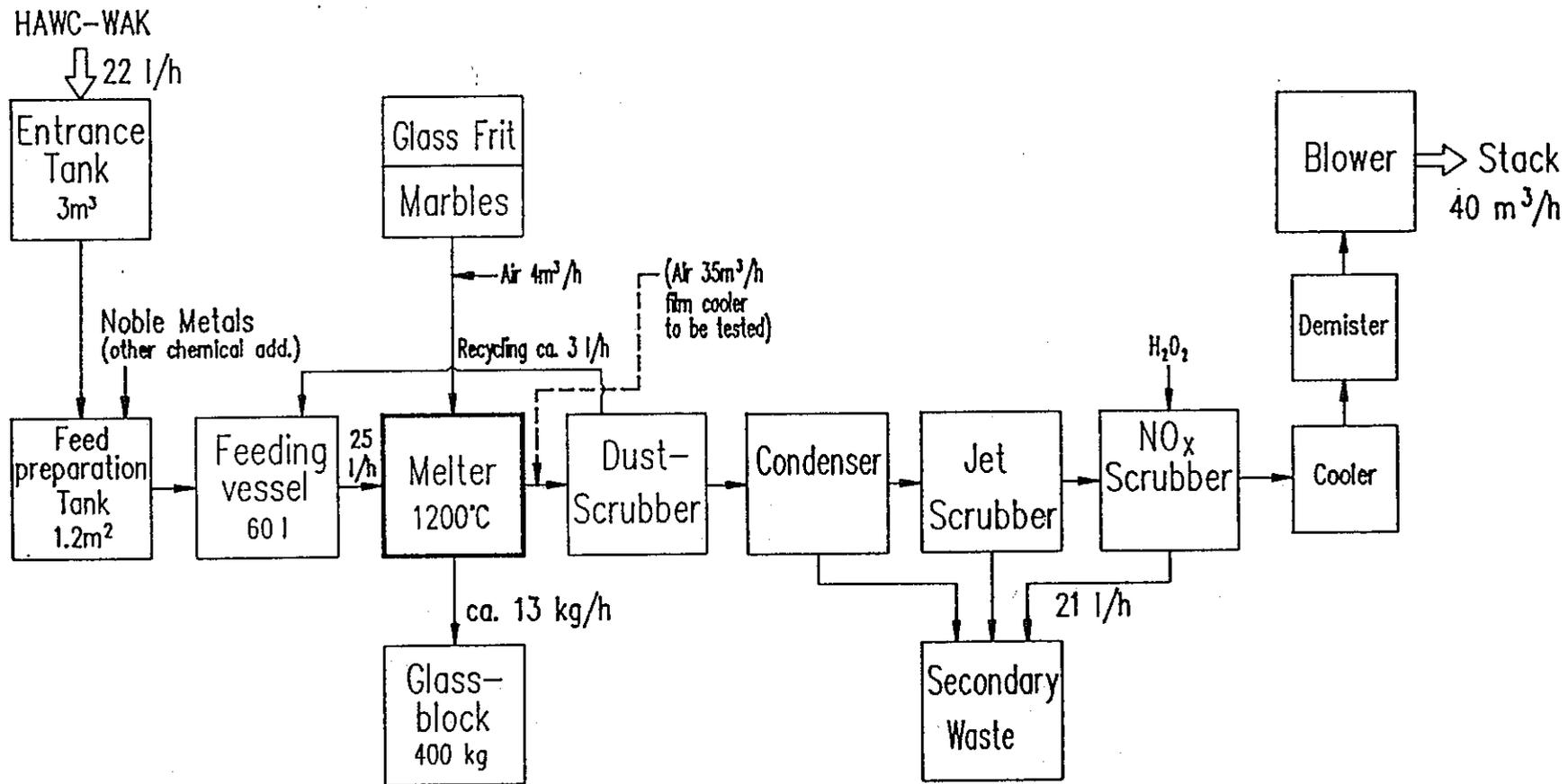
Schedule

- Vitrification test runs with the inactive melter K-6' until 1991
- Construction of the active melter K-6 until end of 1992
- Start Hot operation in 1993

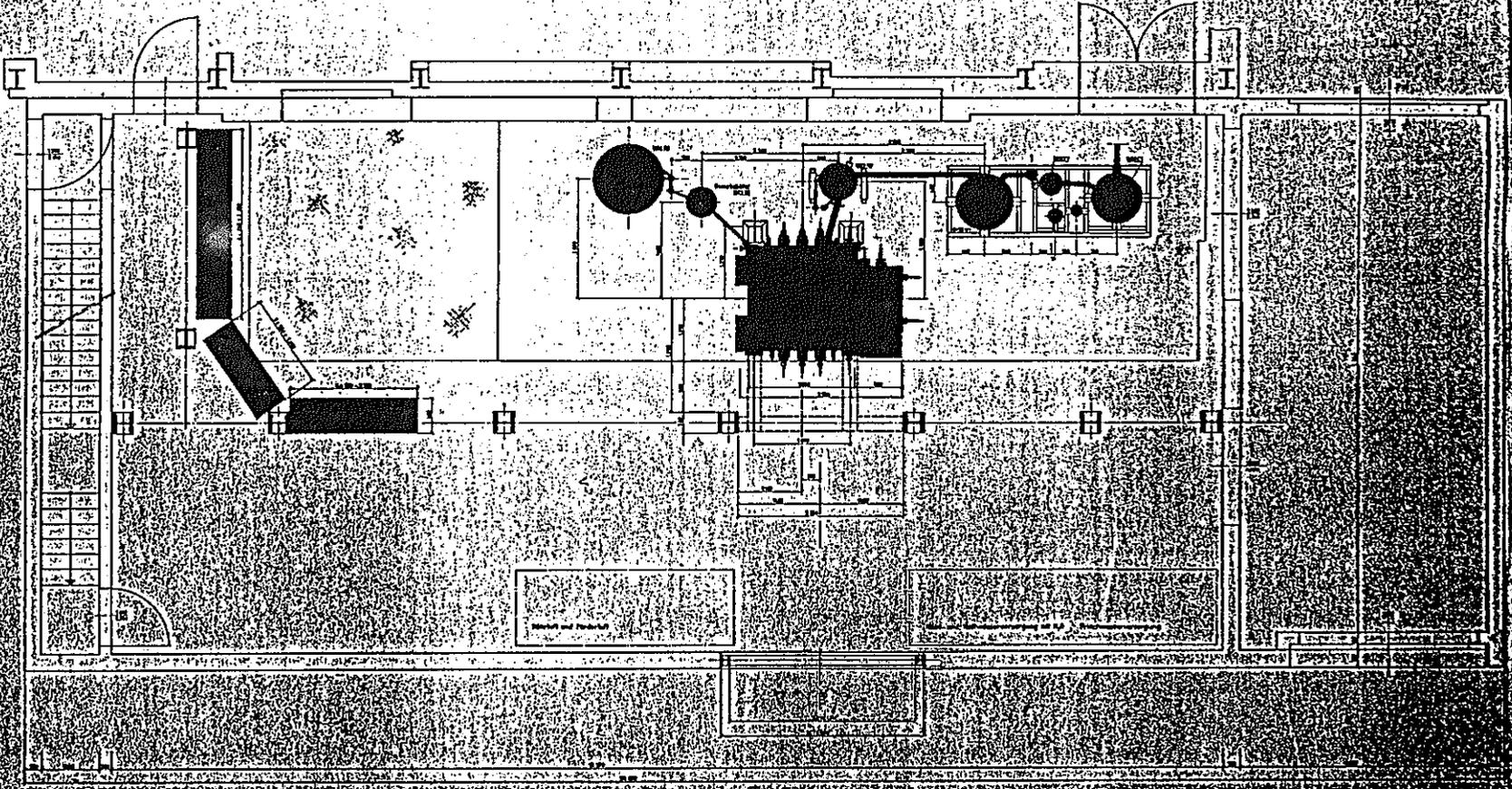
HAWC-WAK Technology Program

Objective: Vitrification plant VA-WAK	1990												1991												1992											
	Ja	Fe	M8	Ap	Ma	Ju	Ju	Au	Se	Ok	No	De	Ja	Fe	M8	Ap	Ma	Ju	Ju	Au	Se	Ok	No	De	Ja	Fe	M8	Ap	Ma	Ju	Ju	Au	Se	Ok	No	De
Completion of the plant construction	—																																			
Plant test operation	—																																			
Noble metal campaign I				—																																
Operation report I				—																																
Noble metal campaign II							—																													
Operation report II							—																													
Design K-6 for <u>radioactive</u> operation										—																										
Documents for K-6 order																—																				
Construction of melter components																			—																	
Operation tests K-6' (glass quality ..)																			—			—														
Manual for melter operation																						—														
Test operation K-6 in PAMELA																															→					

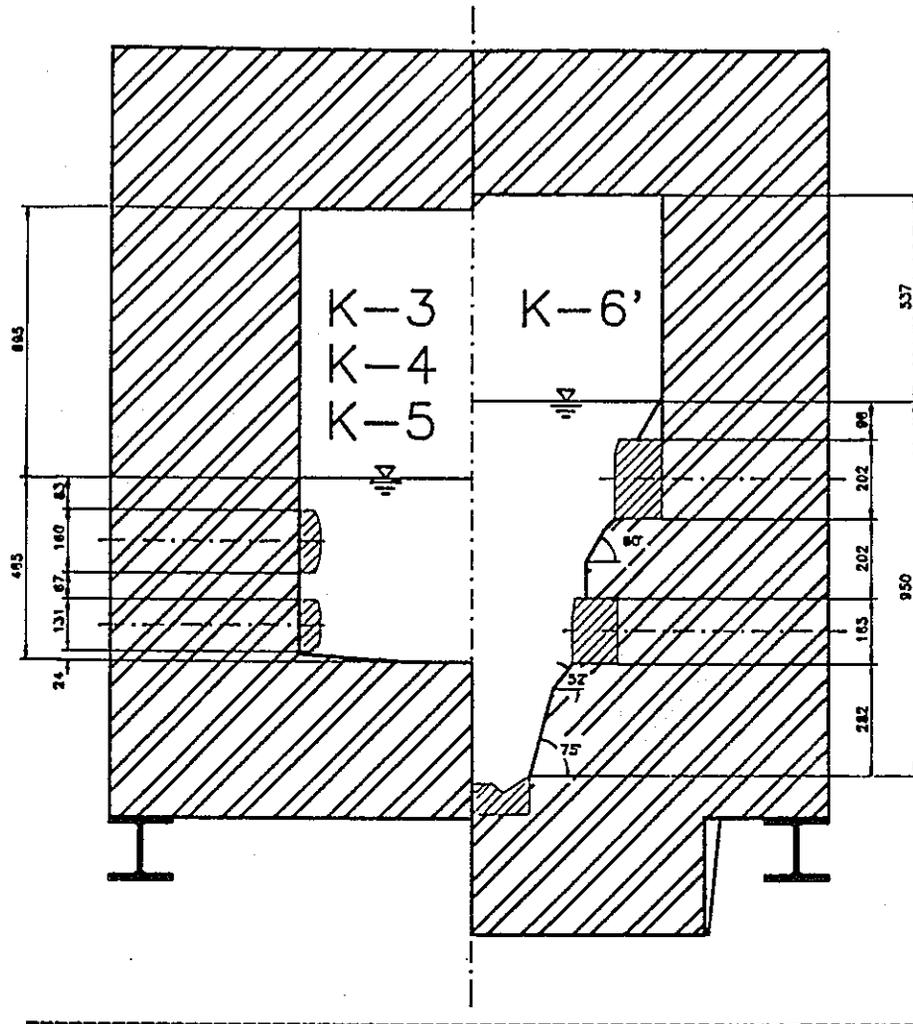
Time schedule of INE activities within the HAWC-WAK technology program



Basic flowsheet mock-up plant VA-WAK for vitrification of HAWC-WAK

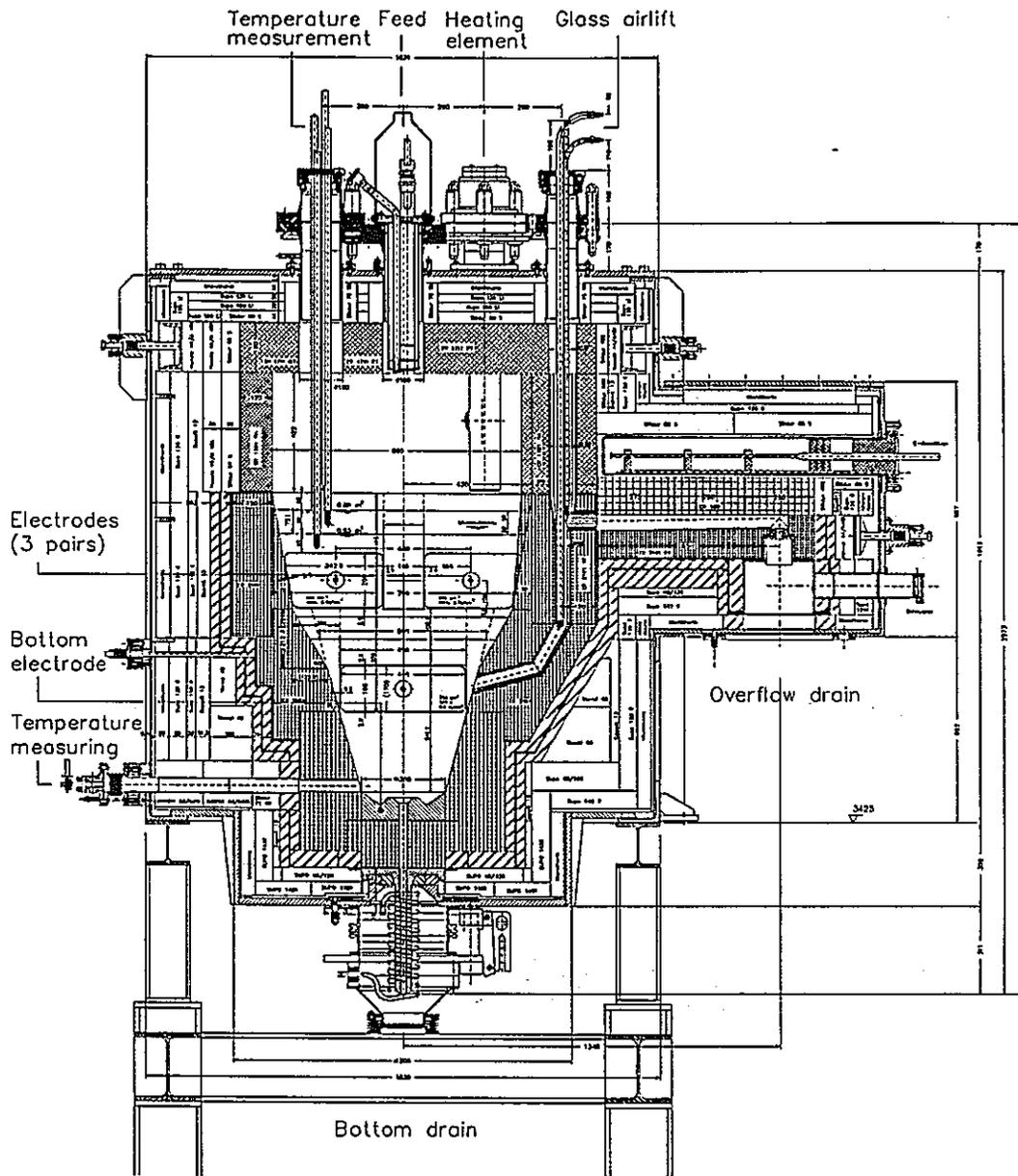


Komponentenanordnung Anlage VA-2

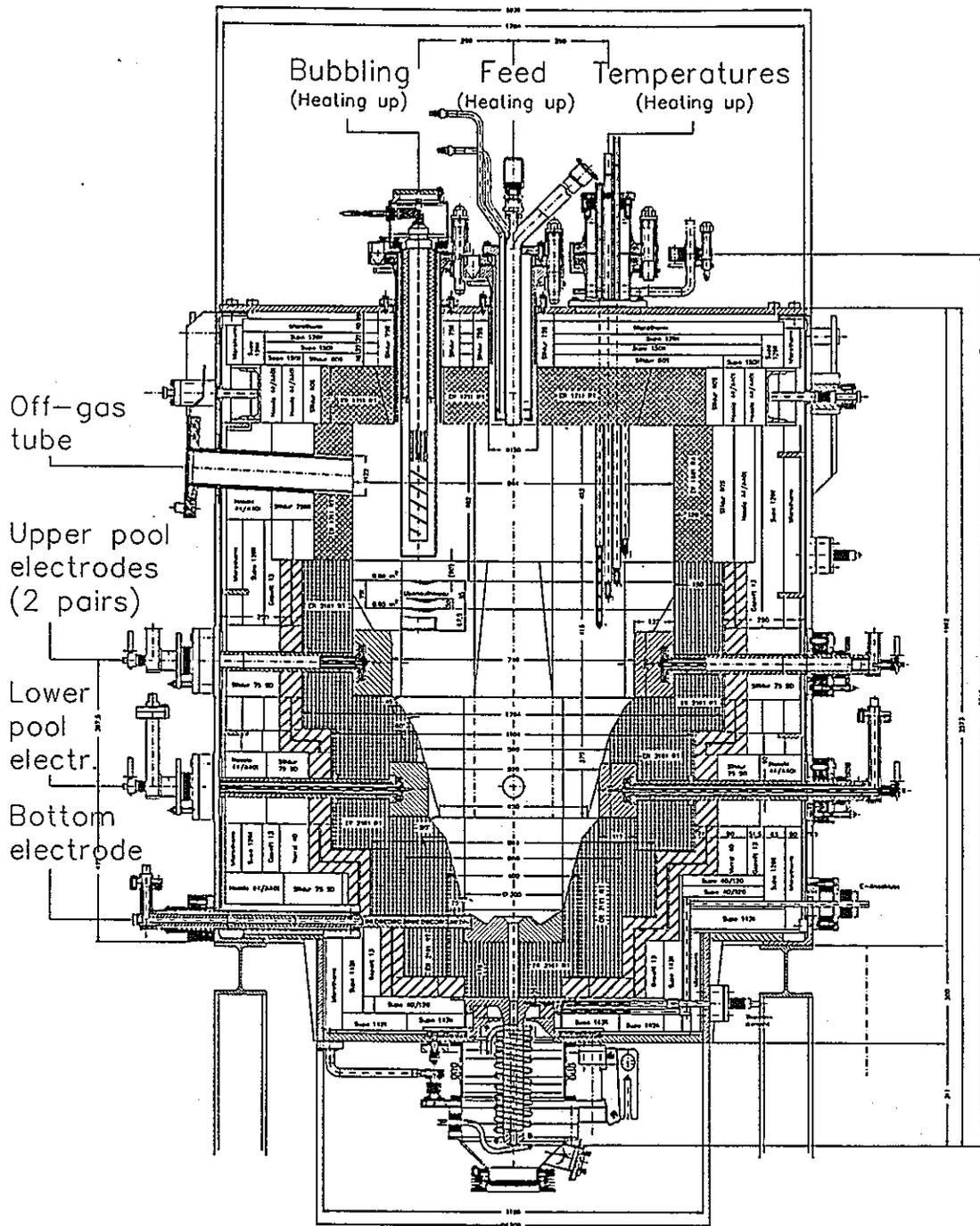


Melter designation	K-3, K-4, K-5	K-6'
Waste type	LEWC, HEWC	HAWC-WAK sim.
Glass pool surface	0.72 m ²	0.88 m ²
Glass pool volume	300 l	ca. 400 l
Discharge volume	60 l (3 x 50 l)	2 x 75 l
Start-up technique	20 MoSi Elements	5 SiC Elements
Containment Height	2.6 m	2.9 m
Total weight	18 to	ca. 20 to

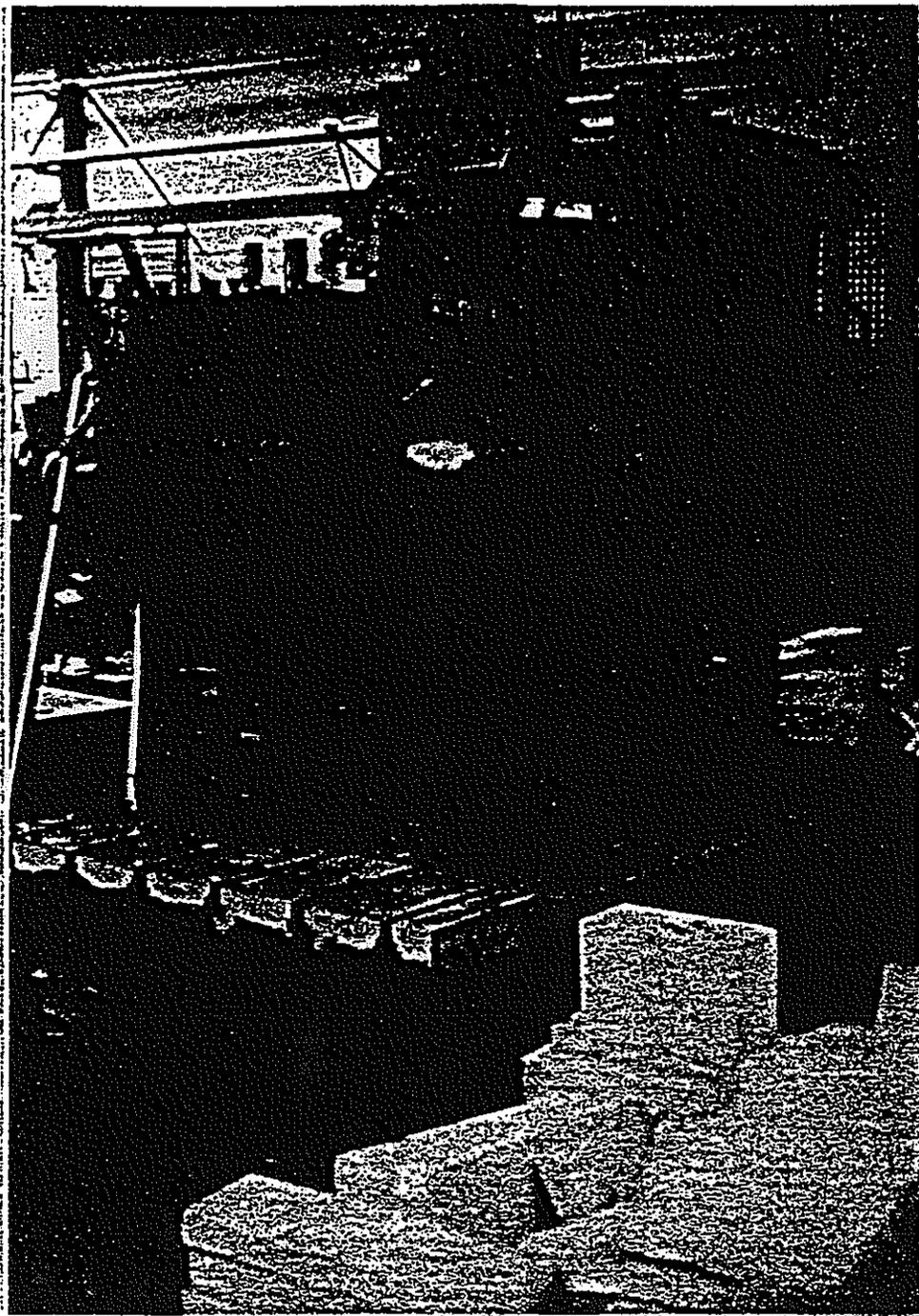
Comparison of PAMELA melter types

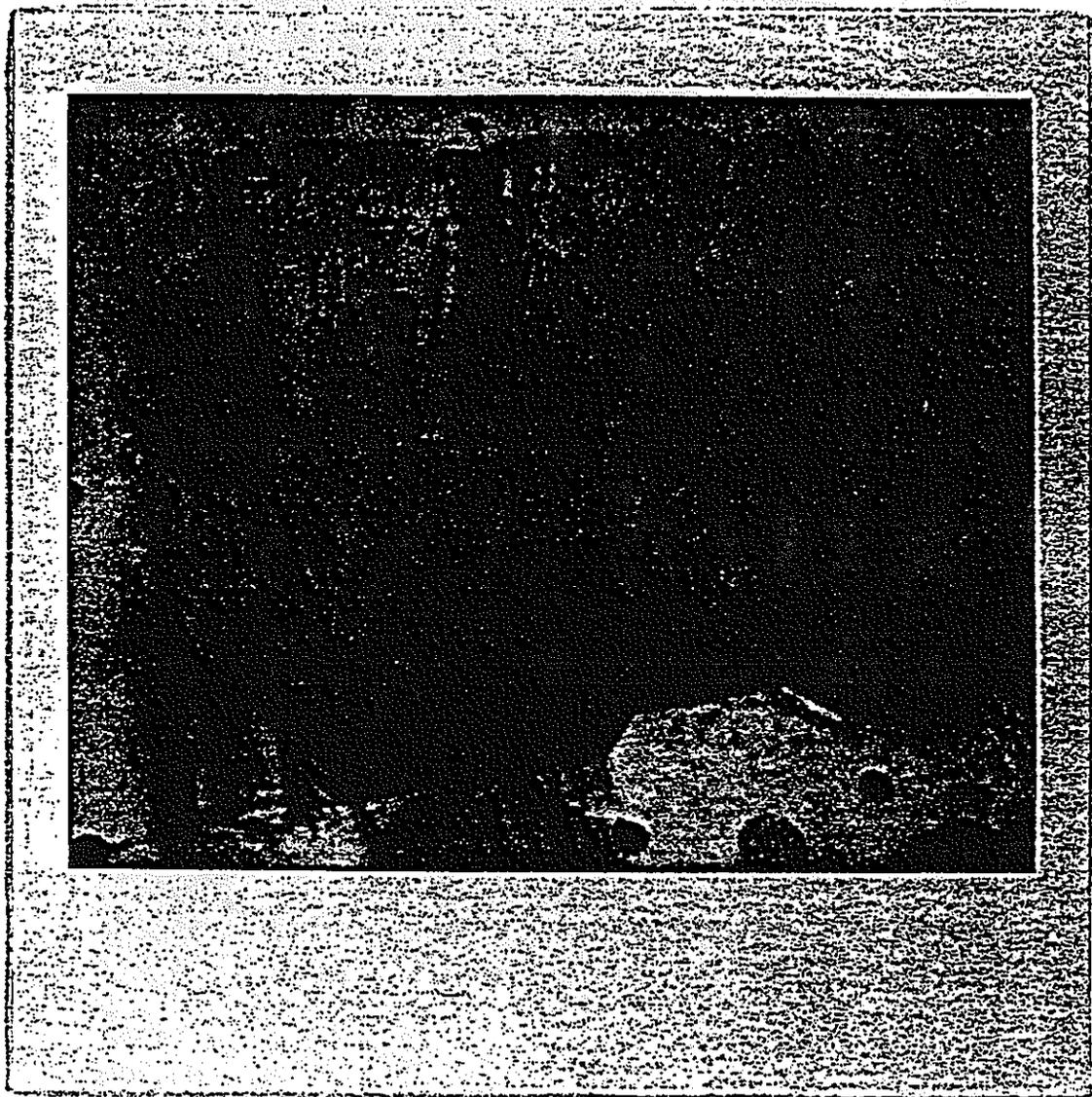


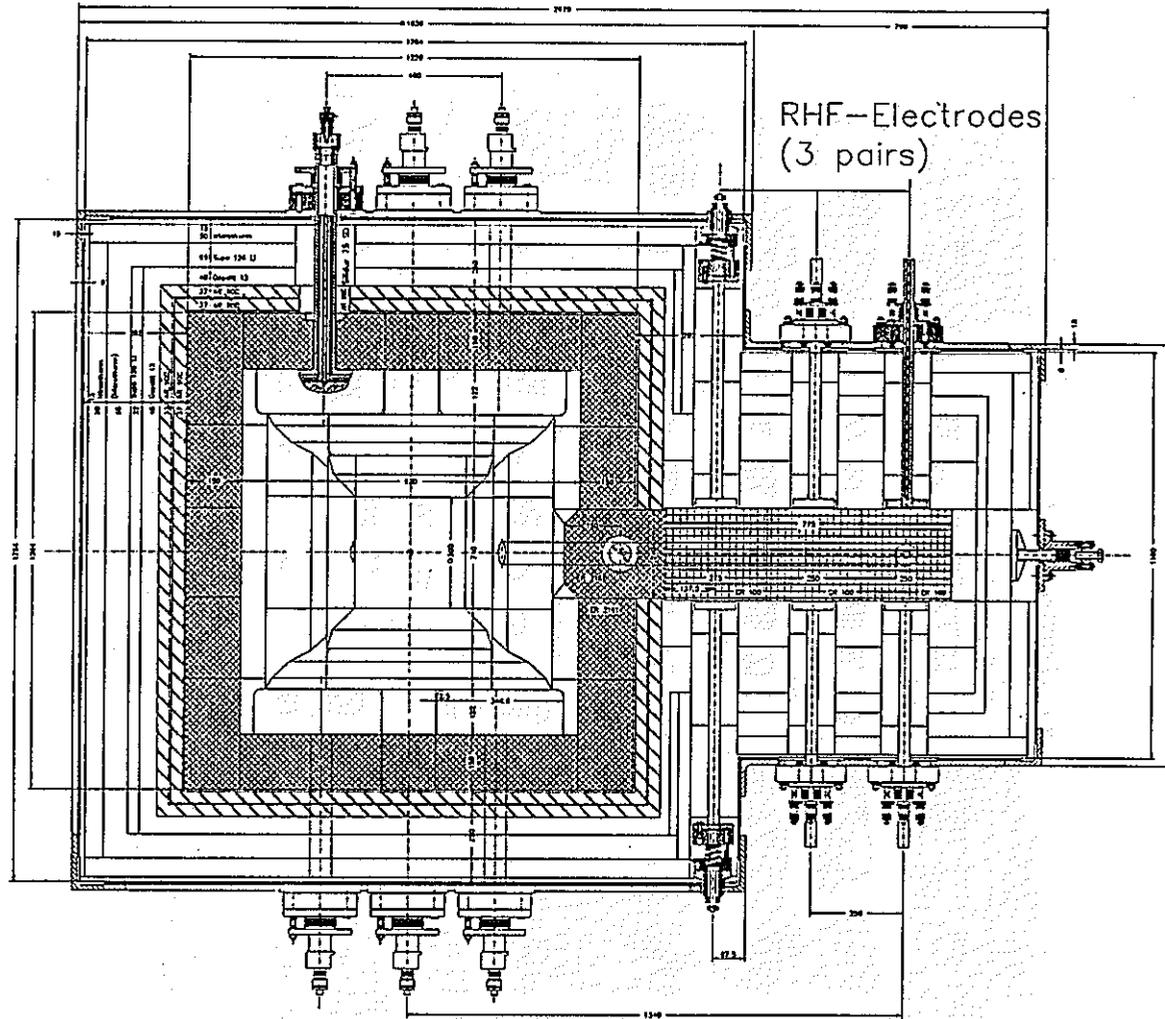
Longitudinal cross section of the K-6' melter



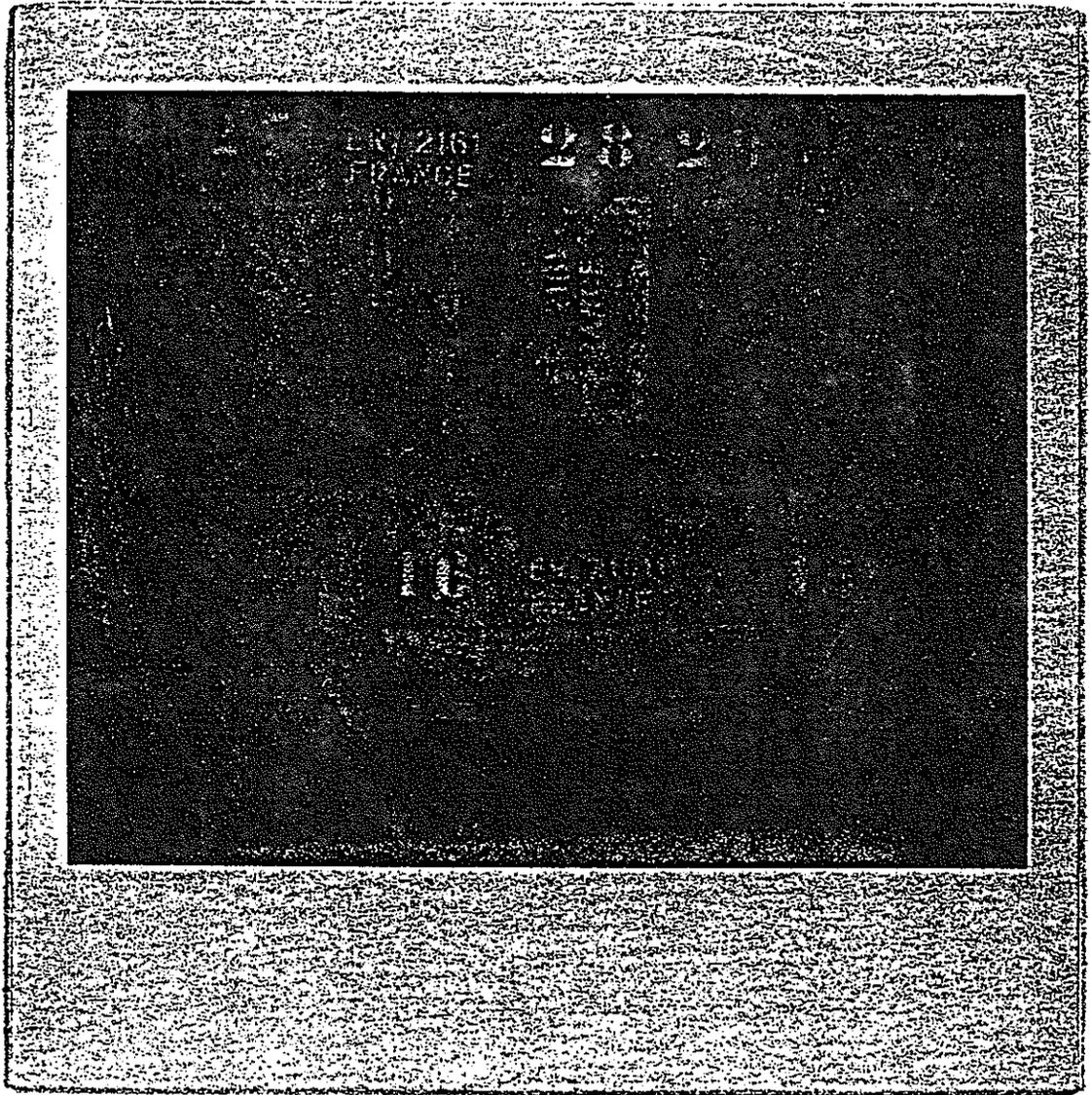
Lateral cross section of the melter K-6'







Horizontal cross section of the melter K-6'



2. P N C 発表分

2.1 Overview on R&D program for Vitrification of High
Level Liquid Waste

Overview on R&D Program for
Vitrification of High
Level Liquid Waste

The 9th Annual KfK-PNC Meeting on
High-Level Waste Management

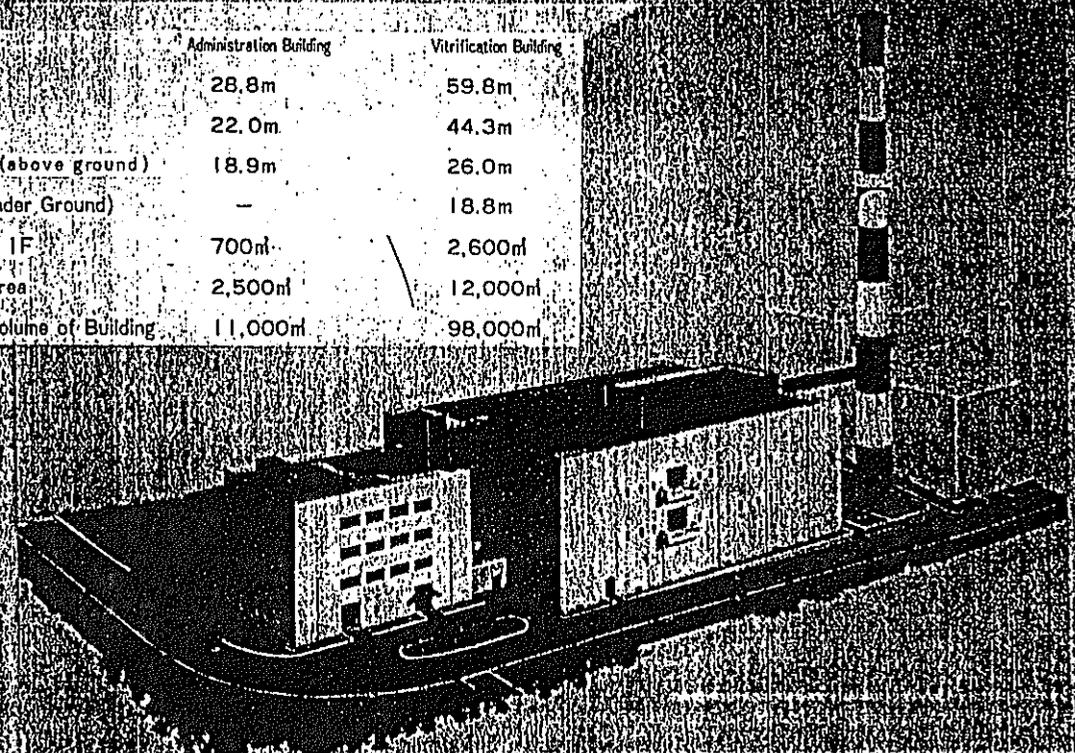
October 9-11, 1989

Power Reactor and Nuclear Fuel Development Corporation

Schedule of TVF

ITEM	1986																		1987					1988					1989					1990					1991					1992					1993				
	F.Y.	2	4	6	8	10	12	2	4	6	8	10	12	2	4	6	8	10	12	2	4	6	8	10	12	2	4	6	8	10	12	2	4	6	8	10	12	2	4	6	8	10	12	2	4	6	8	10	12	2			
Safety License Application and Permission							▼																																														
Building Construction																																																					
Equipment Manufacturing and Installation																																																					
Cold Test Operation																																																					
Hot Operation																																																					

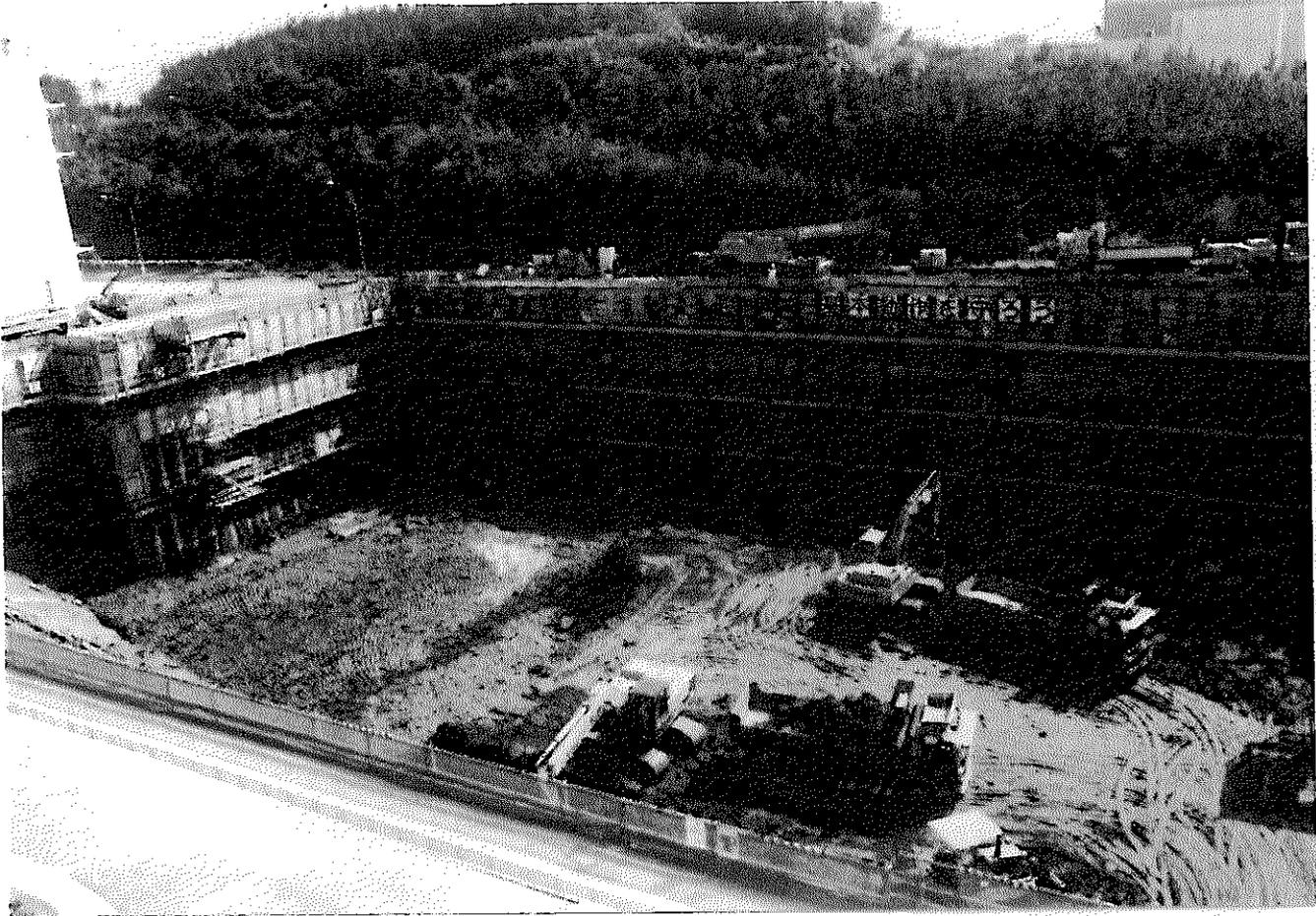
	Administration Building	Vitrification Building
Length	28.8m	59.8m
Width	22.0m	44.3m
Height (above ground)	18.9m	26.0m
(Under Ground)	-	18.8m
Area of 1F	700m ²	2,600m ²
Total Area	2,500m ²	12,000m ²
Total Volume of Building	11,000m ³	98,000m ³



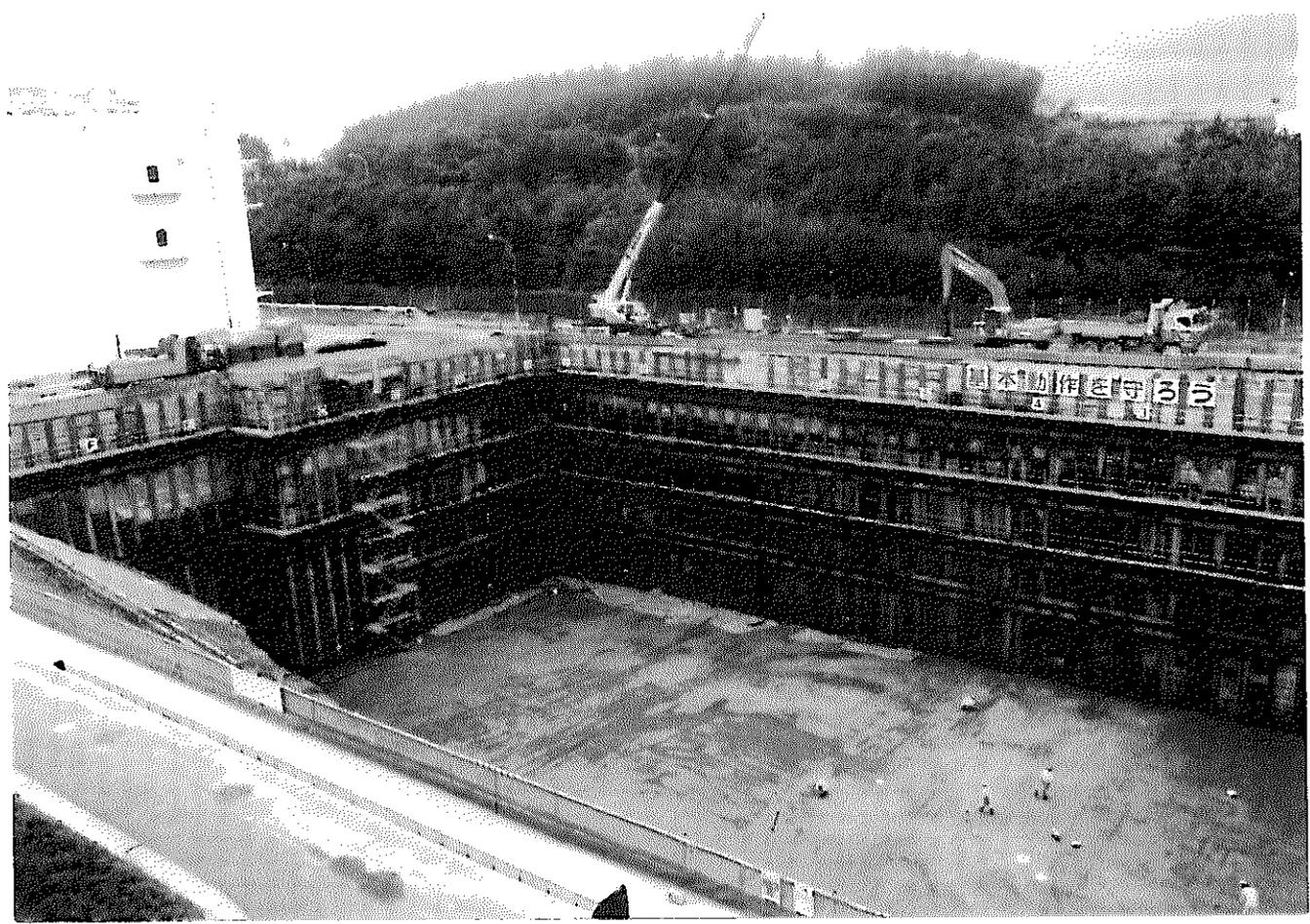
Bird's-eye View of TVF

Construction of Tokai Vitrification Facility (TVF)

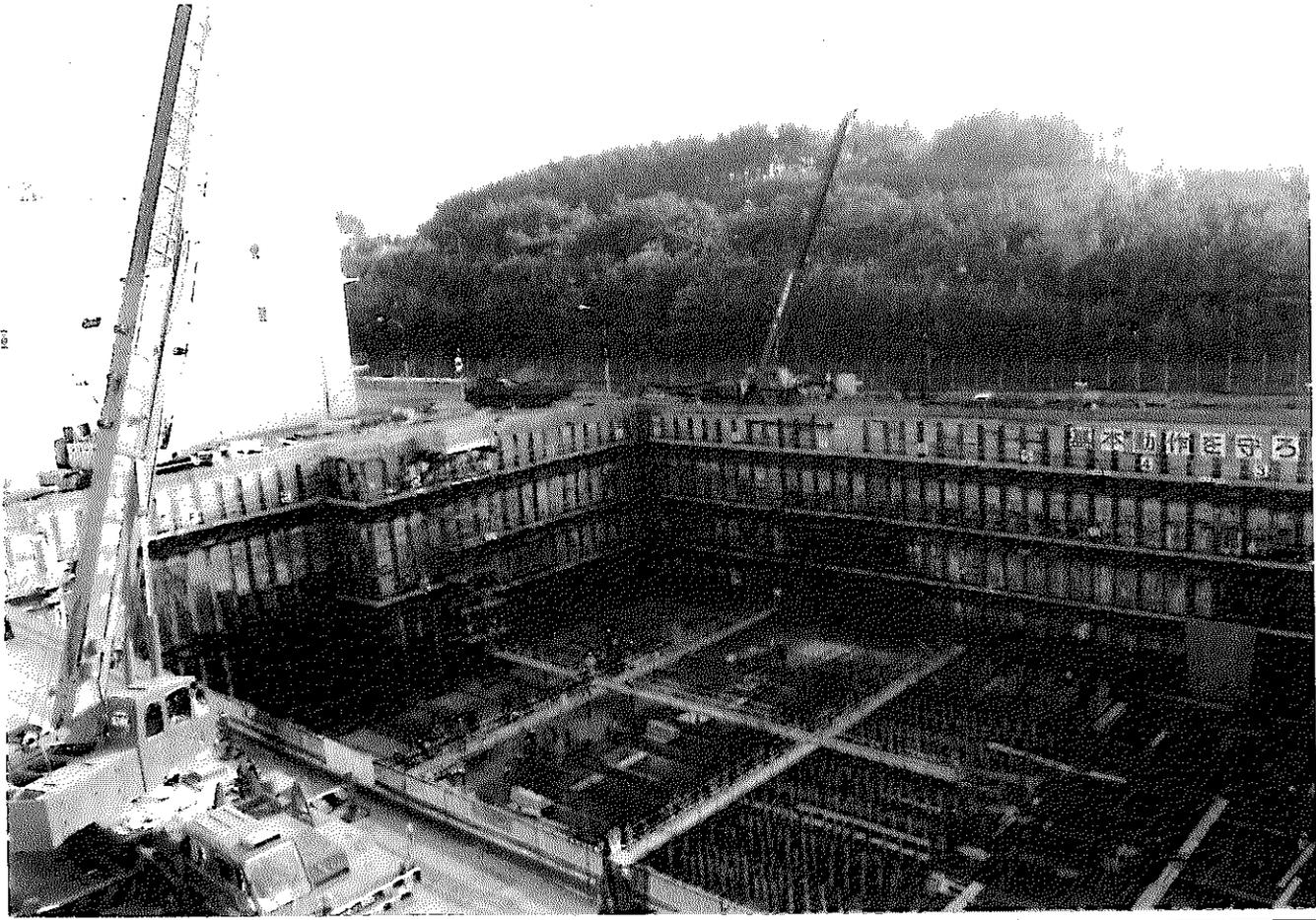
Licensing review for safety	Application	18 Mar. 1987	
	License	9 Feb. 1988	
Permission for design and method of manufacturing		<u>Building</u>	<u>Equipments</u>
	Application	19 May. 1988	10 Nov. 1988
	Permission	16 Jun. 1988	11 Jan. 1989
Construction	Start	June, 1988	
	Main Building	: 48% complete in Sep. 1989 up to ground level	
	Equipment	: ~2% complete melter refractory was cast pit structure for product storage	



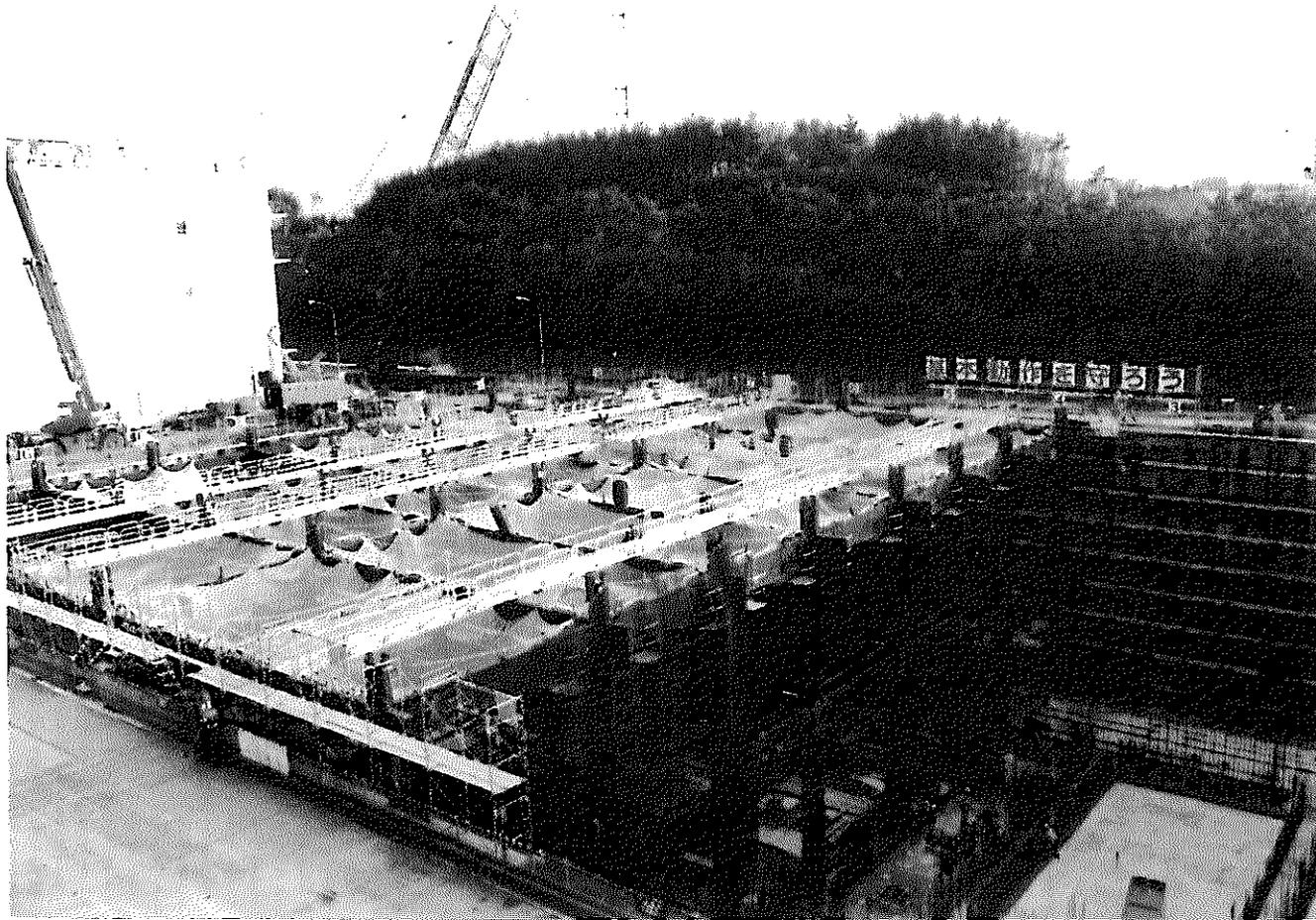
August 1988



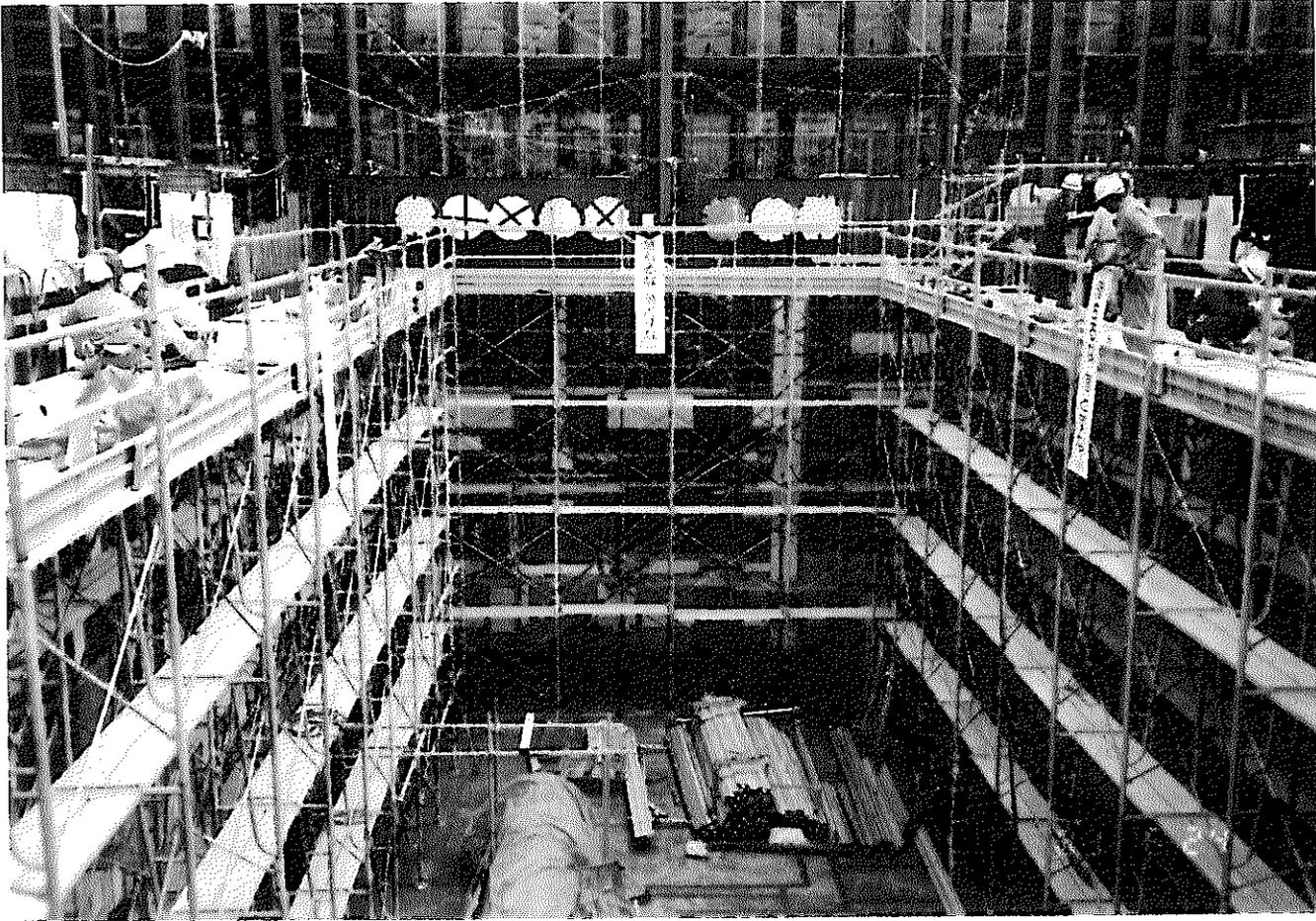
September 1988



December 1988



February 1989



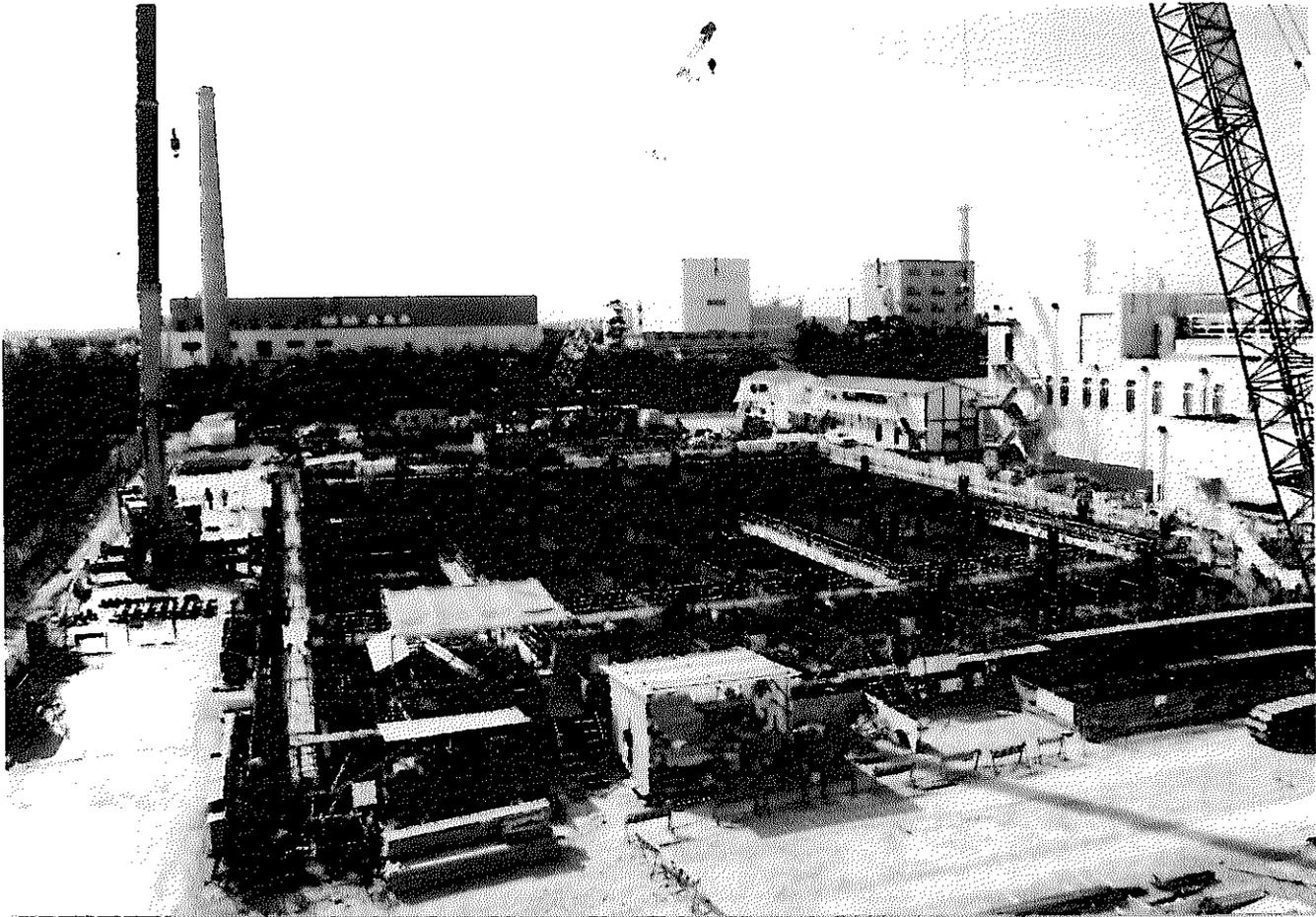
May 1989



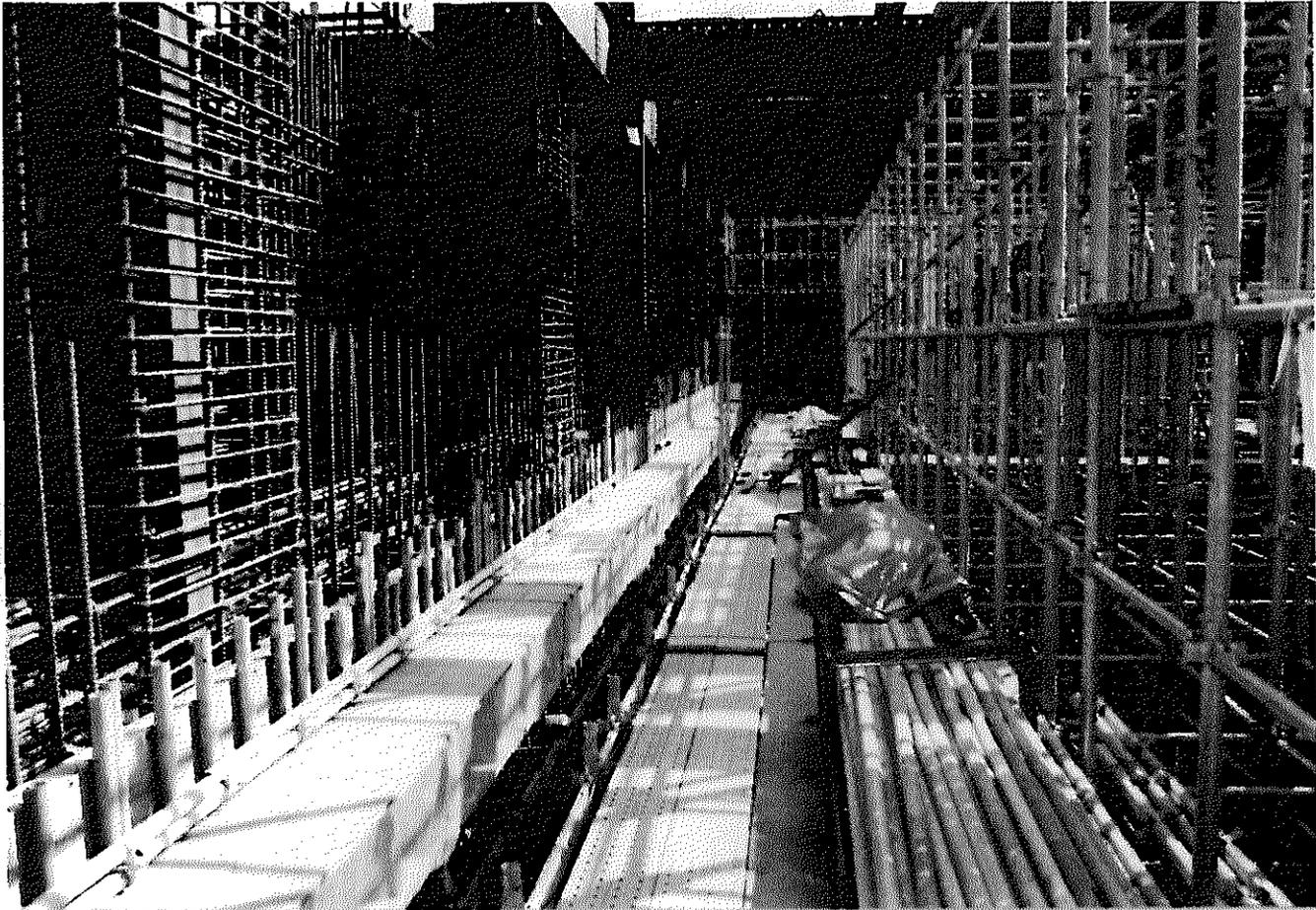
May 1989



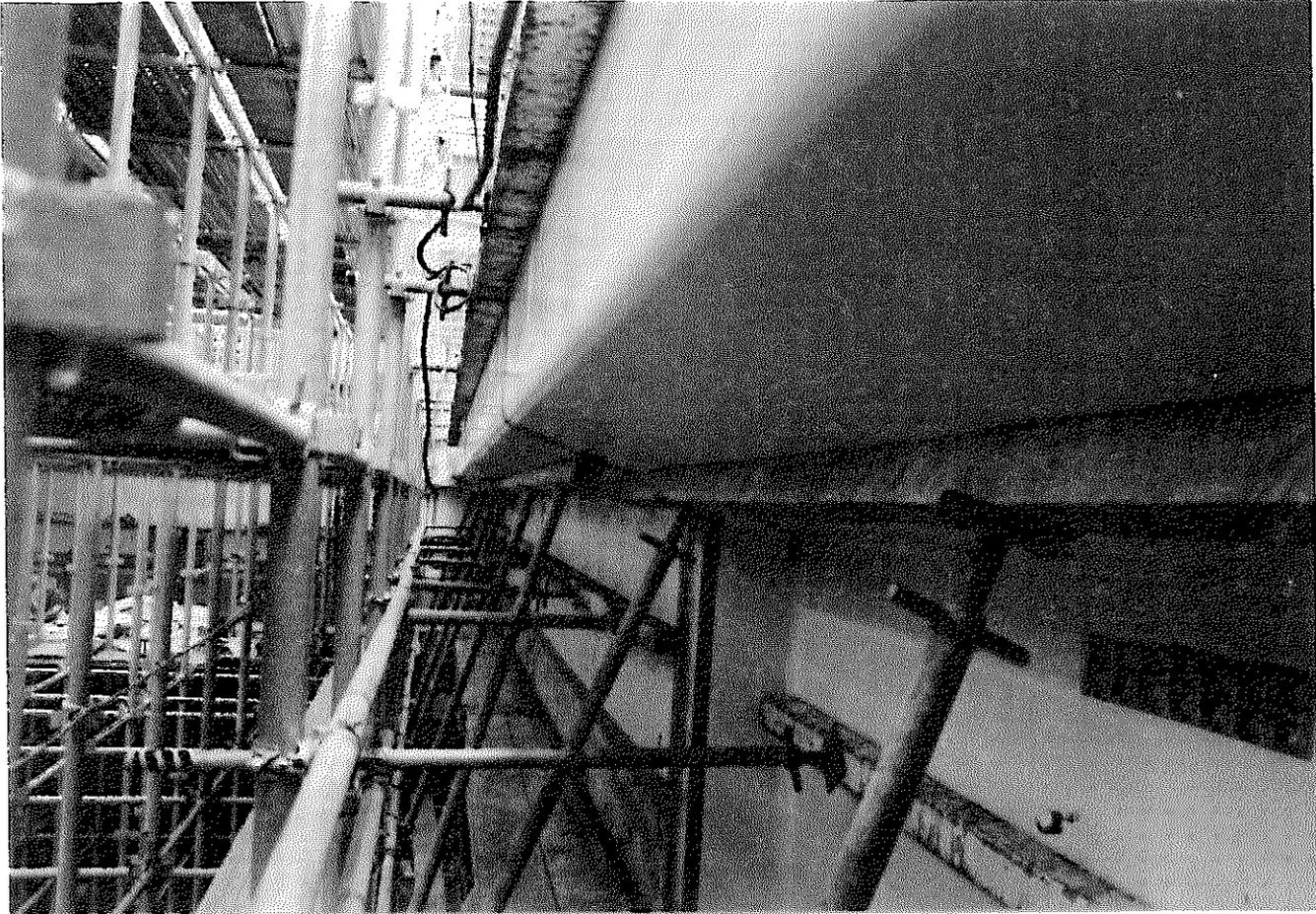
April 1989



July 1989



July 1989



August 1989

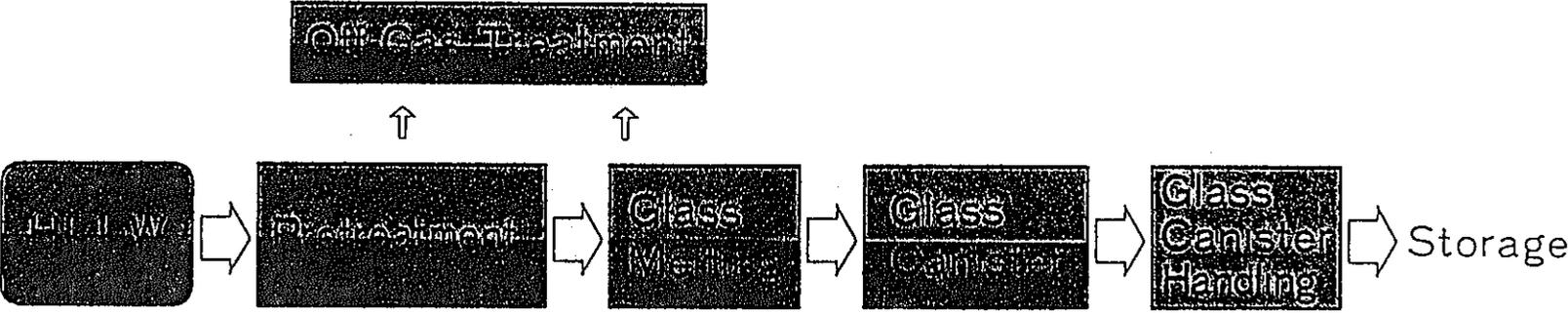
Research and Development Program for High Level Waste Conditioning

PNC SN8600 90-011

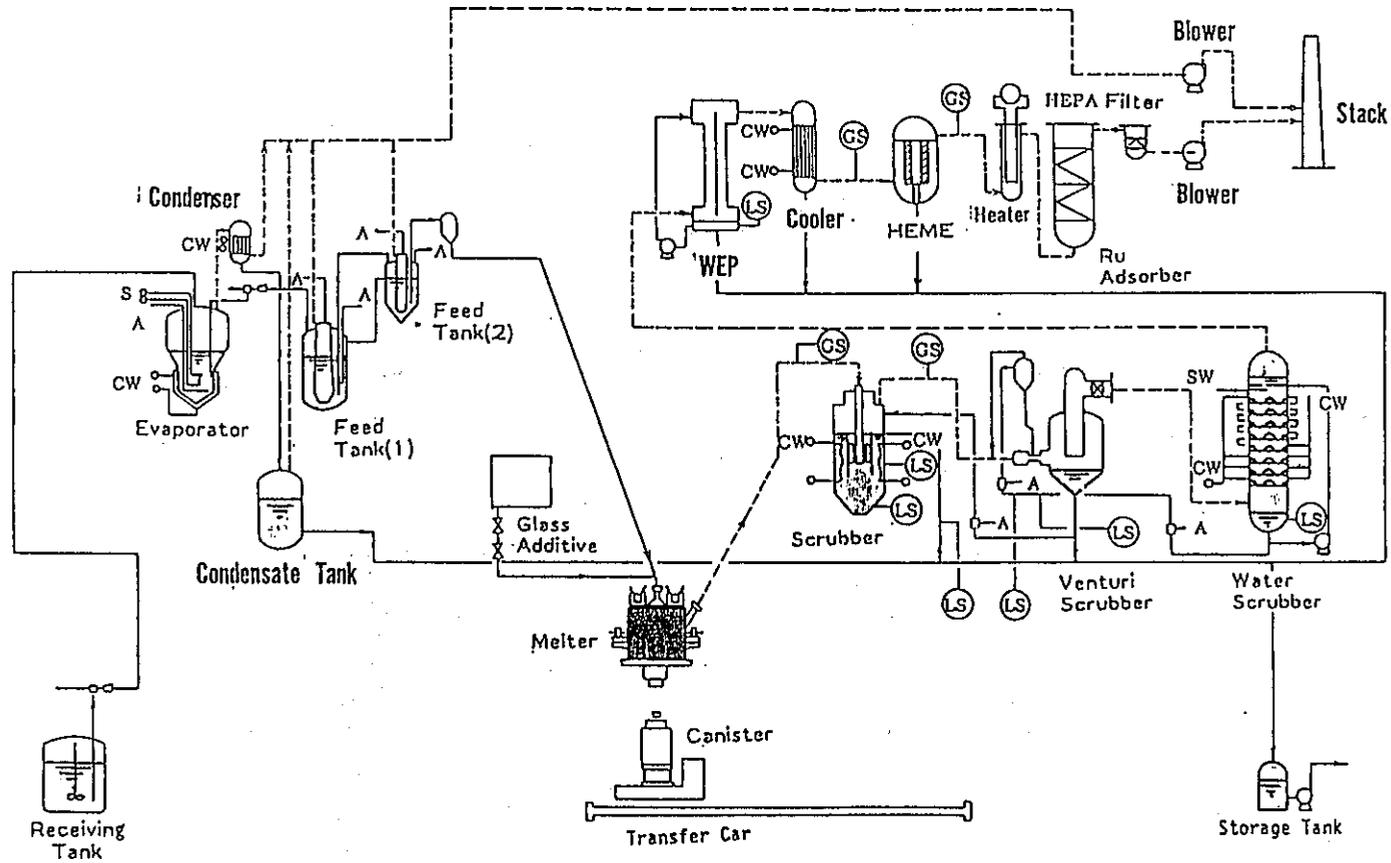
Fiscal Year	1988	1989	1990	1991	1992	1993
TVF	Construction			Cold test	Hot test and operation	
1. Mock up Test (MTF)	Noble metal, Training of operator			Development of 2nd melter		
2. Engineering Test (ETF)	Microwave, Noble metal (Small Melter), New material, Development of 2nd melter					
3. Remote Maintenance (EDF-III)	Model melter, Induction coil, Inspection of TVF component					Back-up for TVF
4. Dismantling	Mock-up I melter, Mock-up II melter, (Fully remote demonstration), Reduce of dismantling wastes					
5. Glass Characterization	Noble Metal, Support for MTF, ETF, Back-up for TVF					
6. Off-gas Behaviour	Iodine, Ruthenium, Technetium			Back-up for TVF		
7. Large-Scale Melter	Study	Design & manufacture		test (for Commercial plant)		
8. Cooperation for Commercial Plant	Study on Products, Support for design and licensing					
9. New Immobilization Technology	Reduce Waste Volume and heat generation, Partitioning of Ru, TRU					
10. Waste Technology for New Type of Fuel Cycle	Metal-, Carbide-, Nitride-, Fuel cycle					
	Metal (Salt, Cd),		Carbide (Organic acid),		Nitride (C-14)	

Vitrification Technology Development

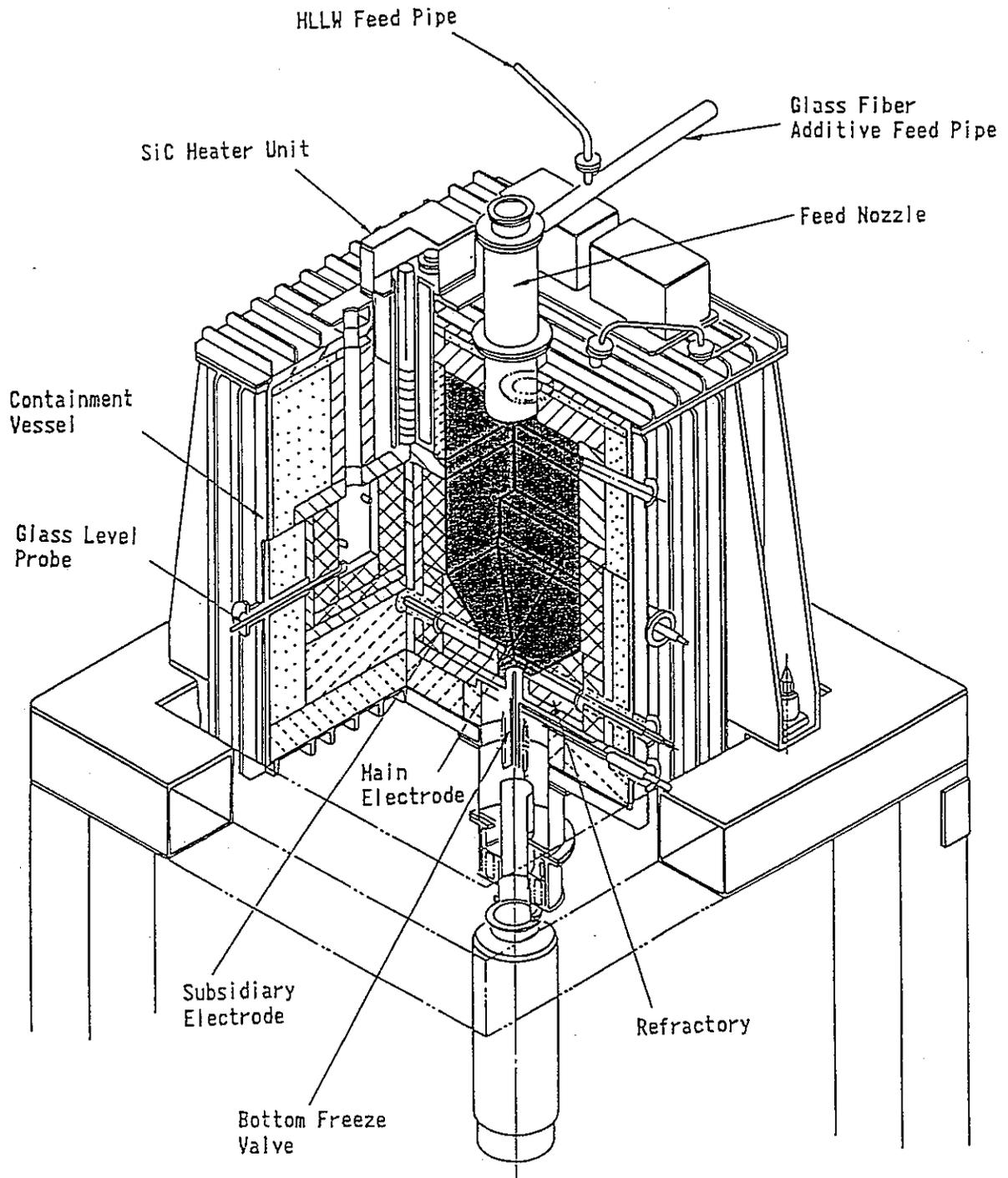
Process Development



- Cold Test (ETF, MTF, EDF-III)
- Hot Labo. Test (CPF)



Vitrification Process Flow



Structure of Melter

Feed Preparation

- Constant feed system → Quality assurance
(Airlift and Control)

Glass Melting Process

- Joule-heated Ceramic Melter → Melter operation with noble metal
→ Melter viewing and inspection
- Glass additive & Feed system → Quality assurance
- Glass drain system → Remote maintenance
- Microwave heating device → Combine with LFCM
- Process materials development → Refractory and electrode materials
- Instrumentations → Molten glass surface detection
→ Glass leak detection

Melter Development Schedule

F. Y. 82 83 84 85 86 87 88 89 90 91 92 93 94

(A-Melter)

Life Test & Thermal Cycle Test
 Resistance Heated Drain Nozzle

(B-Melter)

Microwave Heating-up
 Dismantling Test

(Advanced B-Melter)

Advanced Melter Structure (45° sloped bottom)
 Microwave Heating-up & Boosting
 Induction Heated Drain Nozzle

(C-Melter)

Induction Heated Drain Nozzle
 Bottom Flow Structure

(Mock-up)

Remote Design — Improvement — Dismantling Test
 (I) (II) (III)

Glass Powder & Bead Additive, Glass Fiber Additive
 Replaceable SiC Heater for Heating up
 Resistance Heated Drain Nozzle
 Monitoring Instruments (Glass Level Detection etc.)

Further Improvement for 2nd Operation Melter

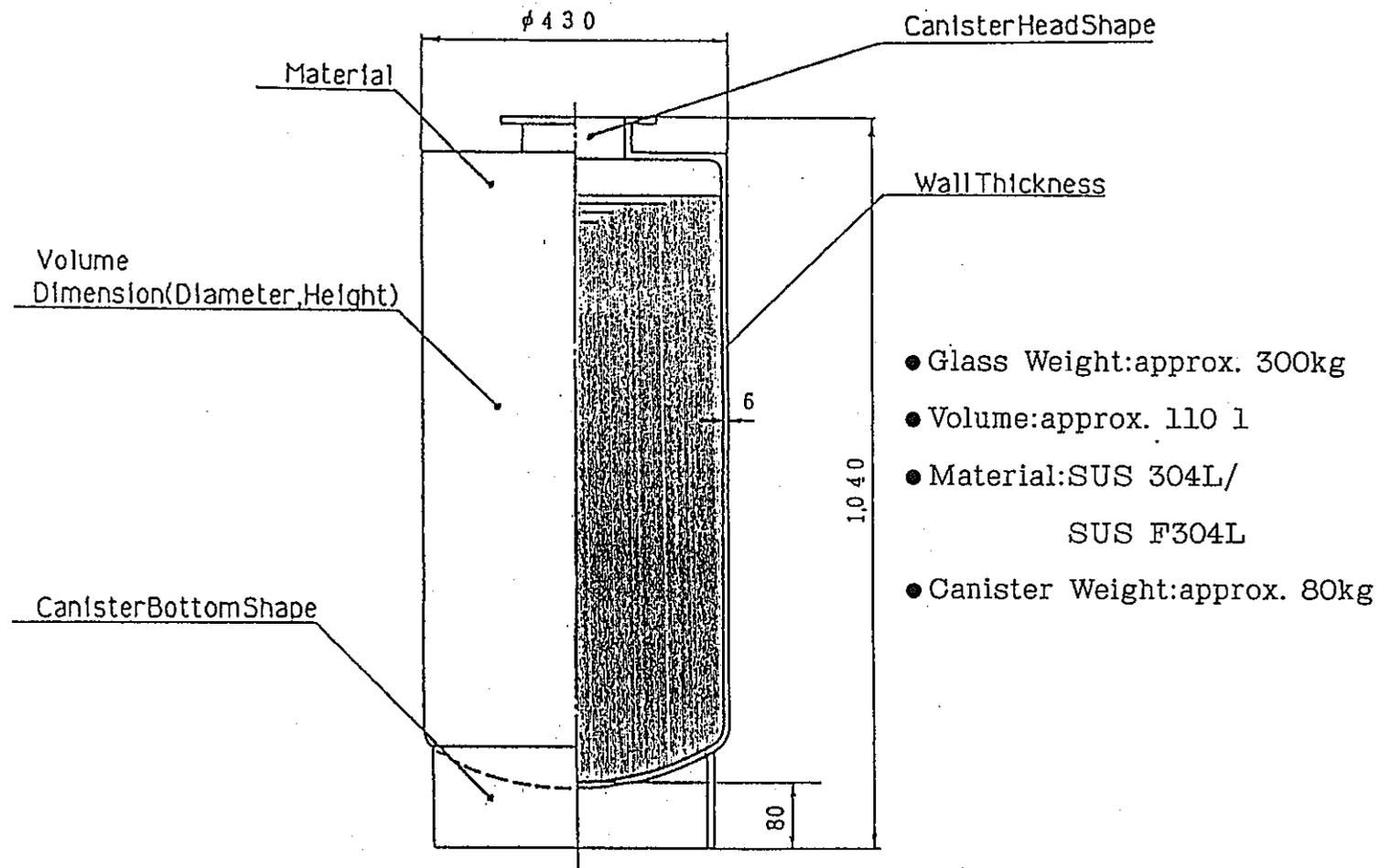
Cold Operation Hot
 Construction

Off Gas Treatment

- Ruthenium removal → Improvement of Ru
removal in wet process
- Investigation of volatile
& semivolatile elements

Canister Handling

- Canister design
- Lid welding technique → TIG welding
- Canister inspection technique → Non-destructive
inspection of welding



Design Factors and Determined Specification of Canister

Cooperative work for Commercial Vitrification Plant

- Conceptual design was completed. Jan. 1988
- PNC-LFCM process was selected. Apr. 1988
- Technical support to design a licensing application.
- Study on glass formulation and characteristics for commercial plant.
- Study on characteristics of vitrified product in canister.
- Full-scale test is planned.

2.2 Operational Experience in Mock-up Melter-III Campaign

Operational Experience in Mock-up Melter-III Campaign

The 9th Annual KfK-PNC Meeting on
High-Level Waste Management

October 9 - 11, 1989

Power Reactor and Nuclear Fuel Development Corporation

- Recent Operational Experiences in Cold Mock-up Test Facility
 - Experience of Melter Operation in the 19th Campaign with Feed of Noble Metals
 - Operational Attempt to prevent the Accumulation of Electroconductive Sludge in 20th Campaign

- Development of Glass Pouring Technique in its Initiation

Summary of Mock-up Melter-III Campaign

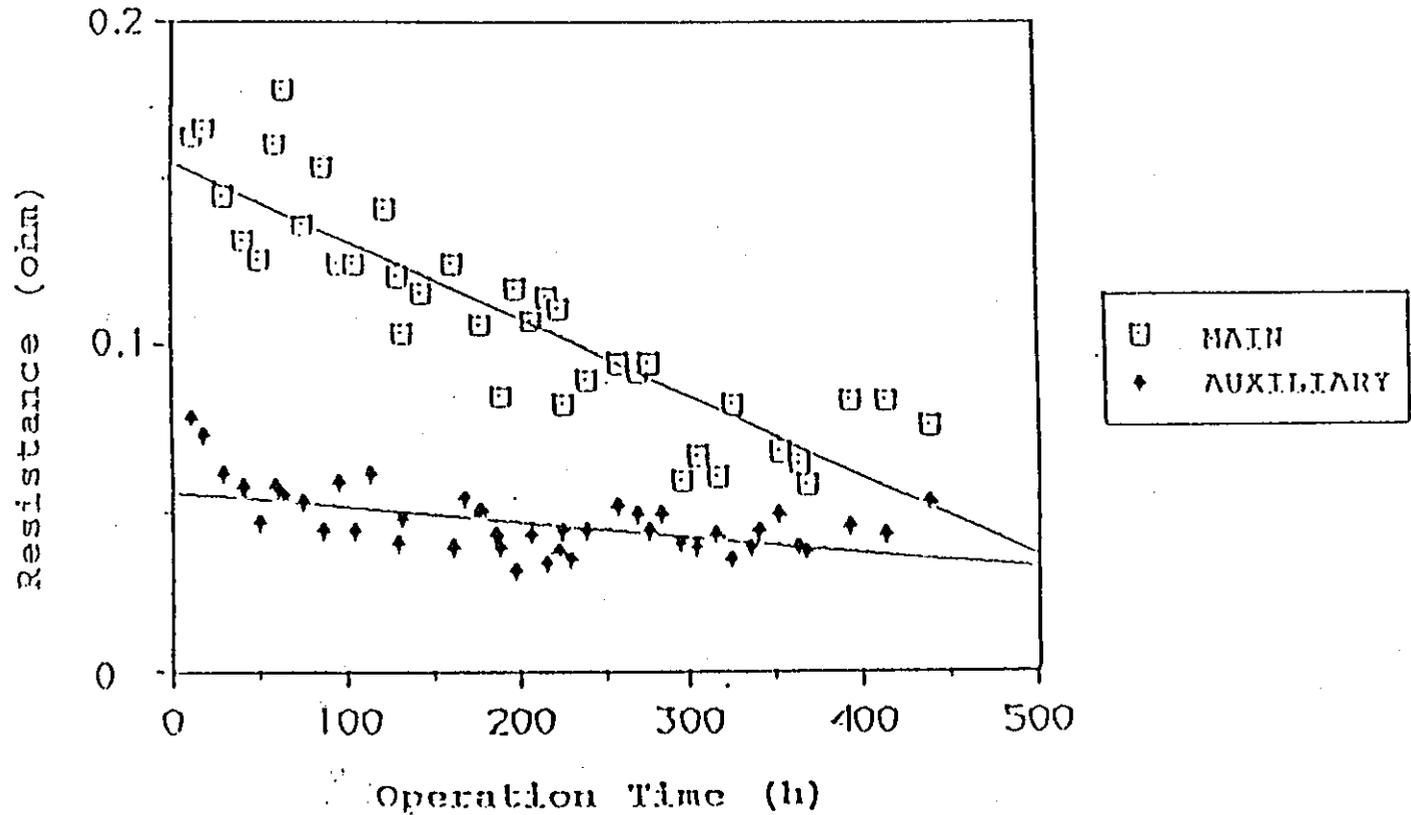
PNC SN8600 90-011

Campaign NO. Item	1 6	1 7	1 8	1 9	2 0
Campaign Object	Test run (start-up of this melter)	Confirmation of melt rate and glass draining	Confirmation of glass melt rate	Production of vitrified glass for high burn-up HLLW	Confirmation of melter system and noble metal behavior
Operation Period	1988.2.10-3.7 9batch(300kg/b)	1988.6.20-6.27 4batch(300kg/b)	1988.7.14-7.22 12batch(150kg/b)	1989.1.10-1.24 6batch(410kg/b)	1989.6.5-7.14 19batch(300kg/b)
Waste Loading	25.0 wt%	25.0 wt%	18.3 wt%	18.4 wt%	25.0 wt%
Simulated HLLW	4.5 m ³	2.1 m ³	2.1 m ³	3.7 m ³	10.0 m ³ (7.4 m ³ including noble metals)
RuO ₂	25.3 kg	12.8 kg	-	24.2 kg	40.8 kg
PdO	14.4 kg	7.7 kg	-	10.5 kg	22.9 kg
Rh ₂ O ₃	-	-	-	3.6 kg	0.7 kg
Glass Additive	2.1 ton	1.0 ton	1.5 ton	2.0 ton	4.4 ton
Glass fiber	2.1 ton	1.0 ton	0.5 ton	1.4 ton	4.4 ton
Glass beads	-	-	1.0 ton	0.6 ton	-
Glass Product	2.6 ton	1.1 ton	1.8 ton	2.5 ton	5.6 ton

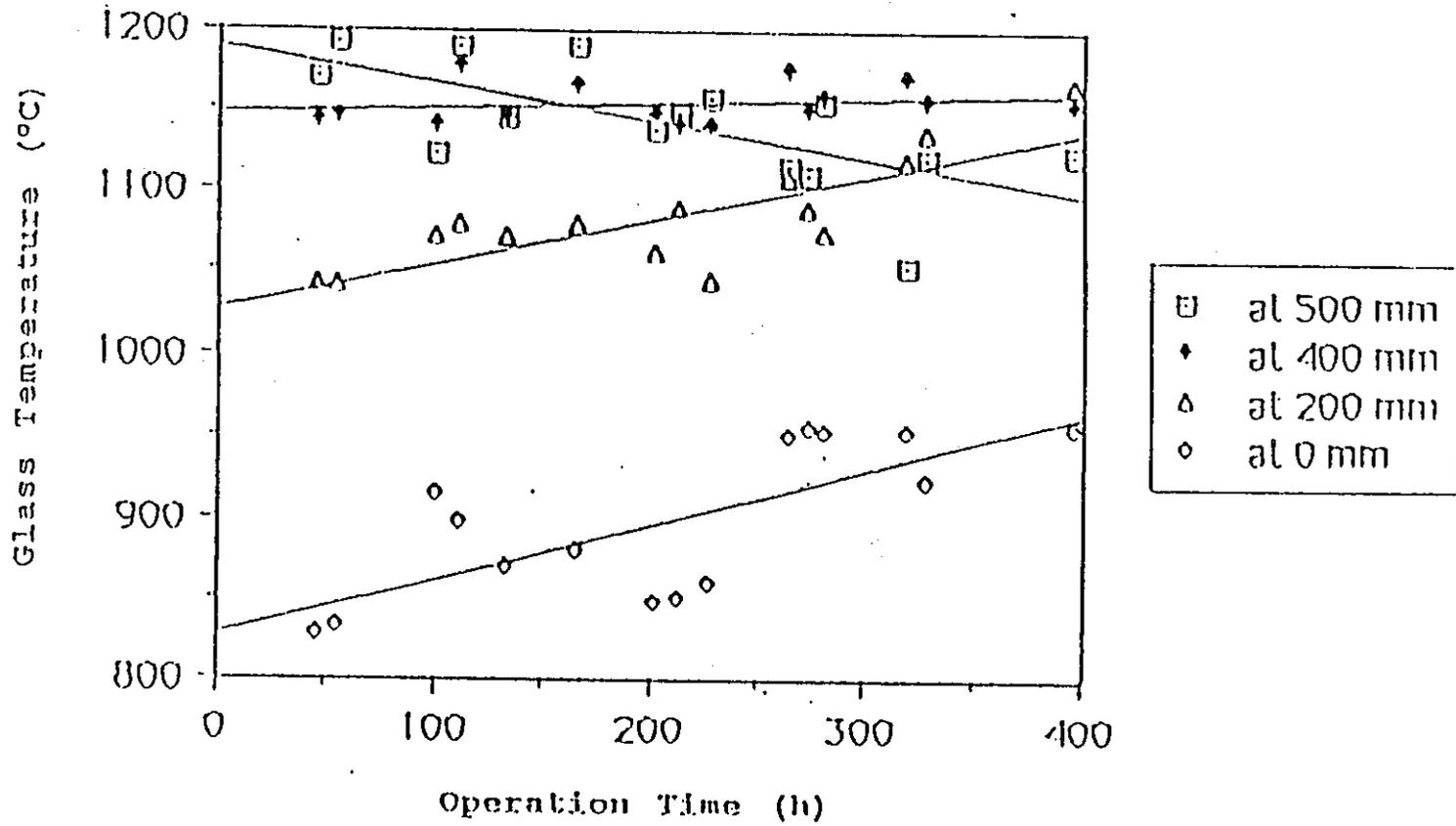
Experience of Melter Operation in the 19th Campaign
with Feed of Noble Metals (Ru, Rh, and Pd)

Noble Metals Fed to the Melter

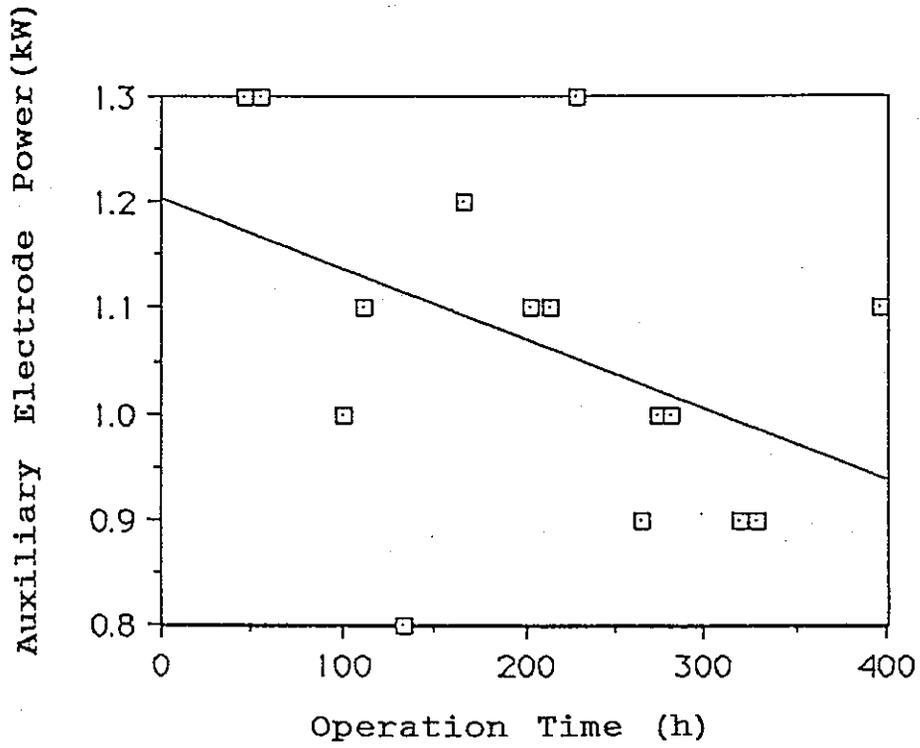
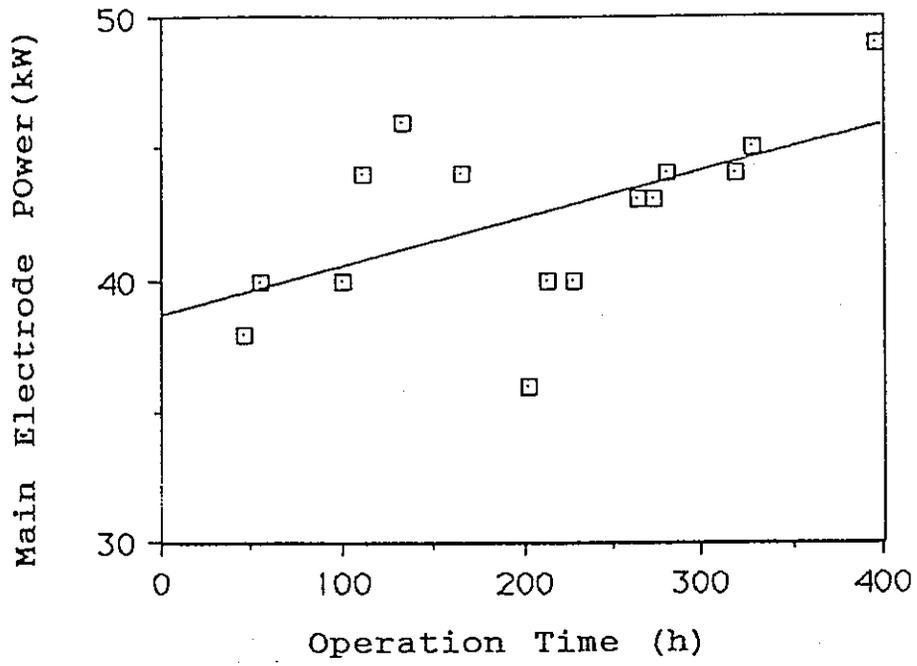
Element	Amount (kg)	Content in Feed (g/l)	Content in Glass (wt %)
RuO ₂	24.2	6.50	0.90
Rh ₂ O ₃	3.6	0.97	0.13
PdO	10.5	2.82	0.39



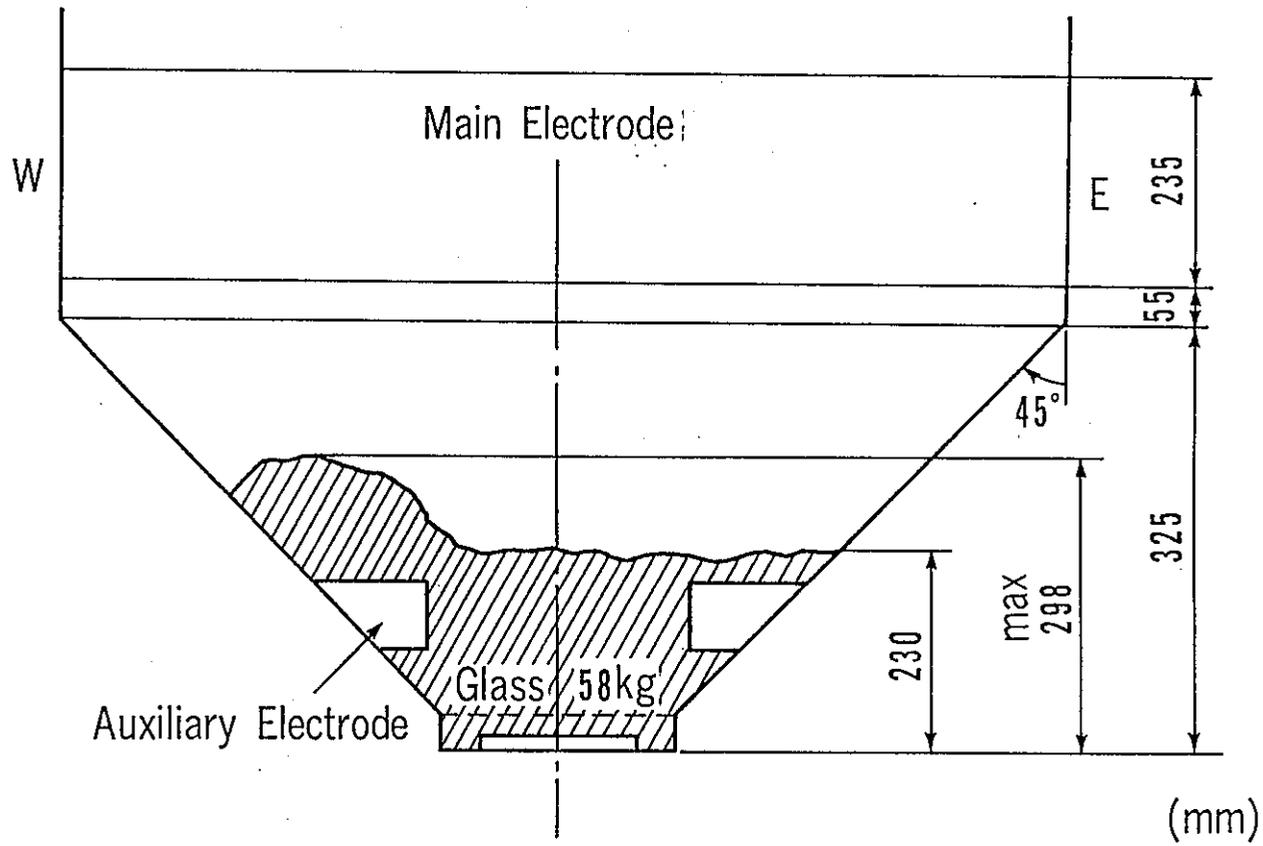
Change of Resistance between Electrodes during the Operation (19th Campaign)



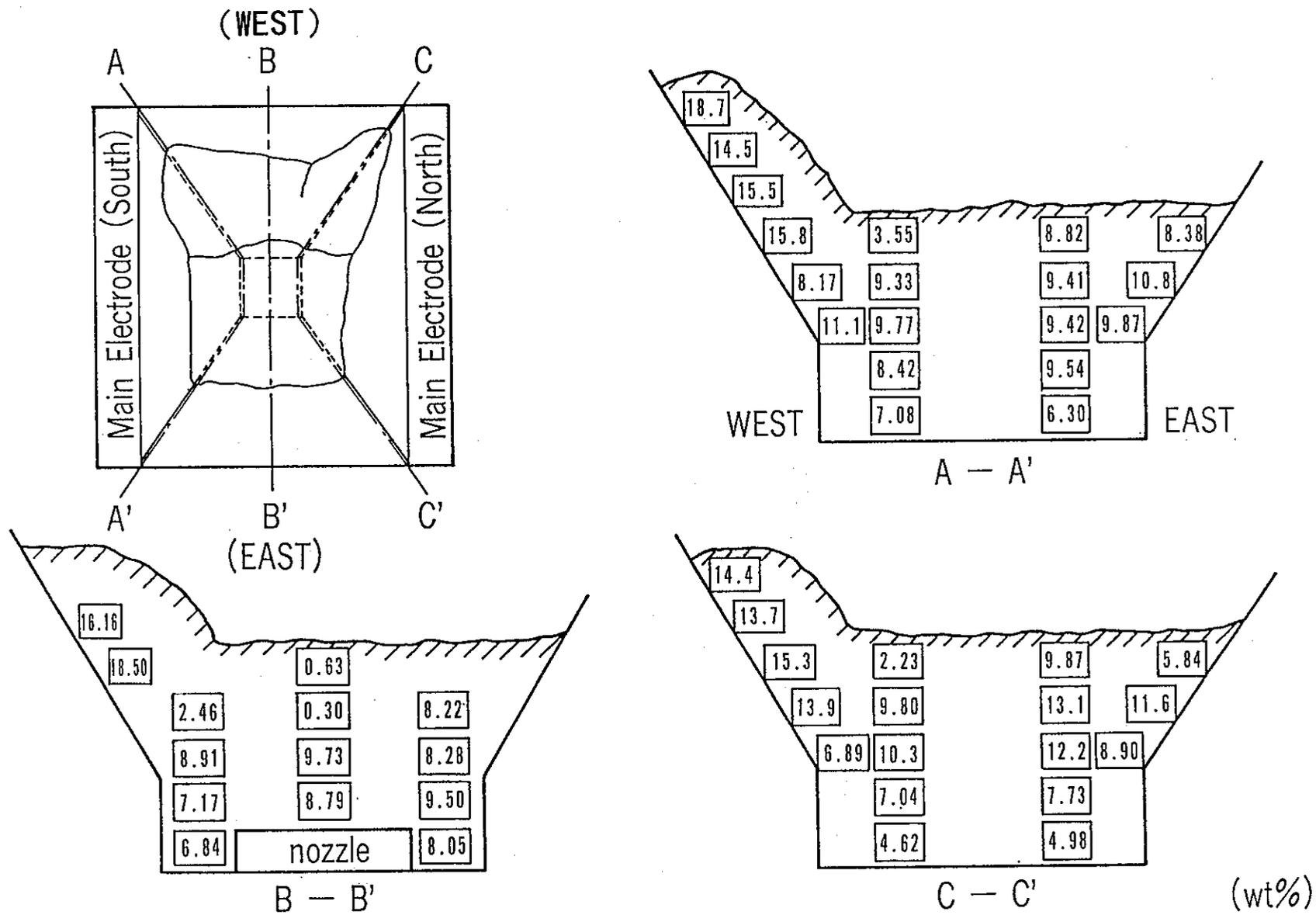
Helder Bottom Temperature during the Operation
(19th Campaign)



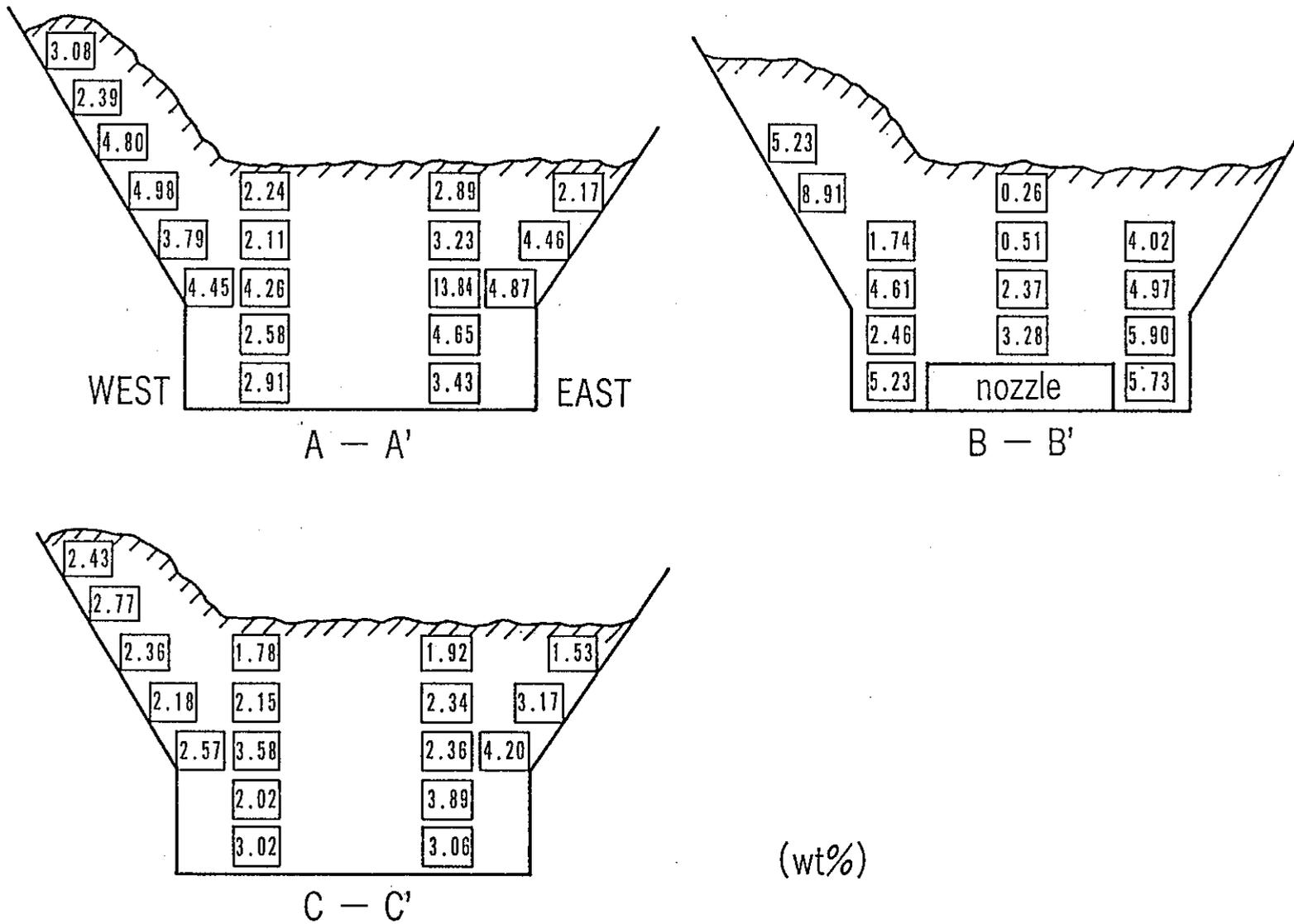
Change of Electrodes Power during the Melter Operation (19th Campaign)



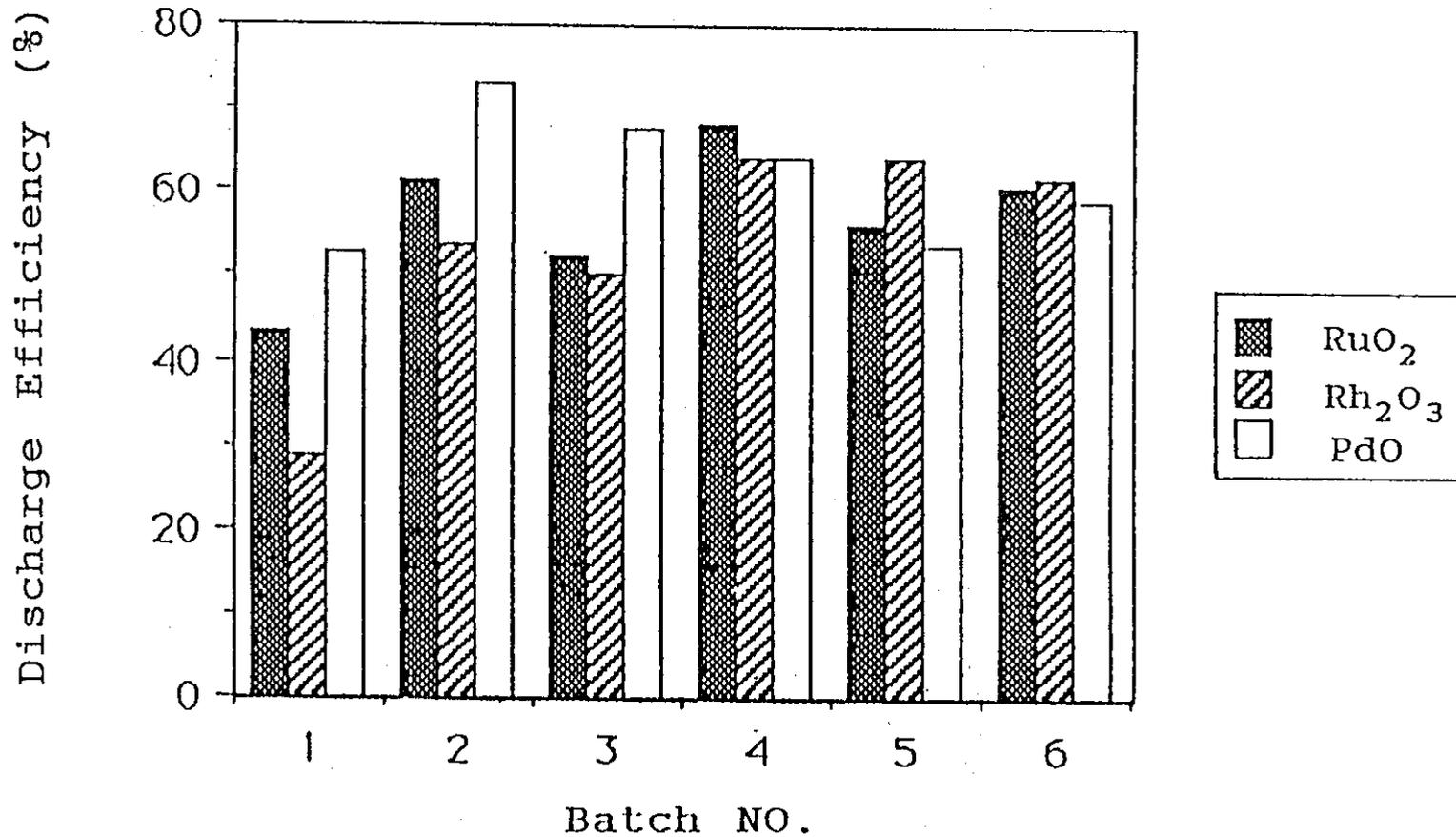
Accumulation of Noble Metals after Drain out



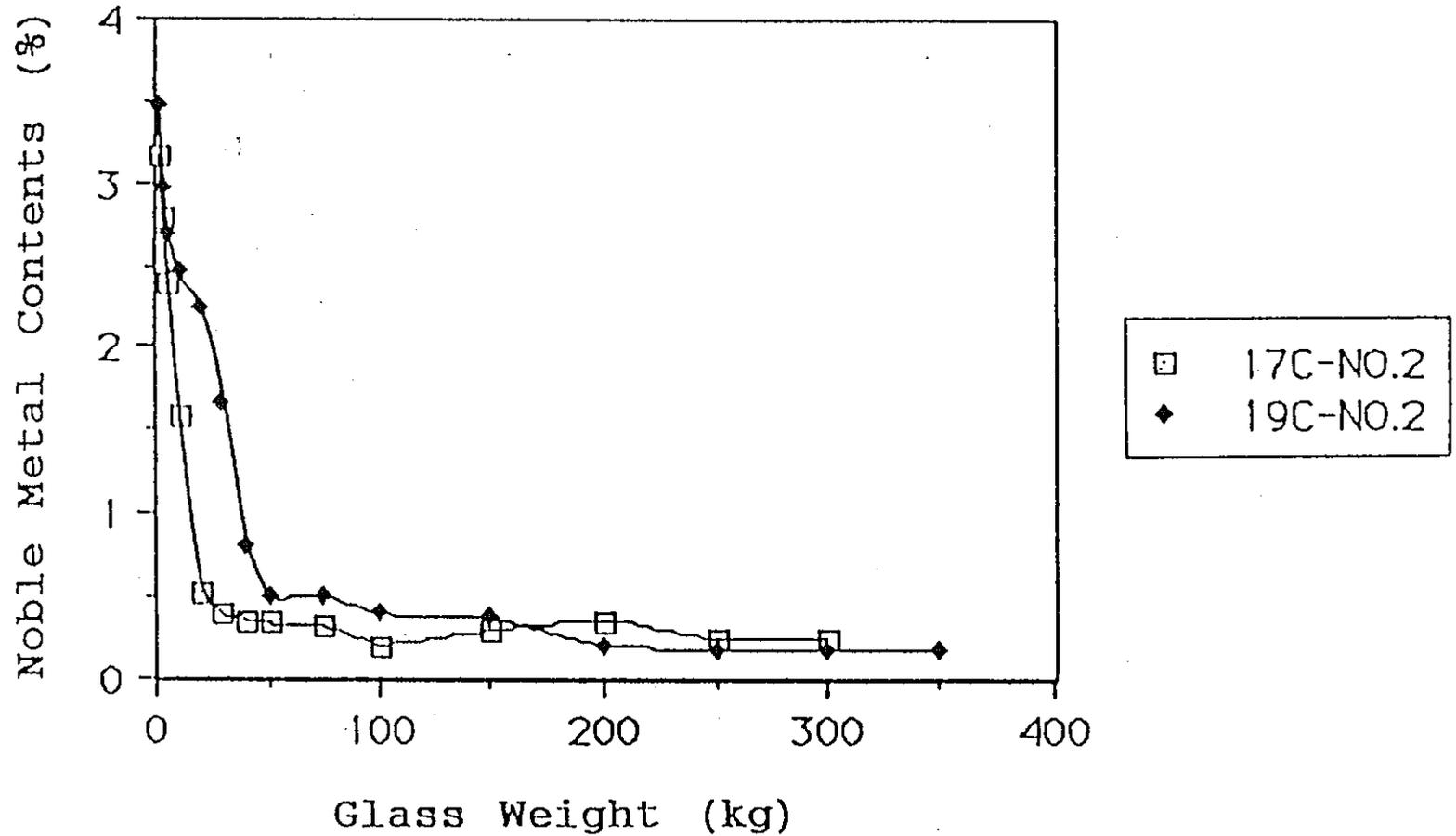
Content Distribution of RuO₂ in Melter Bottom



Content Distribution of PdO in Melter Bottom



Discharge Efficiency of Noble Metals during the Melter Runs (19th Campaign)



Change of RuO₂ Content during the Glass Pouring (17th, 19th Campaign)

Experience of Melter Operation in the 20th Campaign with Feed of Noble Metals (Ru, Pd)

— Operational Attempt to prevent the Accumulation
of Electroconductive Sludge —

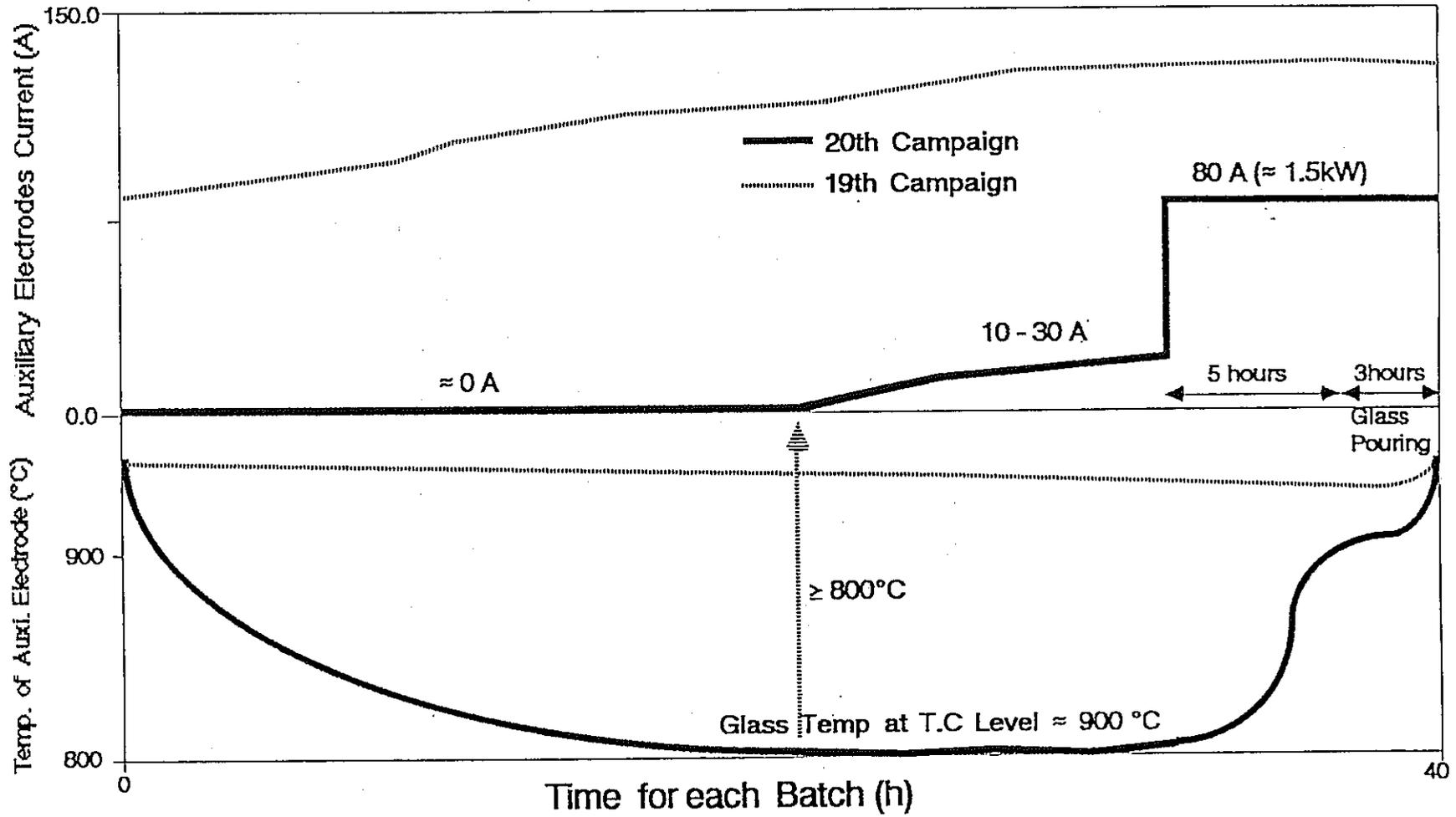
Noble Metals Fed to the Melter

Element	Amount (kg)	Content in Feed (g/l)	Content in Glass (wt %)
RuO ₂	40.8	6.13	0.89
PdO	22.9	3.45	0.50

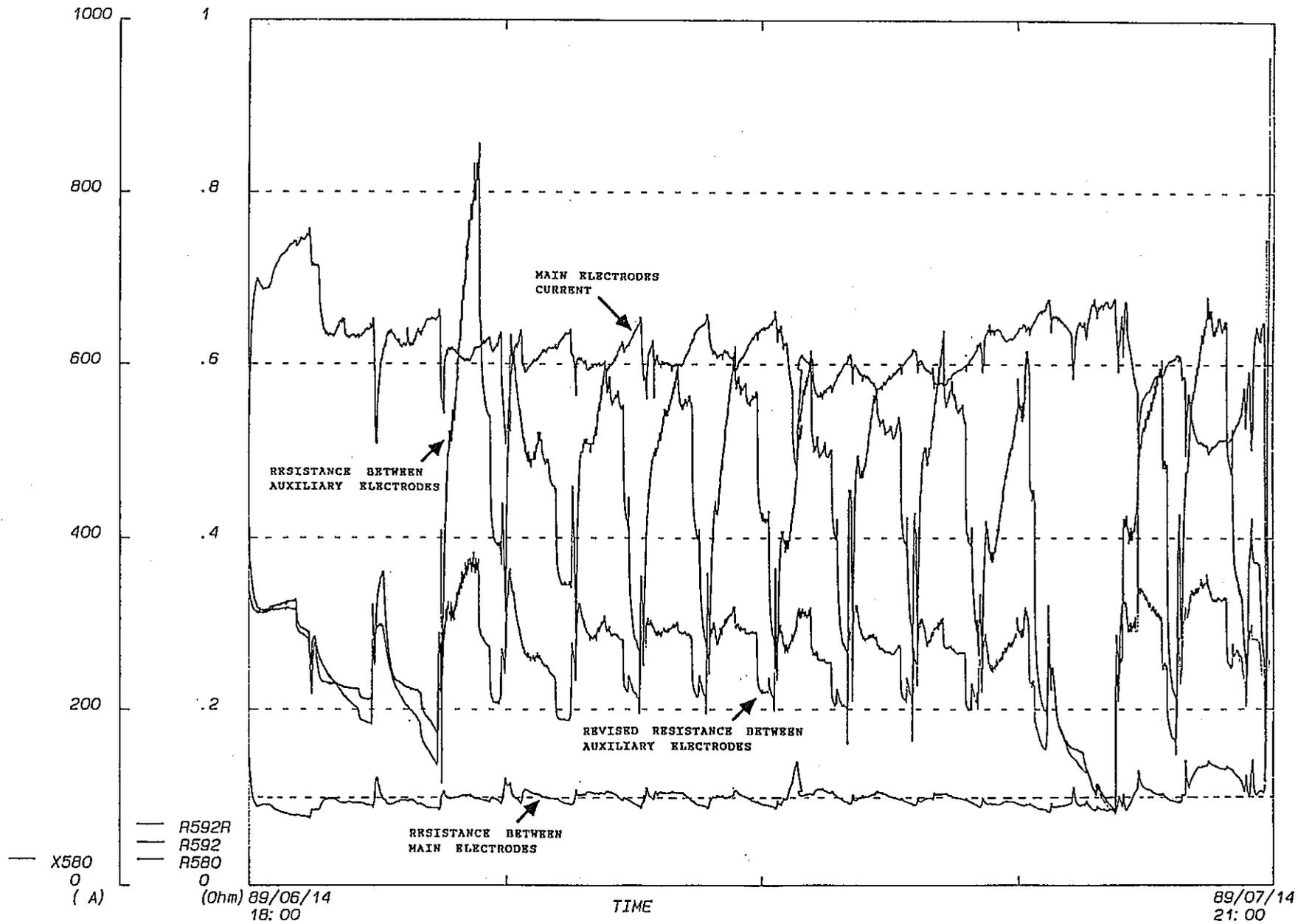
Operational Conditions and Results in the 20th Campaign

PNC SNR600 90-011

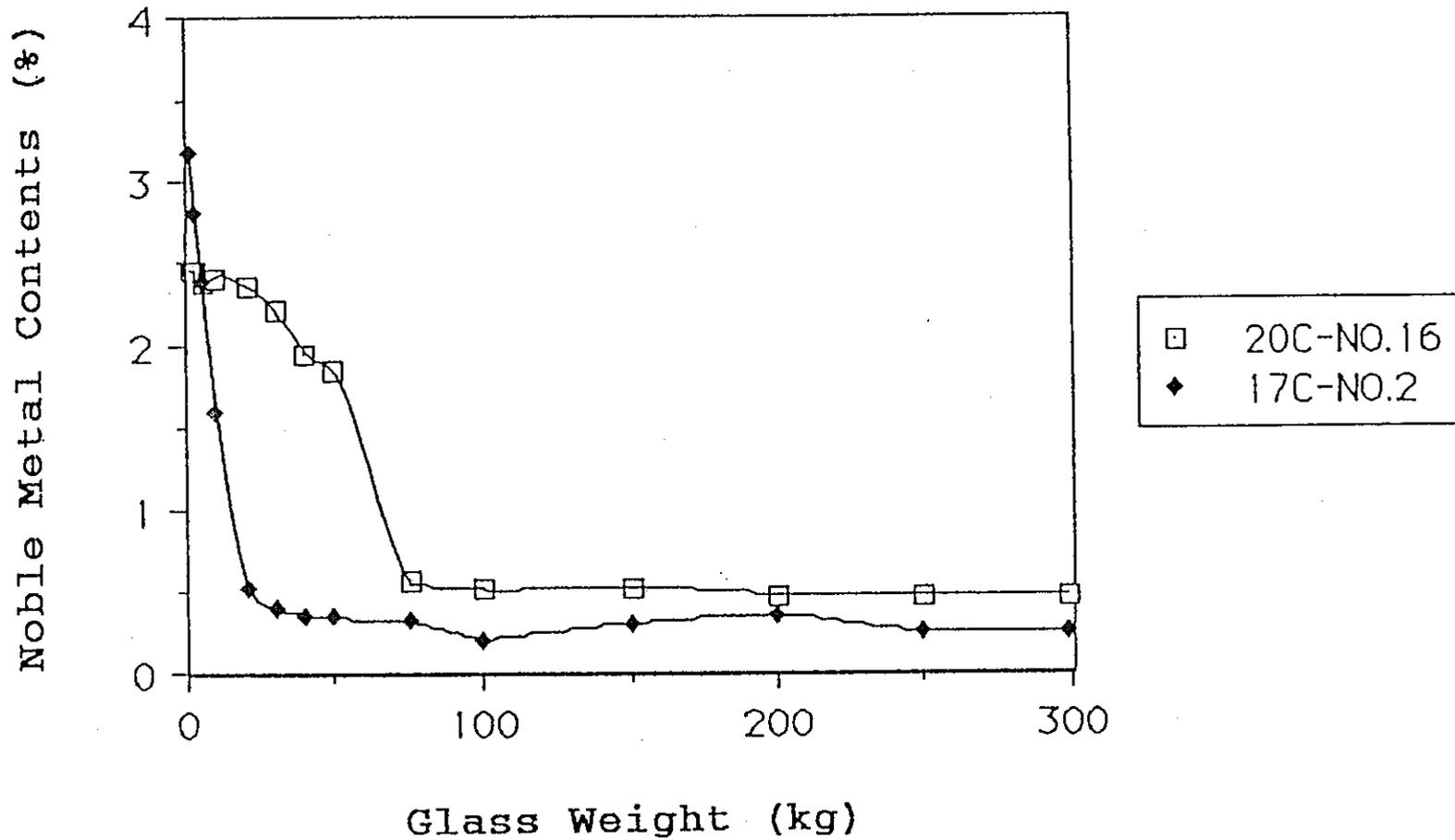
Batch No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
Operation Period	1989 6/5 ~7	7 ~9	9 ~10	10 ~12	14 ~16	16 ~18	18 ~20	20 ~22	22 ~24	24 ~26	26 ~28	28 ~30	30~7/2	2 ~4	4 ~6	6 ~8	8 ~10	10 ~12	12 ~14
Kind of Feed	SW-15				SW-17												SW-30		
Glass Production Rate (kg/h)	7.0				7.0				6.5						7.1				
Feed Rate (t/h)	15.5				12.0				10.9						9.2				
Amount of Feed (t)	2641				2148				4506						781				
Glass Production (kg)	1189				1259				2717						608				
Main Electrodes Power (kW)	41.0	41.8	40.8	41.7	42.0	37.4	35.8	35.3	35.3	35.9	35.8	36.0	34.1	33.0	33.0	36.0	35.9	33.4	33.0
Auxiliary Electrodes Power	80A				80A				0 A						940°C (max 120A)		0 A		
Power between Main Electrode and Nozzle	<ul style="list-style-type: none"> •Control the temperature of auxiliary electrode over 800°C •Turn on a current of 80A at 5hours before pouring •Turn on a current of 30A at 10 hour before pouring •Turn off a current at the termination of pouring 																		



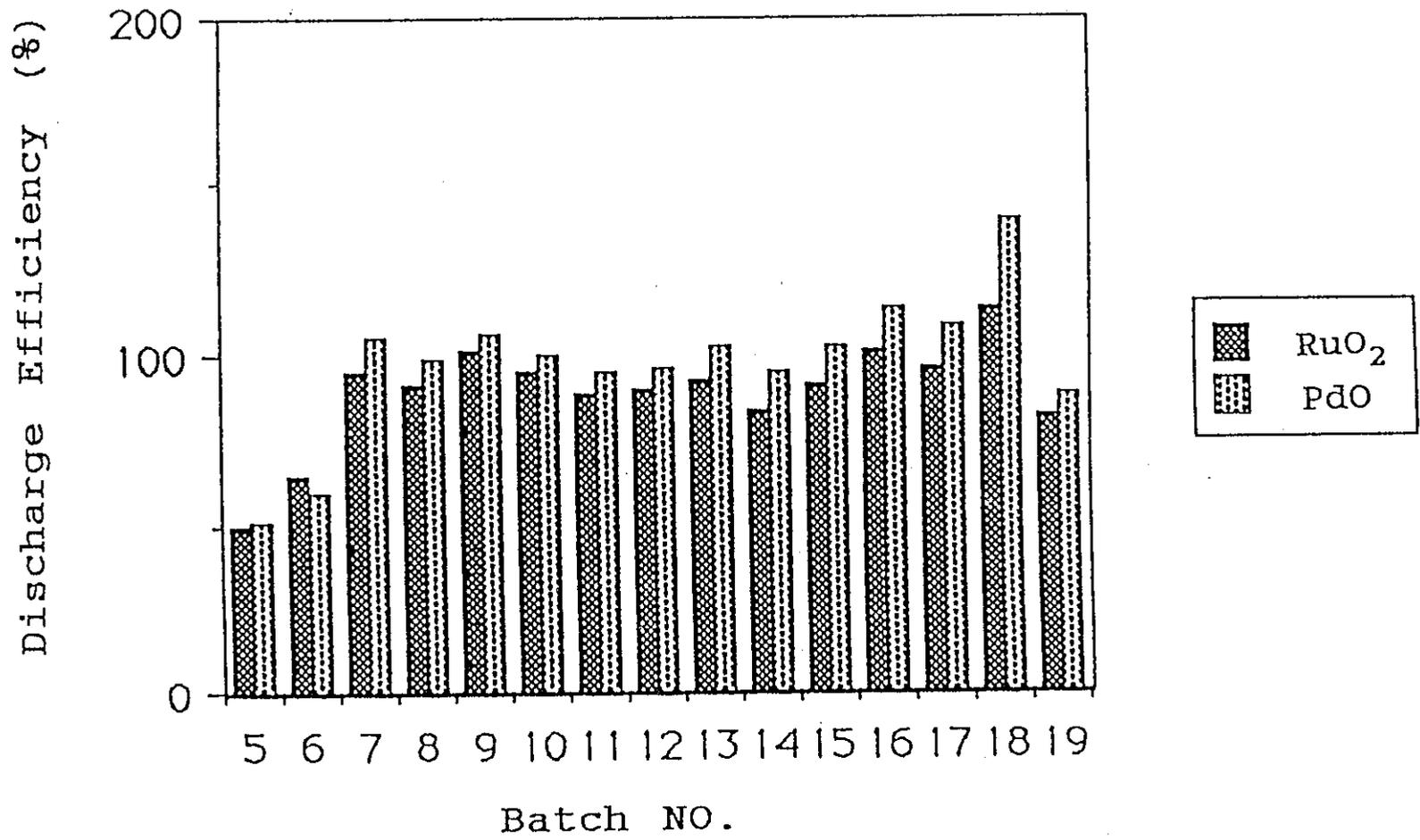
Operational Difference in Control of Melter Bottom Temperature



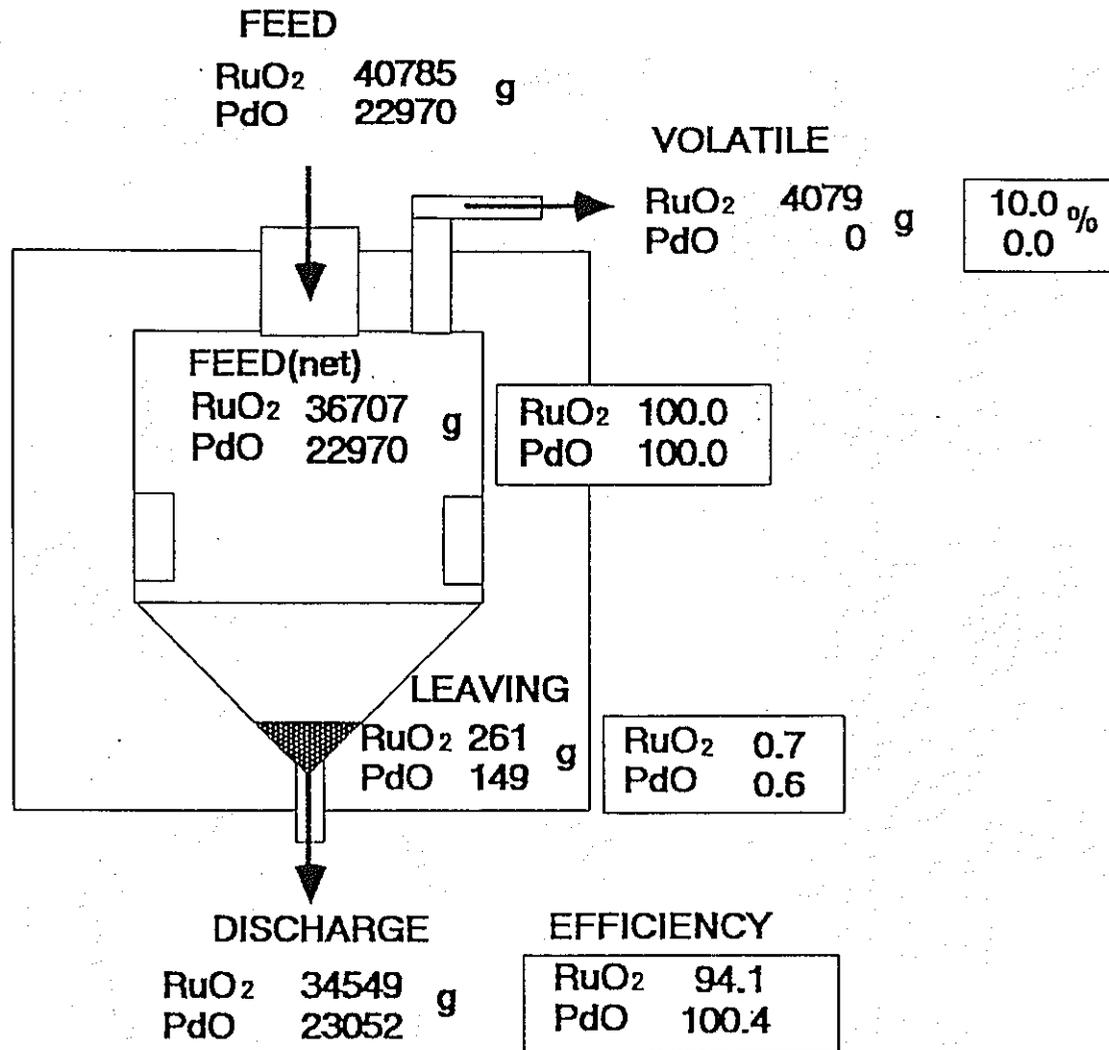
Meller Operation during the Feed of Noble Metals



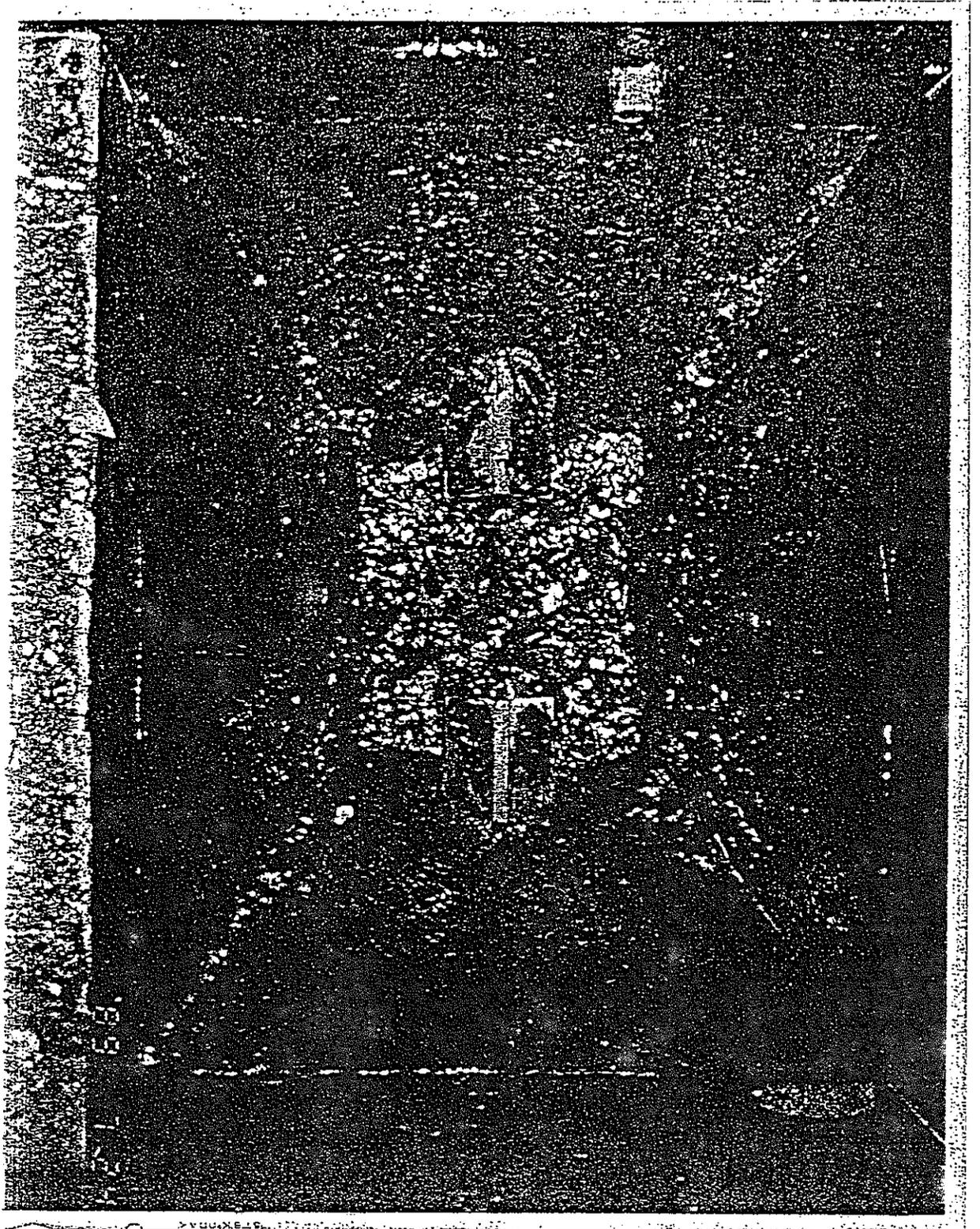
Comparison in Change of RuO₂ Content during the Glass Pouring

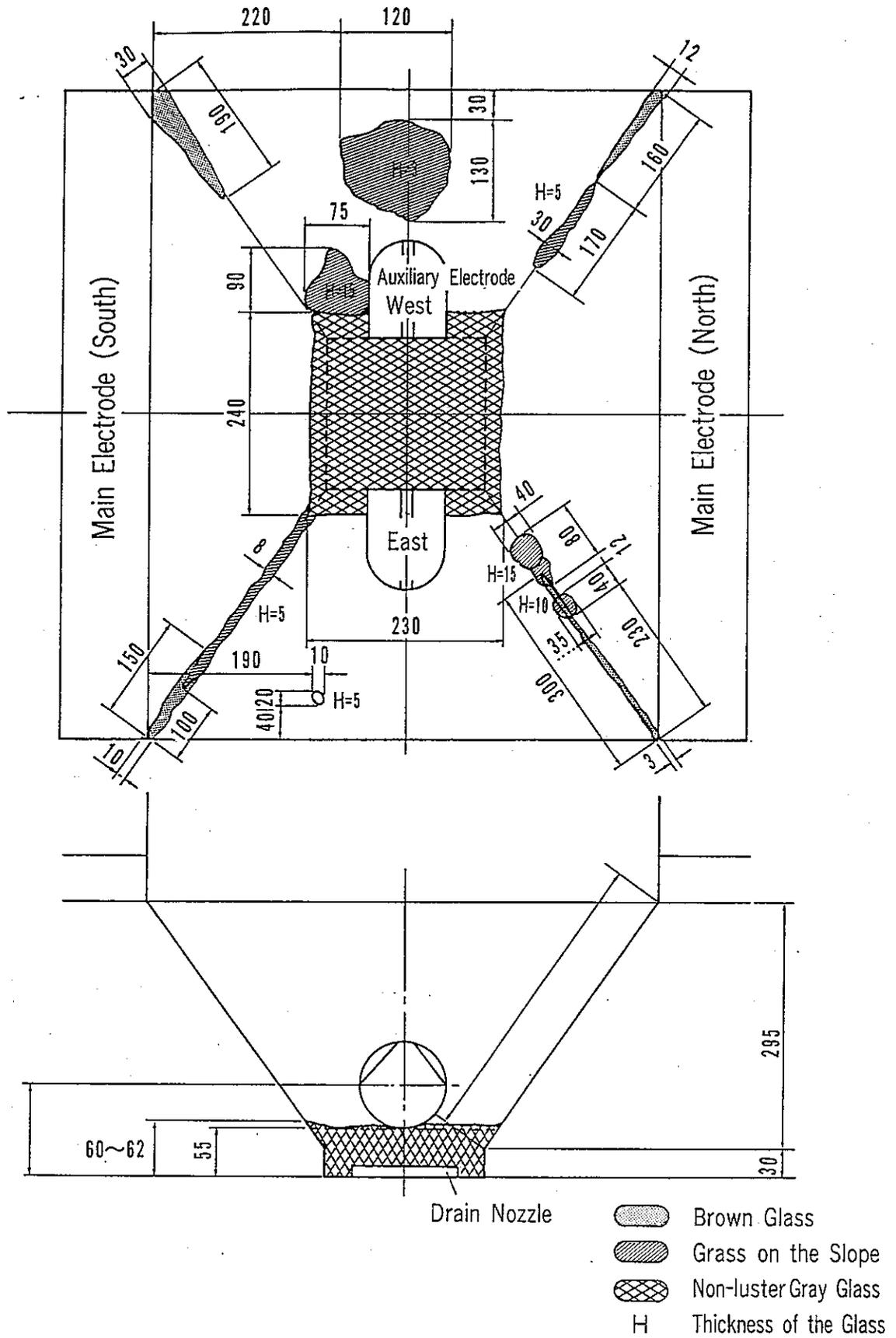


Discharge Efficiency of Noble Metals during the Melter Runs (20th Campaign)

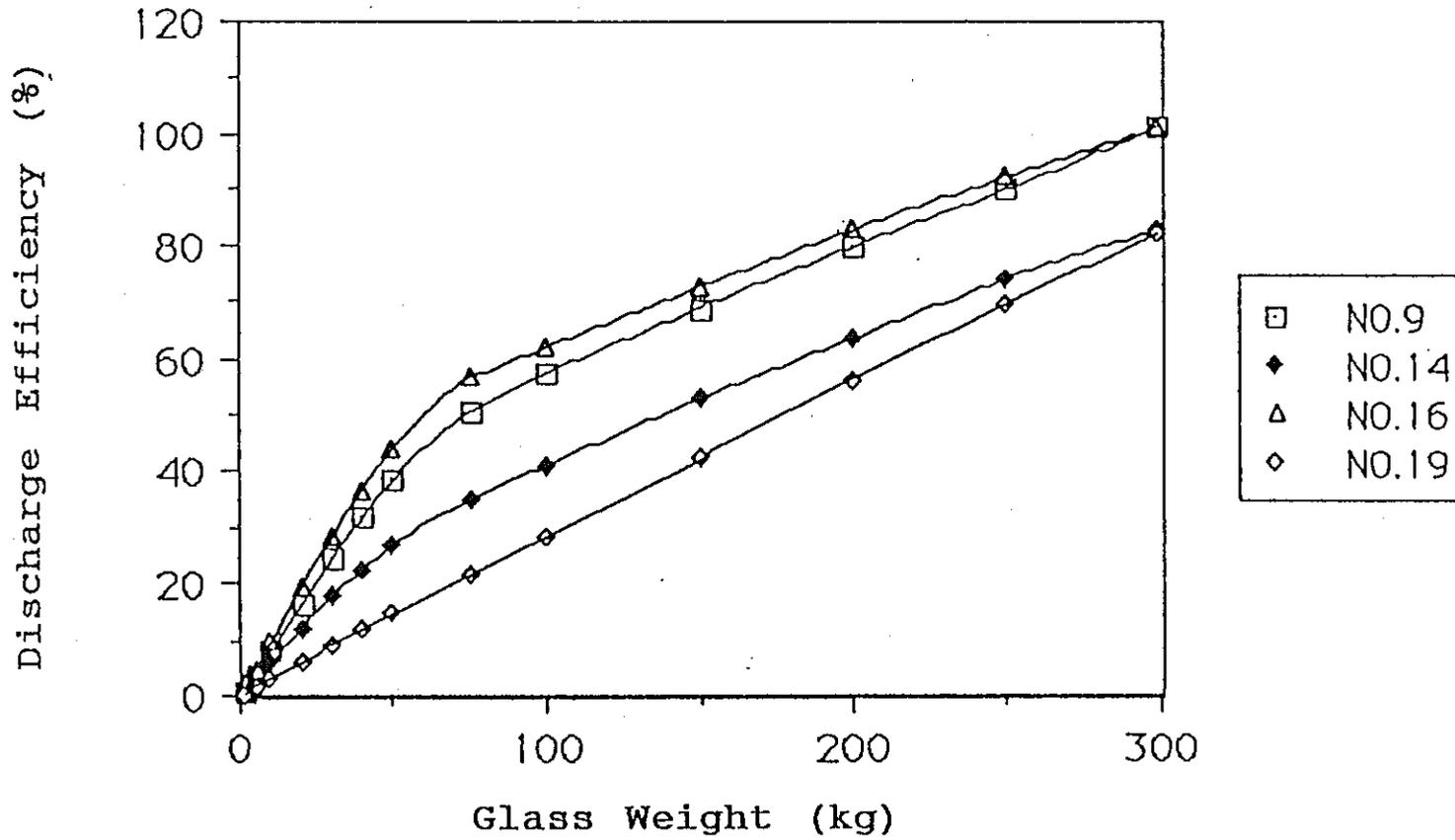


Balance of the Noble Metals in Melter
(20th Campaign)

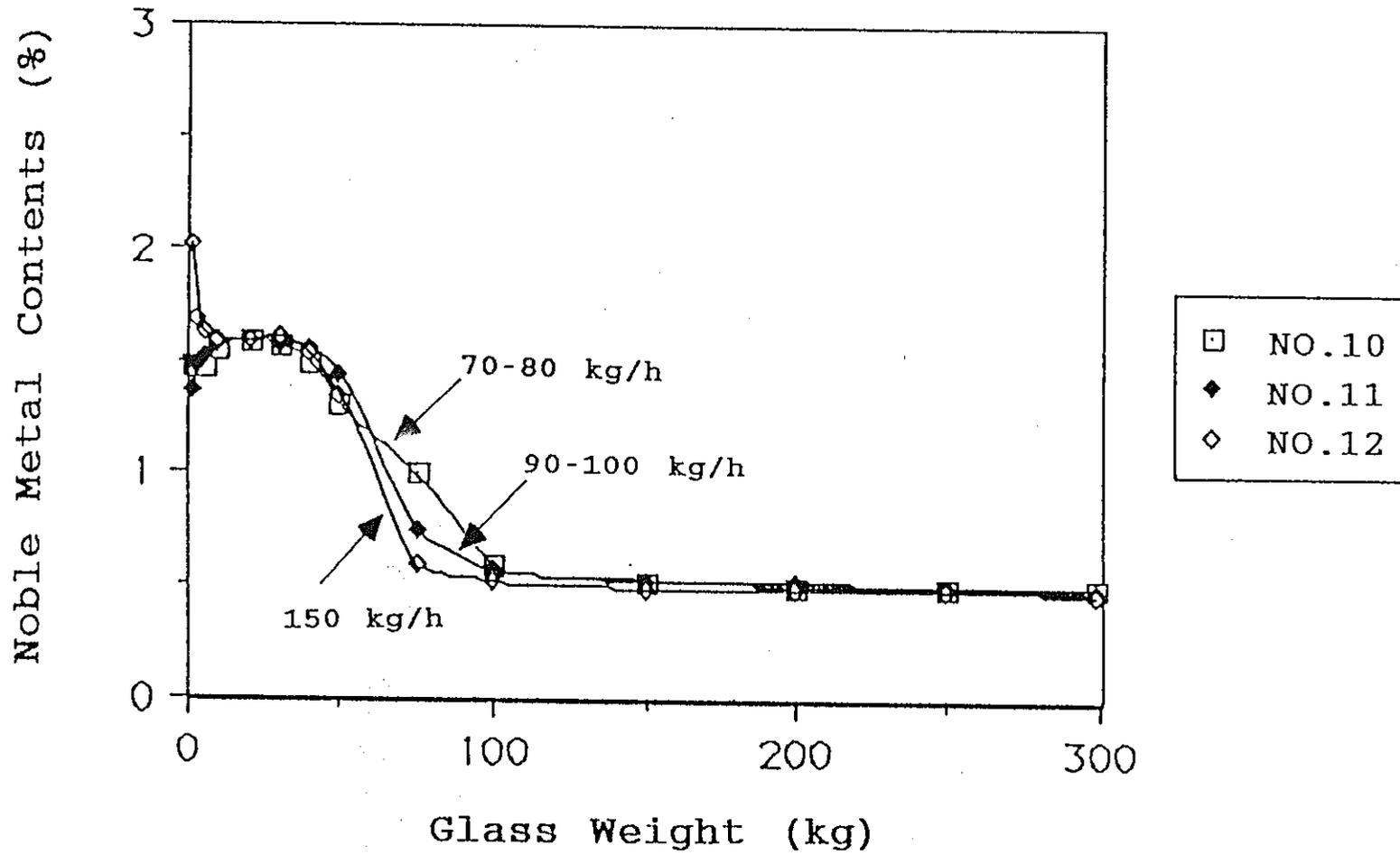




Glass Left on the Melter Bottom after Drain Out (20th Campaign)



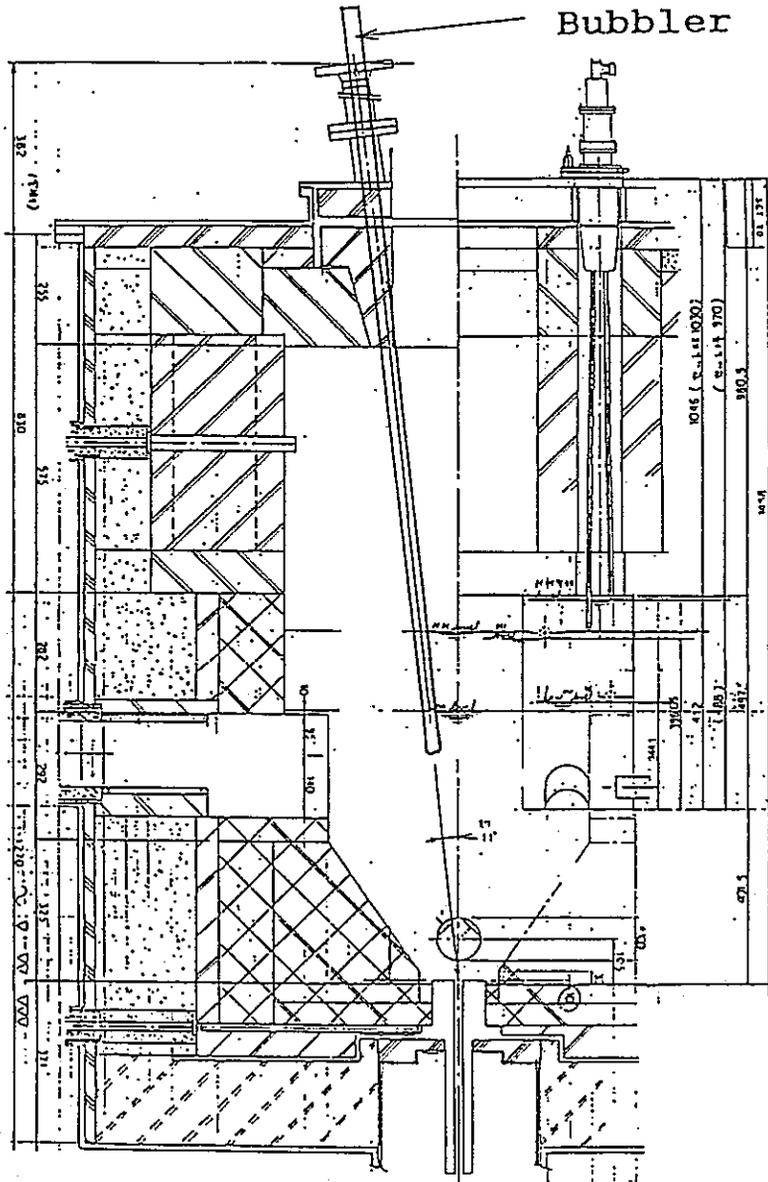
Change of Discharge Efficiency of RuO₂ during the Glass Pouring



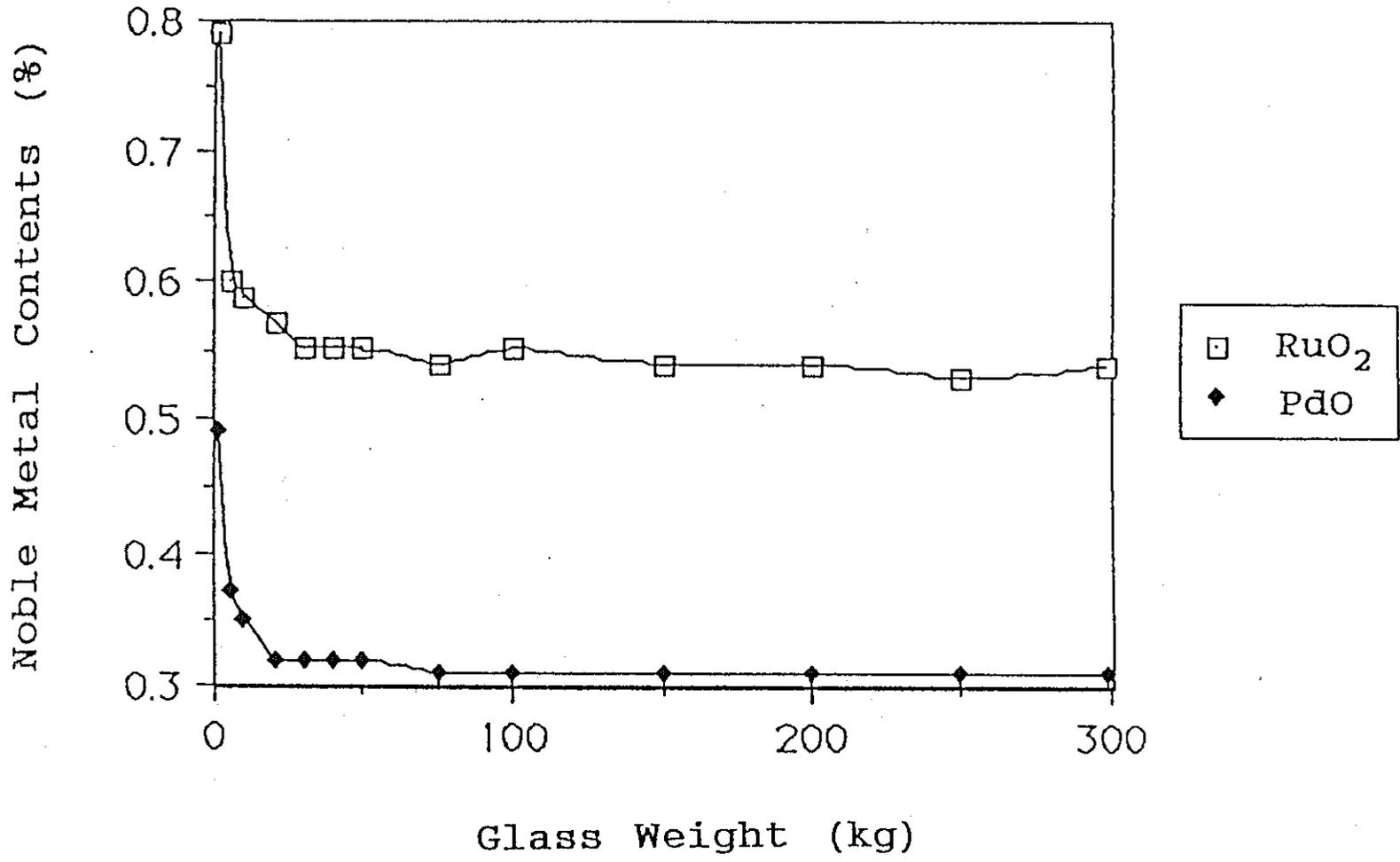
Effect of Pouring Rate to RuO₂ Content

Bubbling Condition

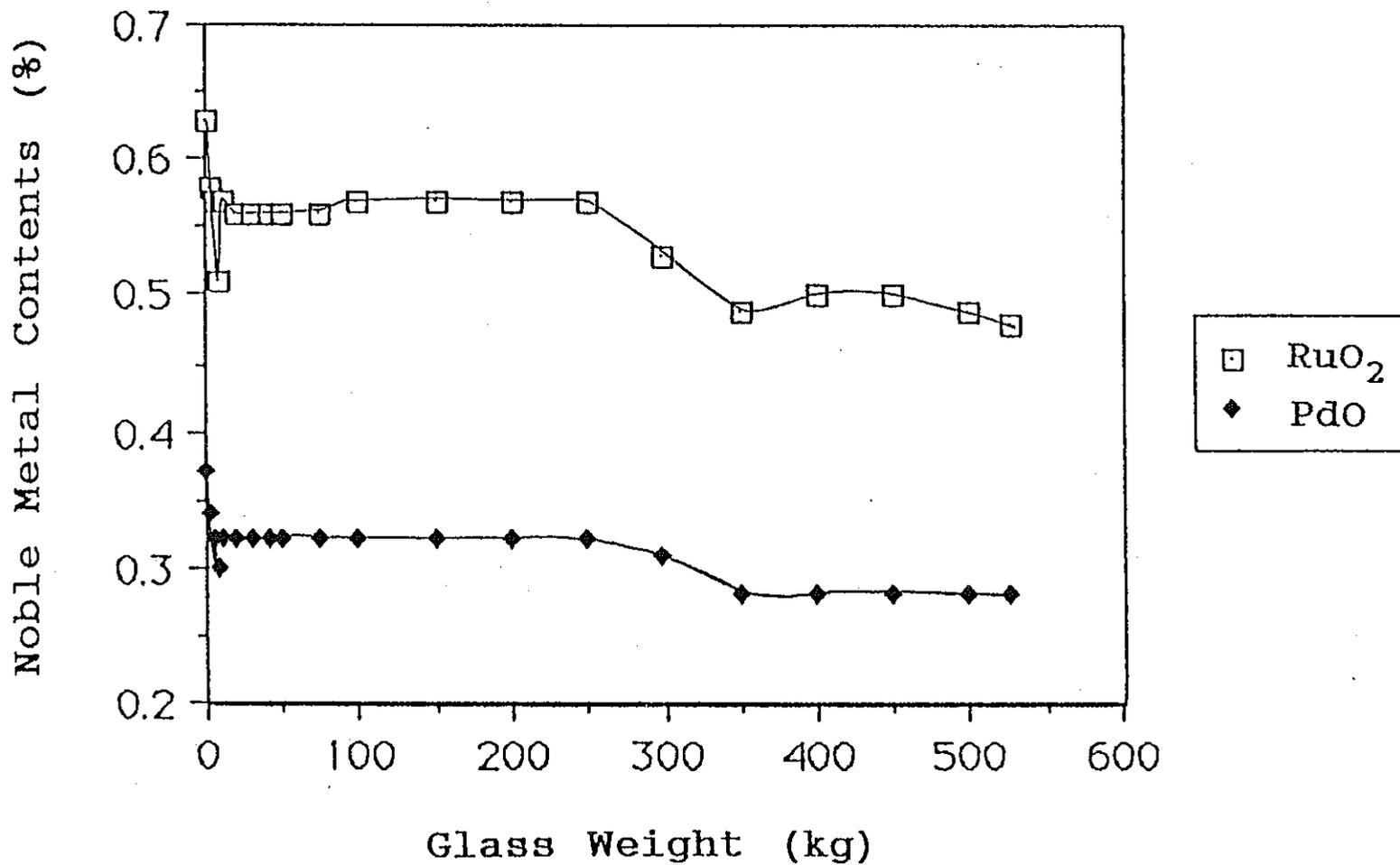
Fluid : Compressed Air
Flow Rate : 800 l/h
Pressure : 3 kg/cm²
Dia. of Bubbler : 18 mm
Bubbling Level : 500 mm from Bottom
Bubbling Time : 47.7 h



Bubbling in the Melter Operation

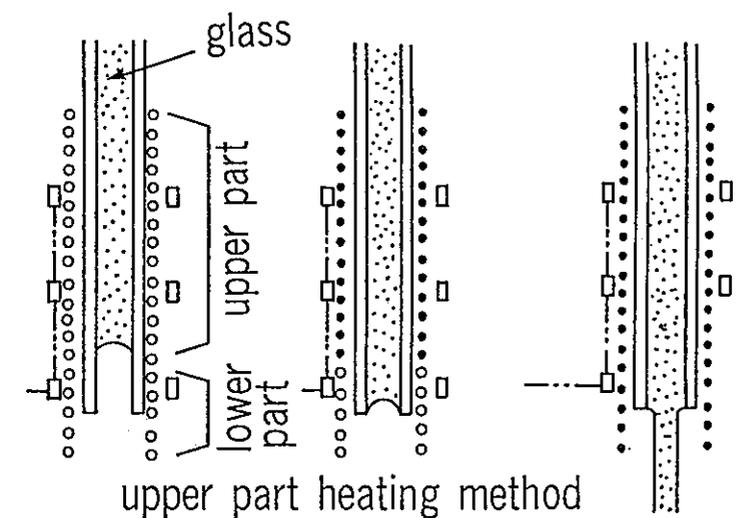
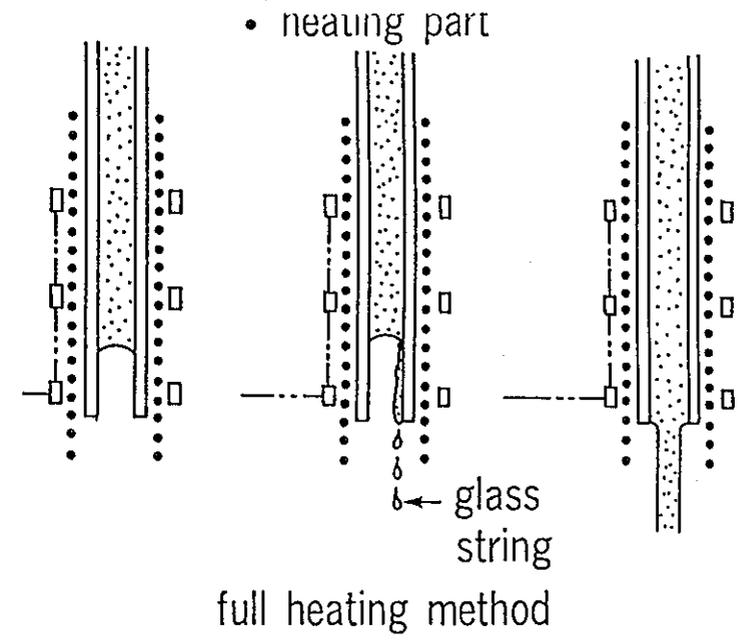
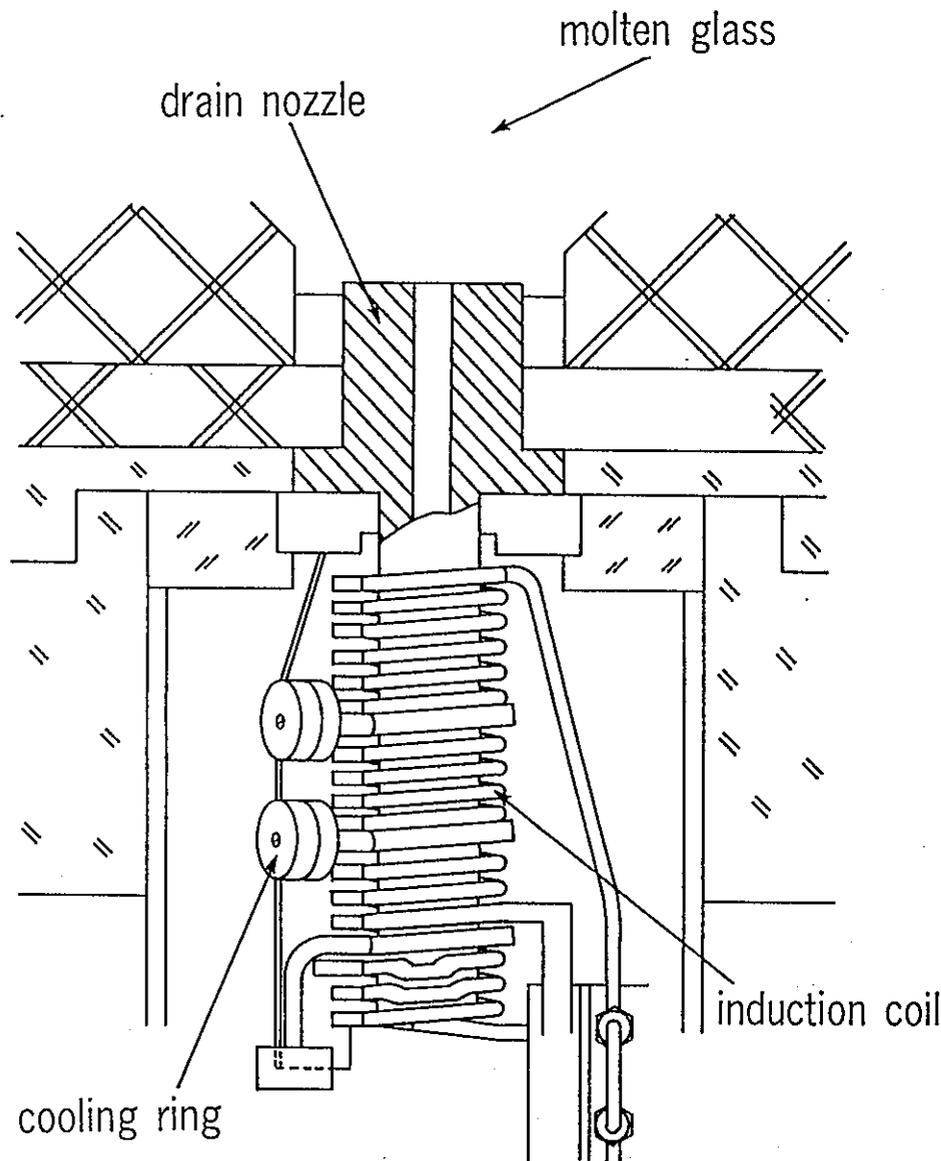


Change of Noble Metal Contents during the Bubbling

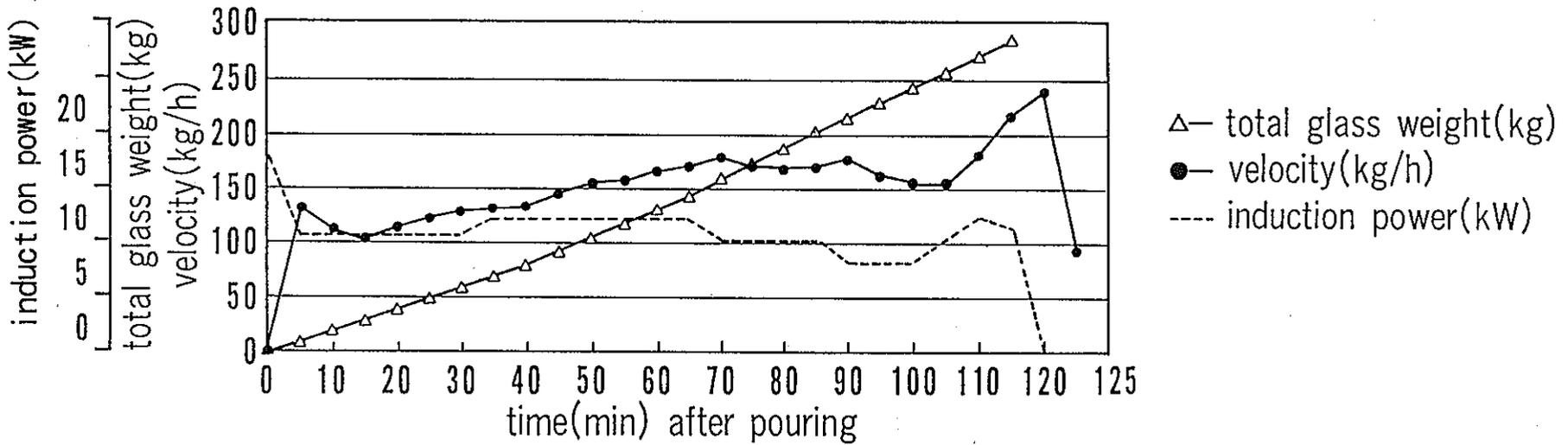


Change of Noble Metal Contents during the Drain Out

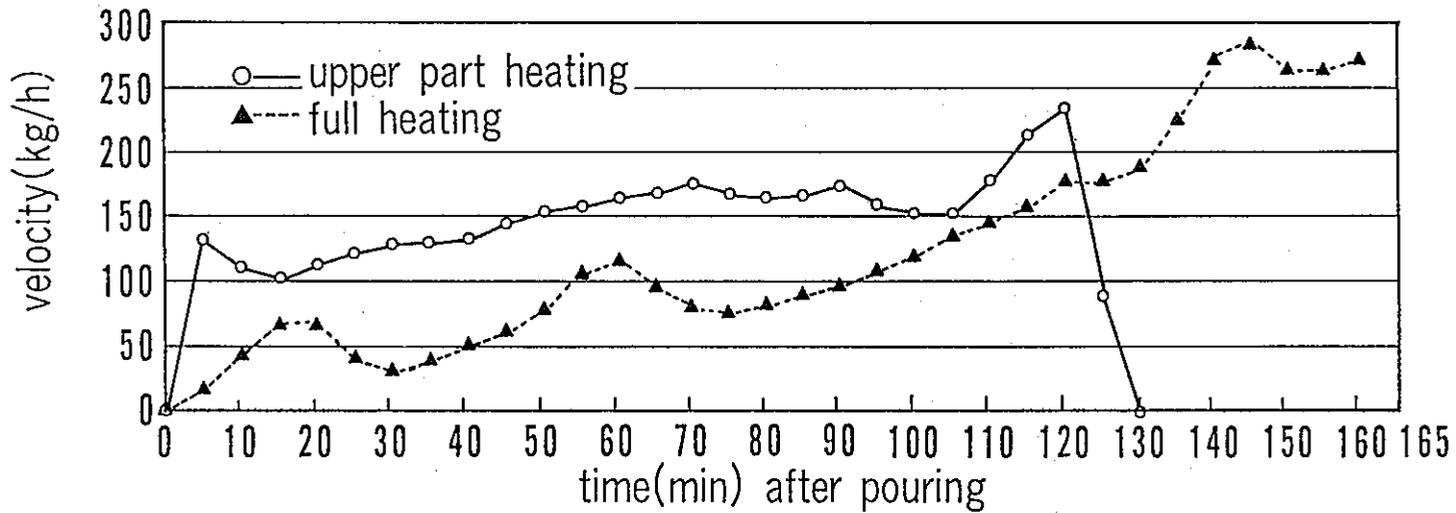
- Development of Glass Pouring Technique in its Initiation
 - Prevention of Pluggage at the Initiation of Glass Pouring
 - Controllability of Glass Flow Rate



Schematic picture of the construction of drain nozzle and nozzle heating method



Velocity control by nozzle induction power



Difference of velocity between upper part heating and full heating

2.3 Characterization of glass containing noble metal element

CHARACTERIZATION OF GLASS CONTAINING NOBLE METAL ELEMENT

The 9th Annual KfK-PNC Meeting on
High-Level Waste Management

October 9-11, 1989

Power Reactor and Nuclear Fuel Development Corporation

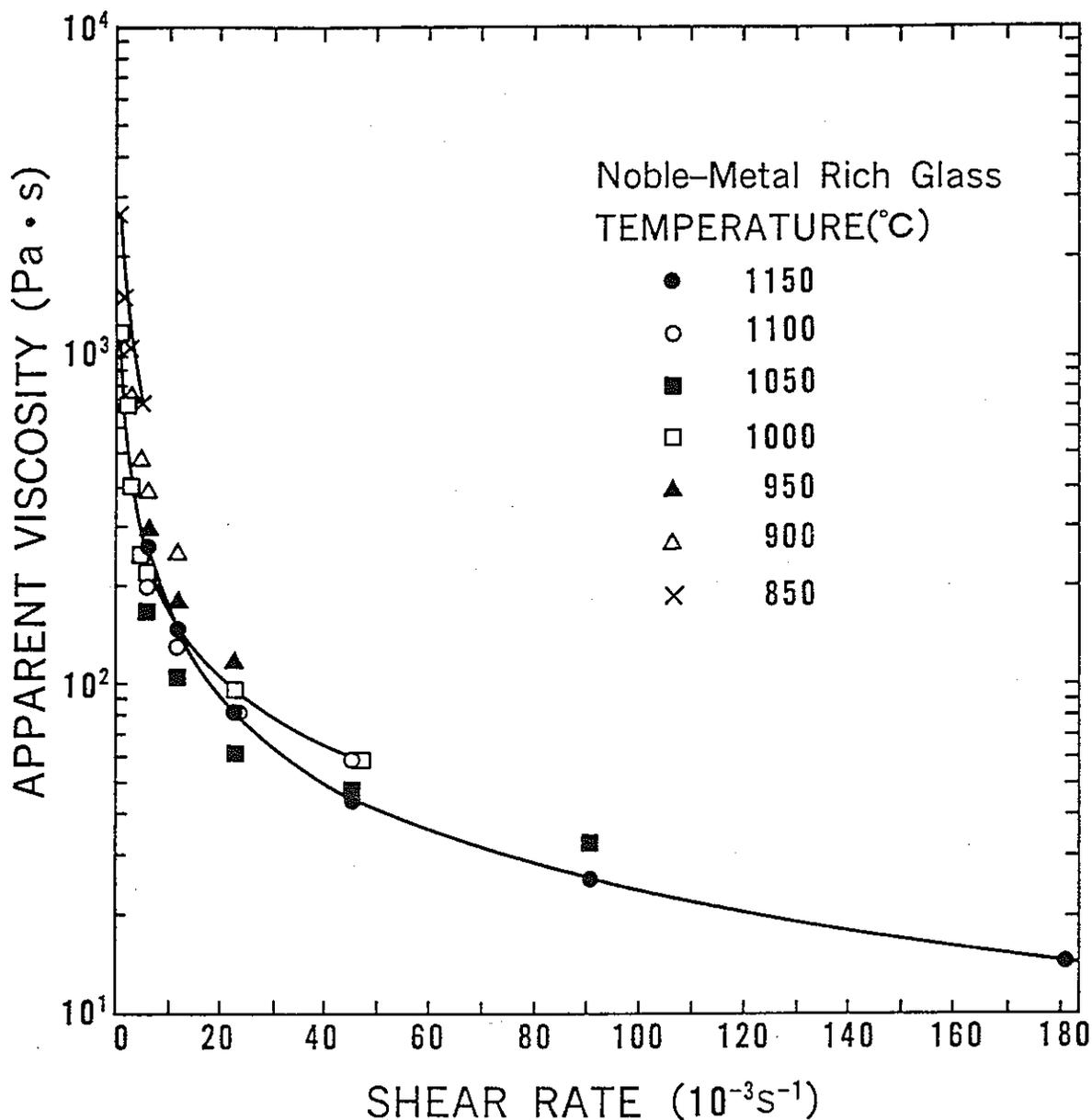
CHARACTERIZATION OF GLASS CONTAINING NOBLE METAL ELEMENT

1. VISCOSITY
2. ELECTRICAL RESISTIVITY
3. SEM/EDX
Melter-Bottom Glass from 19th, 20th Campaign
4. DTA/TG
Pd-Rh-Te Alloy
5. OTHERS
Crystal Growth, Recovery of Noble Metal

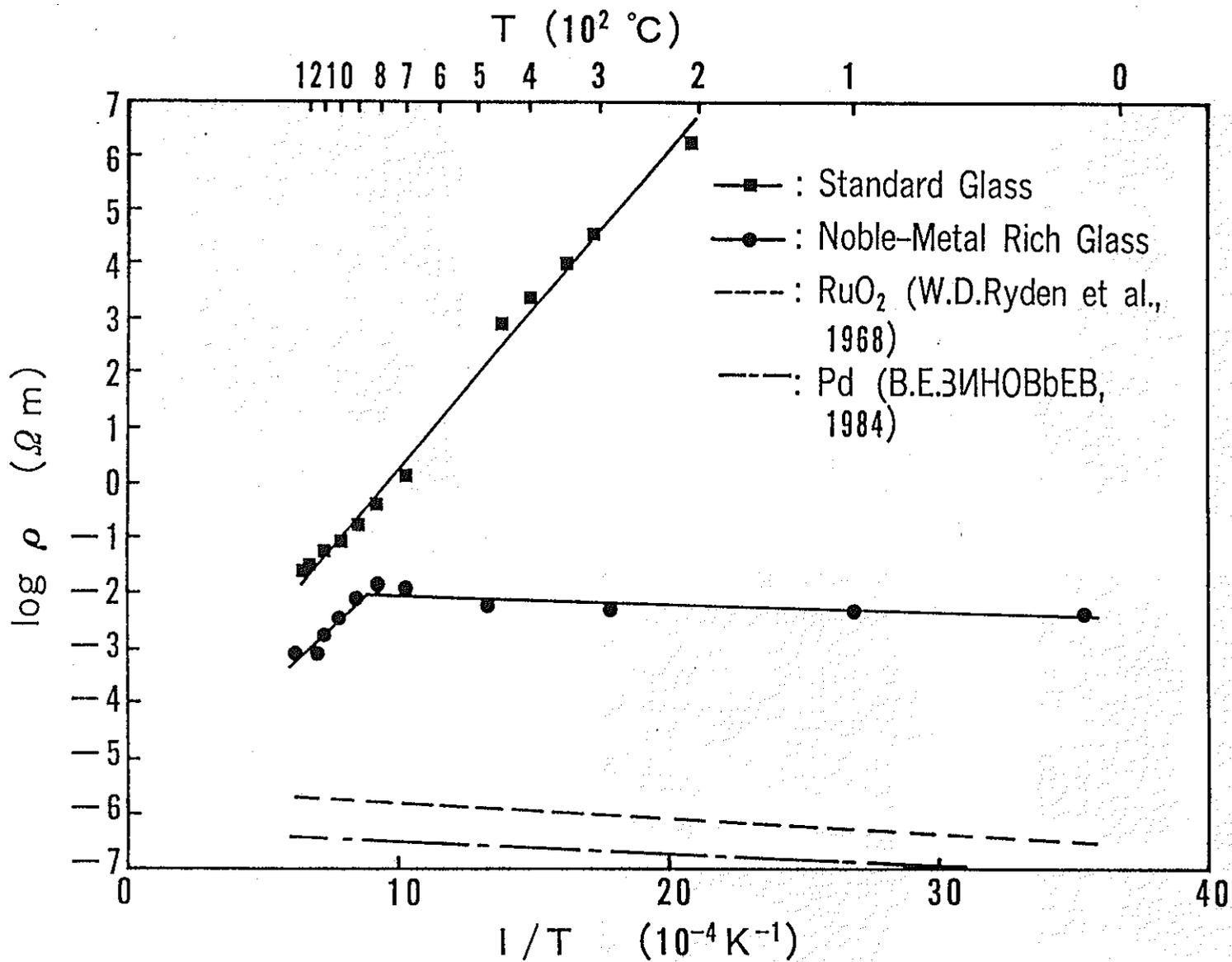
2. SPECIMEN SIMULATED HIGH LEVEL WASTE GLASS

COMPOSITION (wt%)	STANDARD WASTE GLASS	NOBLE-METAL RICH GLASS
GLASS ADDITIVE	75	65
SiO ₂	47	40
B ₂ O ₃	14	12
Al ₂ O ₃	5	4
Li ₂ O	3	3
CaO	3	3
ZnO	3	3
WASTE	25	35
Na ₂ O	10	8
NOBLE-METAL	1	17
RuO ₂	0.7	13.0
Rh ₂ O ₃ (Rh)	0.2 (0.1)	—
PdO (Pd)	0.3 (0.3)	3.5(3.0)
OTHERS	14	10
TOTAL	100	100

6. VISCOSITY

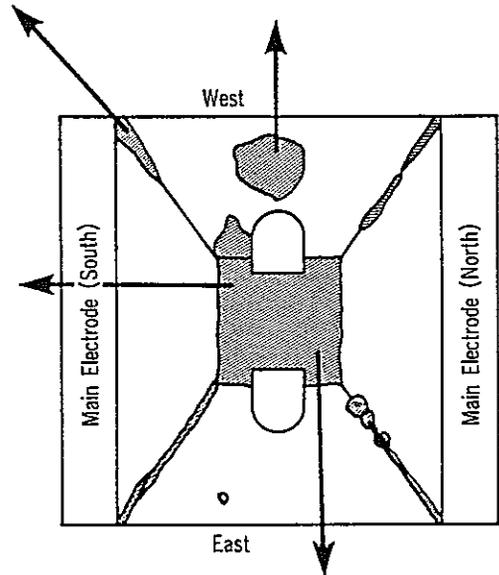
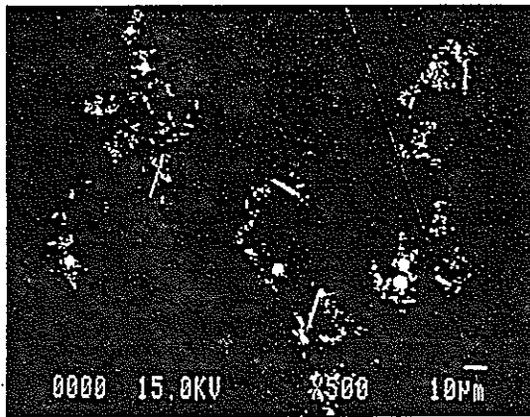
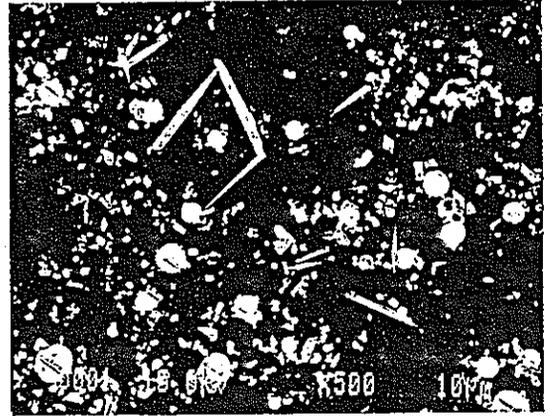
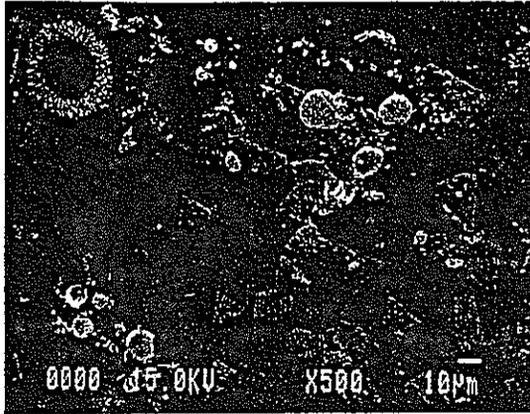


Apparent viscosity of noble-metal rich glass. (改造Bメルタ 第4キャンペーン)

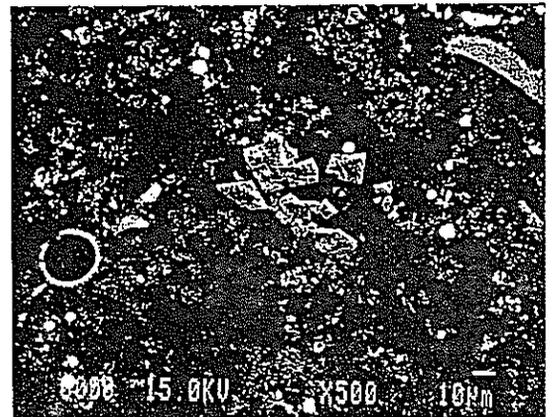


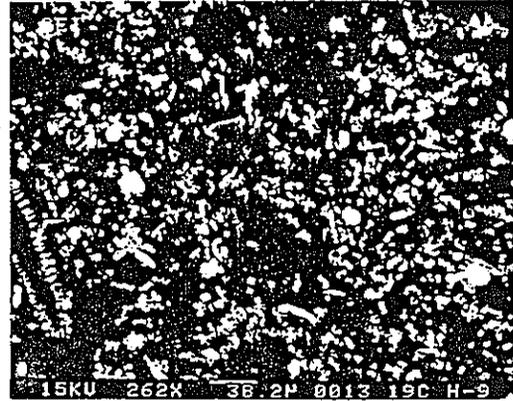
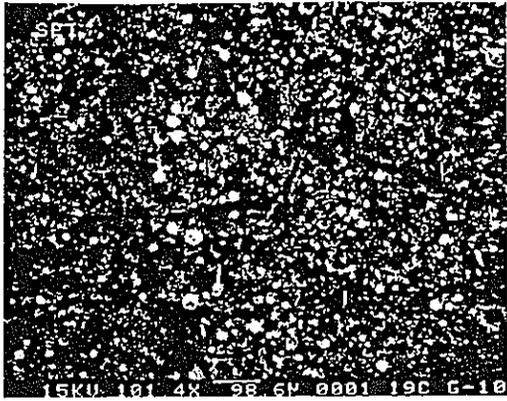
Electrical resistivity of waste glasses and noble metals.

(改造Bメルト 第4キャンペーン)



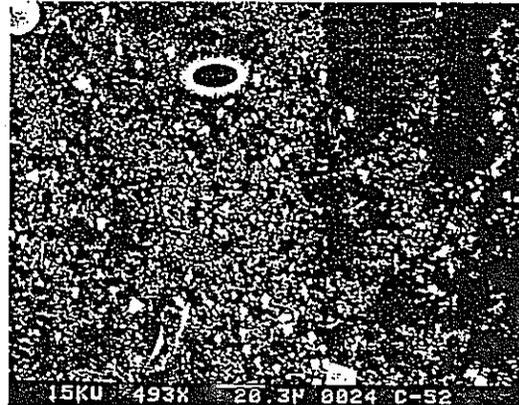
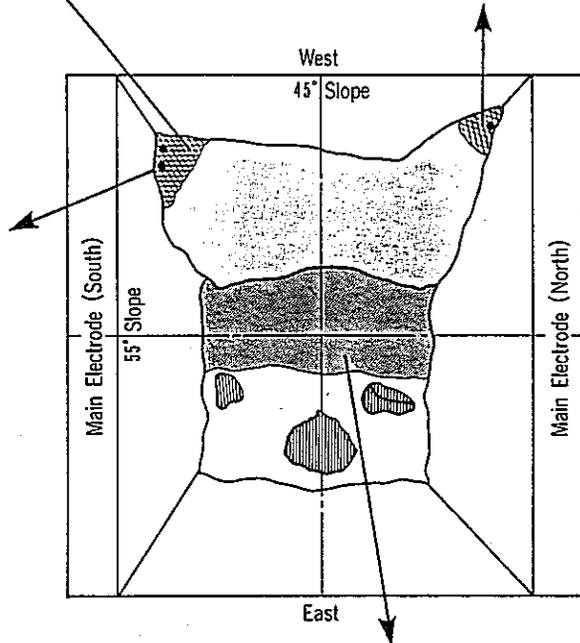
20th Campaign
Melter-Bottom Glass



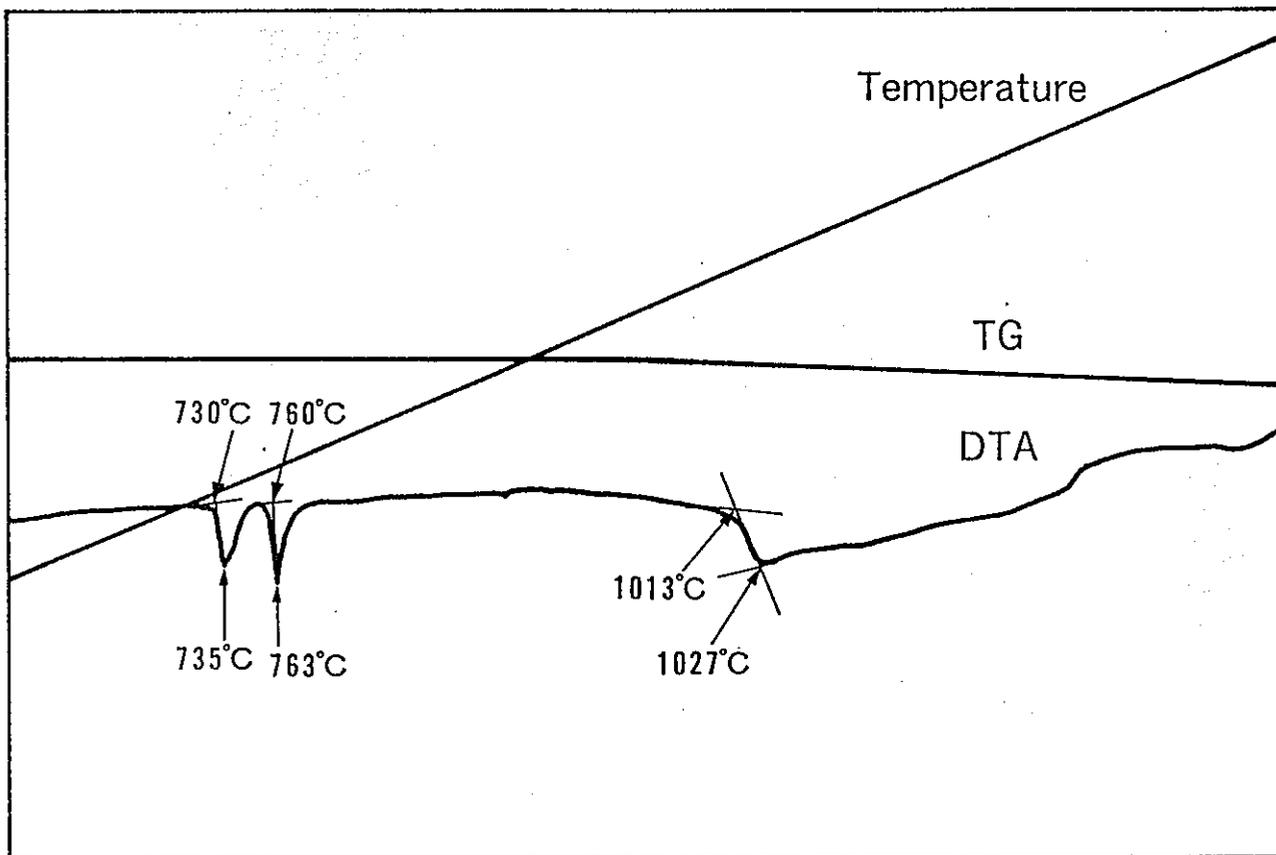


77Pd-12Rh-8 Te
3.2g, 10.6Mg/m³

19th Campaign
Melter-Bottom Glass and
Nodules of Noble Metal Alloy



19th Campaign Noble Metal Nodule
DTA/TG





—|—
0.1mm

19th Campaign Noble Metal Nodule
aqua regia etching, 5s, R.T.

white part : 40Pd—56Rh— 1 Te— 3 Ru

gray part : 84Pd— 1 Rh— 9 Te

black part : 87Pd— 1 Rh— 8 Te

2.4 Study on Absorption of Volatile Ruthenium into Water

Study on Absorption of Volatile Ruthenium into Water

The 9th Annual KfK-PNC Meeting on
High-Level Waste Management

October 9-11, 1989

Power Reactor and Nuclear Fuel Development Corporation

Objective

Treatment of volatile ruthenium in the off gas
from the melter of vitrification process

- Removal by absorption in wet scrubbing process
 - Adsorption to silica gel column
- Study of the influence of variation in the operation of the process

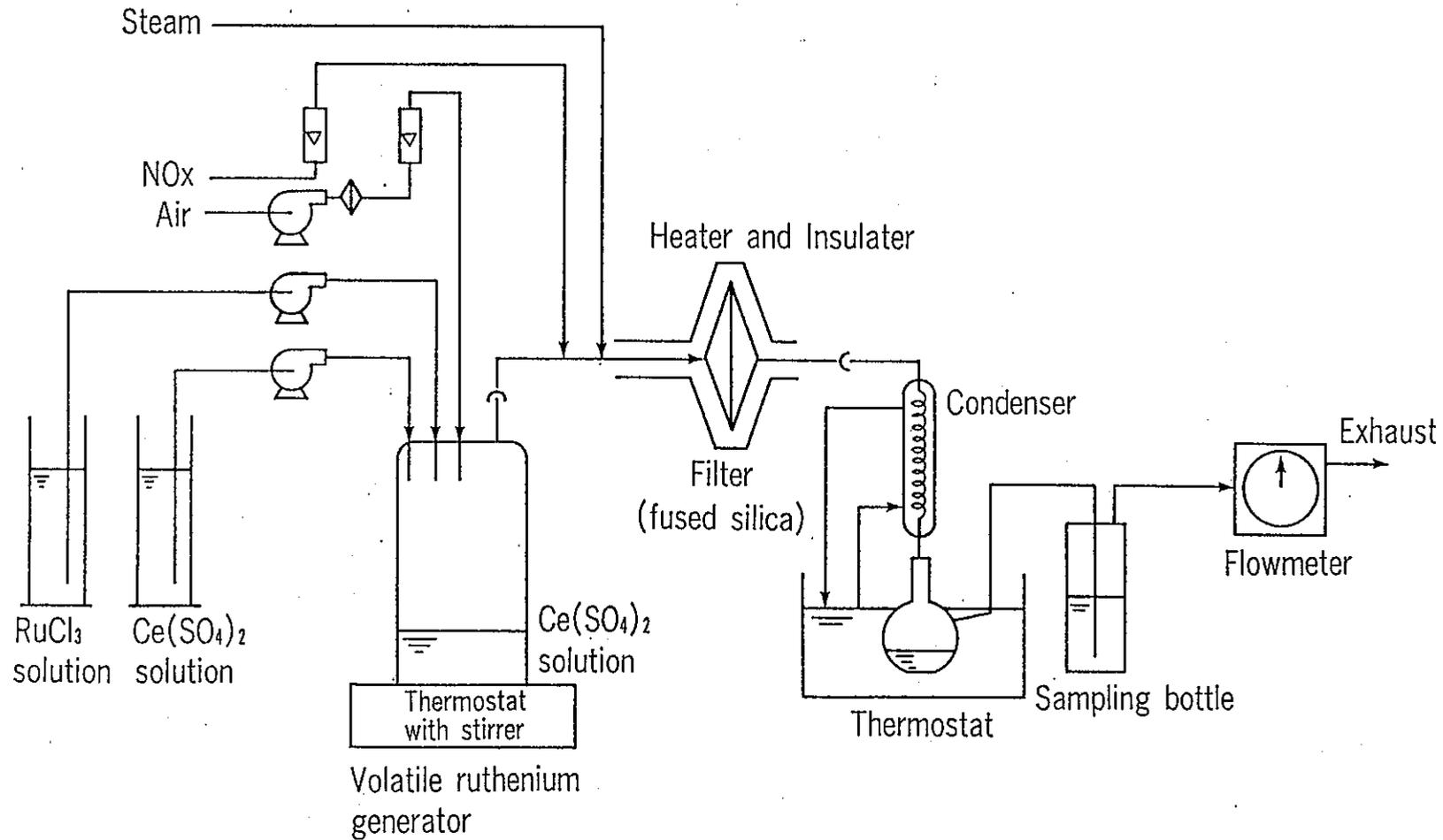
Absorption behaviour of volatile ruthenium

— Absorption behaviour into condensates for steam condensation —

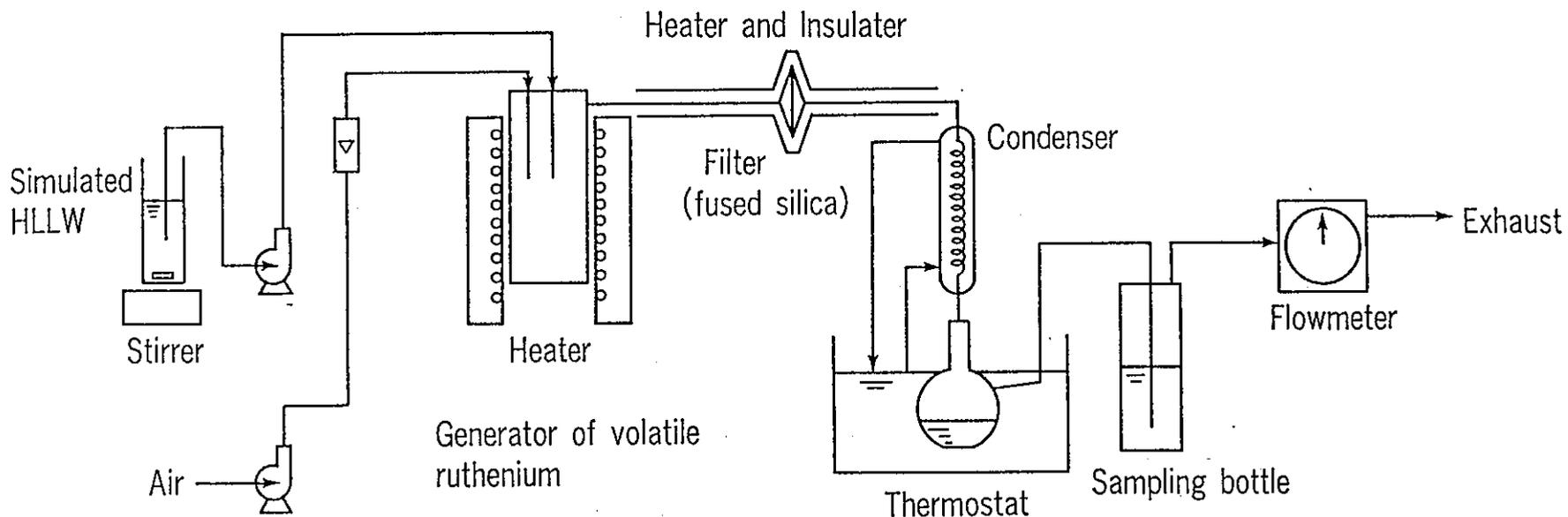
1. Effect of NO_x in off gas on absorption of RuO₄
2. Effect of condensation temperature on absorption of volatile ruthenium from calcining process

Experimental Method

Generation of Volatile Ru	RuO ₄	Oxidation of RuCl ₃ by Ce (SO ₄) ₂
	Volatile Ru from melter	Calcining of simulated high-level liquid waste (~300°C)
Trap of Ru	Aerosol Ru	Filter made of fused silica fiber
	Volatile Ru	(1) Absorbtion into condensates by chilling (2) Absorbtion into 6N-HCl with 1%C ₂ H ₅ OH
Evaluation for absorption of volatile Ru	Concentration ratio of Ru betwen gas and water : m $m = \frac{\text{Ru in off gas after condenser (mol fraction)}}{\text{Ru in condensates (mol fraction)}}$	



RuO_4 absorption test apparatus



Volatile ruthenium absorption test apparatus for off-gas from calcining simulated high-level liquid waste (HLLW)

Experimental Condition

(Effect of NO₂ in off gas on absorption of RuO₄)

Experimental Gas					Condensation temperature (°C)	Liq/Gas ratio (mol/mol)
Air (l/min)	H ₂ O (cc/min)	NO ₂ (ppm)	Ru(gas) (ppm)	NO ₂ /Ru ratio		
2.0	1.0	0	0.81	0	21	0.7
1.0	1.0	10.8	0.63	17	20	1.1
1.0	1.0	26.6	0.79	34	20	1.2
1.0	1.0	66.2	0.52	126	20	1.6
1.0	0.9	112	0.59	189	20	1.2
1.0	0.9	301	0.77	391	20	1.4

Experimental Condition

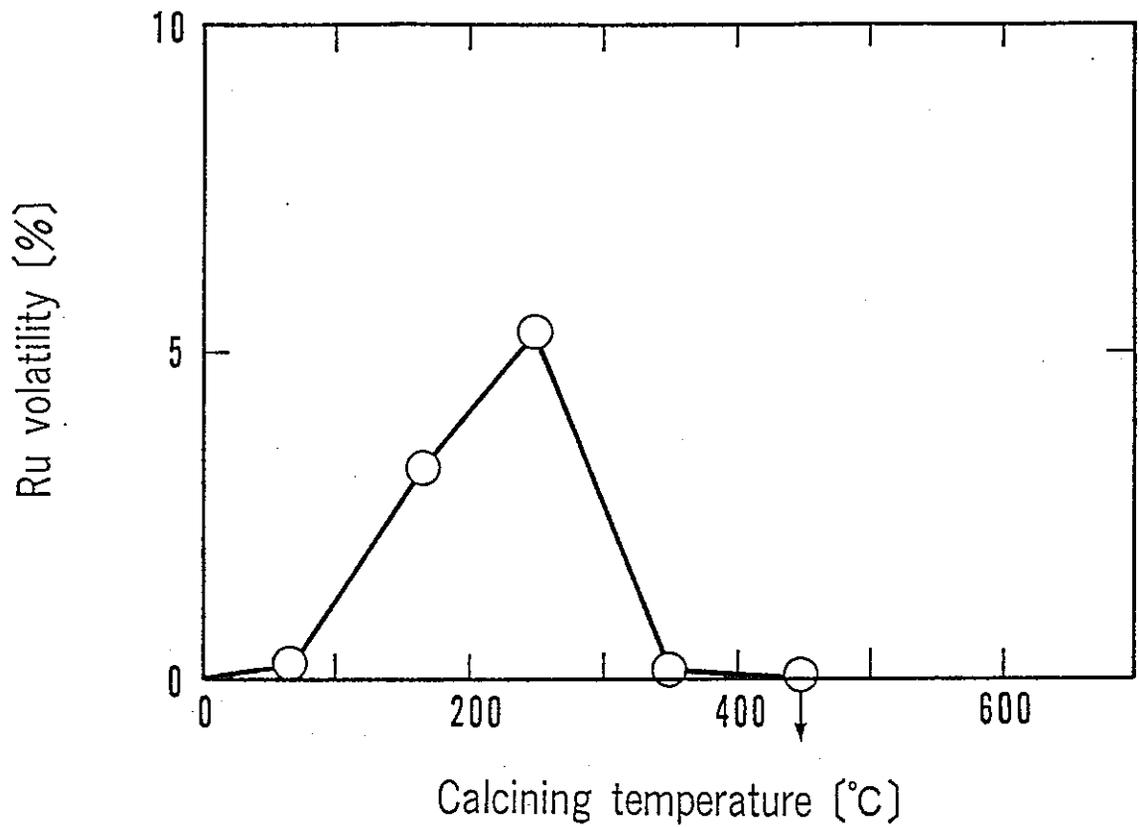
(Effect of NO in off gas on absorption of RuO₄)

Experimental Gas					Condensation temperature (°C)	Liq/Gas ratio (mol/mol)
Air (l/min)	H ₂ O (cc/min)	NO (ppm)	Ru(gas) (ppm)	NO/Ru ratio		
1.0	1.0	0	22.6	0	16	1.3
1.0	0.8	51	4.9	10	15	0.9
1.0	0.9	100	6.9	14	15	1.0
1.0	0.8	163	7.3	22	16	0.8

Experimental Condition

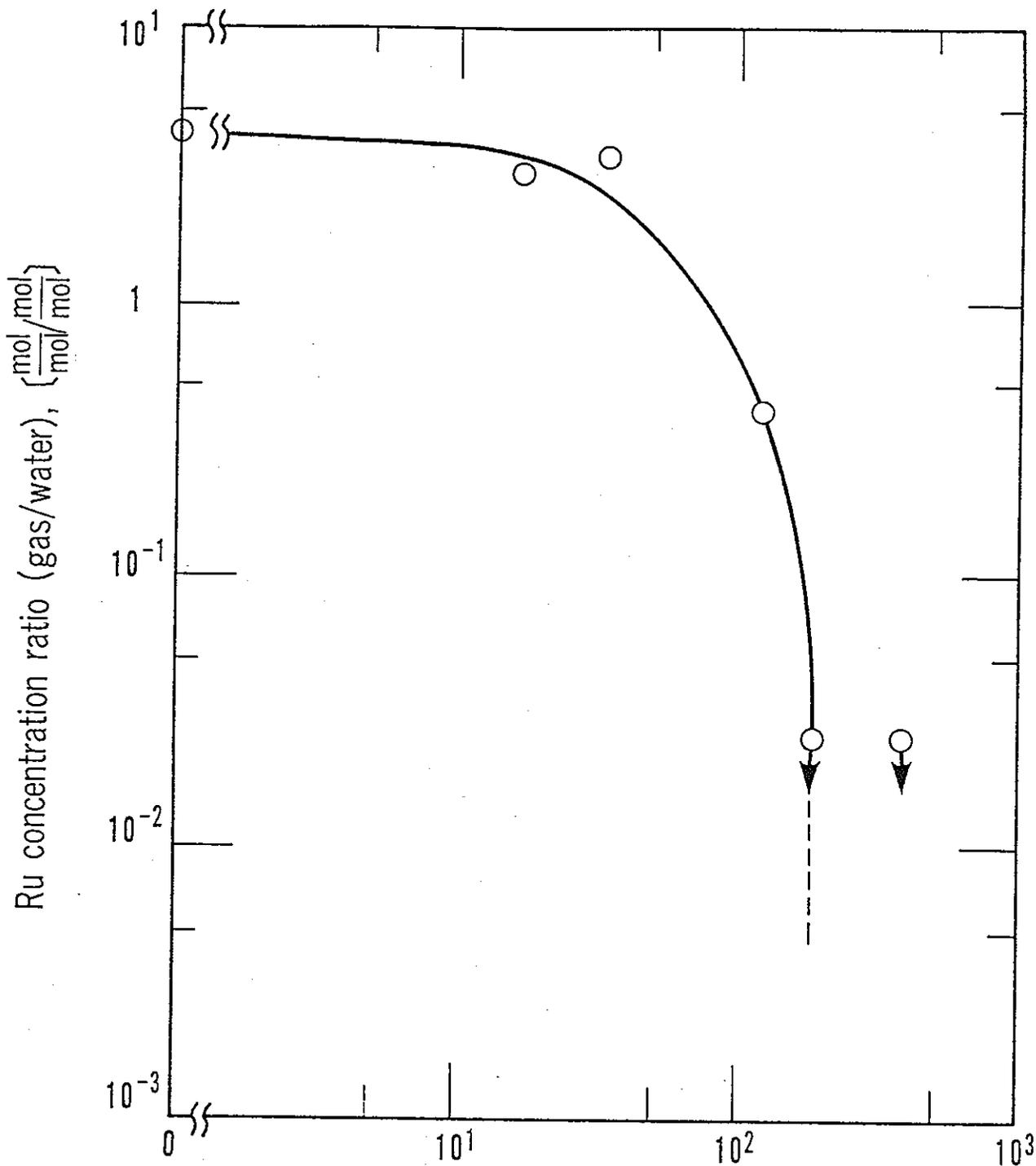
(Effect of condensation temperature)

Simulated HLLW feed rate (cc/min)	Calcining temperature (°C)	Purge air feed rate (l/min)	Ru(gas) concentration (ppm)	Condensation temperature (°C)	Liq/Gas ratio (mol/mol)
1.0	300	0.5	99	20	2.3
1.1	300	0.5	78	41	2.2
1.5	300	0.5	116	62	2.7
1.6	300	0.5	72	83	2.2



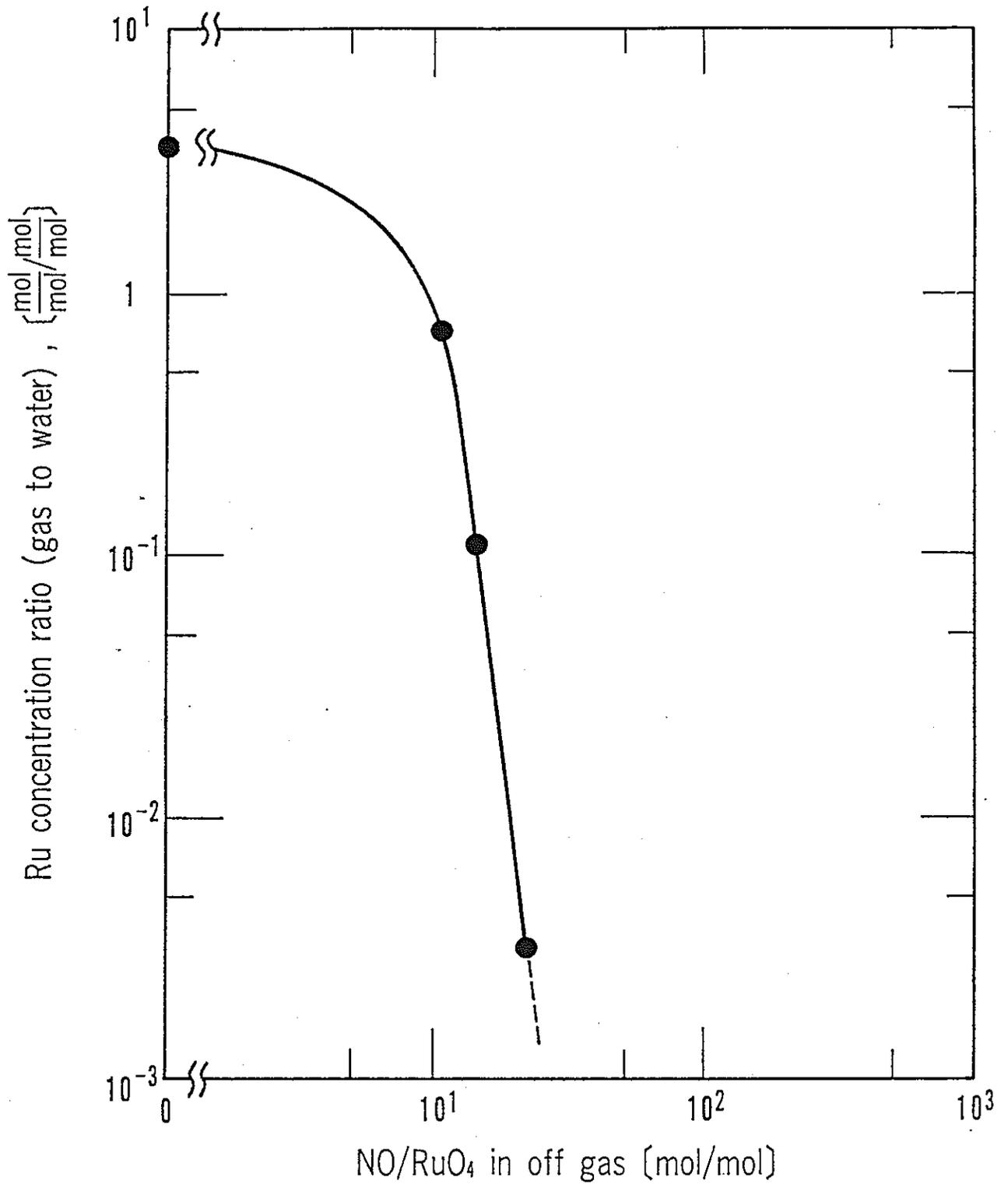
Effect of calcining temperature on Ru volatility for simulated high-level liquid waste

○ denotes under detection limit

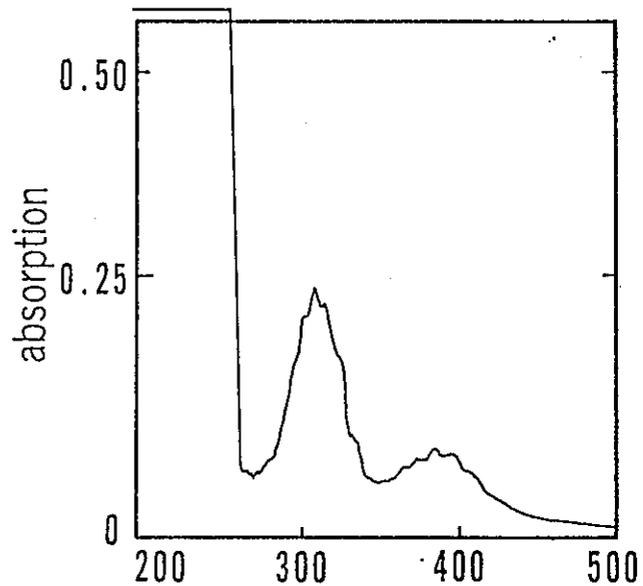


Effect of NO₂ in off gas on RuO₄ concentration ratio (gas to water)

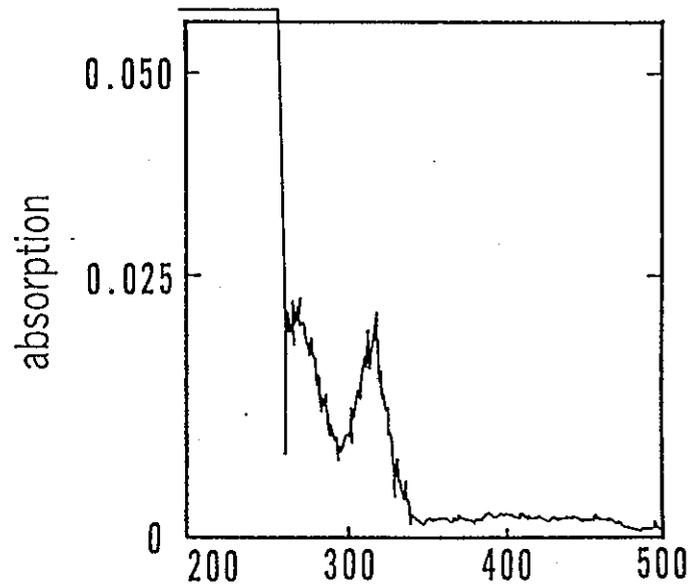
⊕ denotes under detection limit



Effect of NO in off gas on RuO₄ concentration ratio (gas to water)

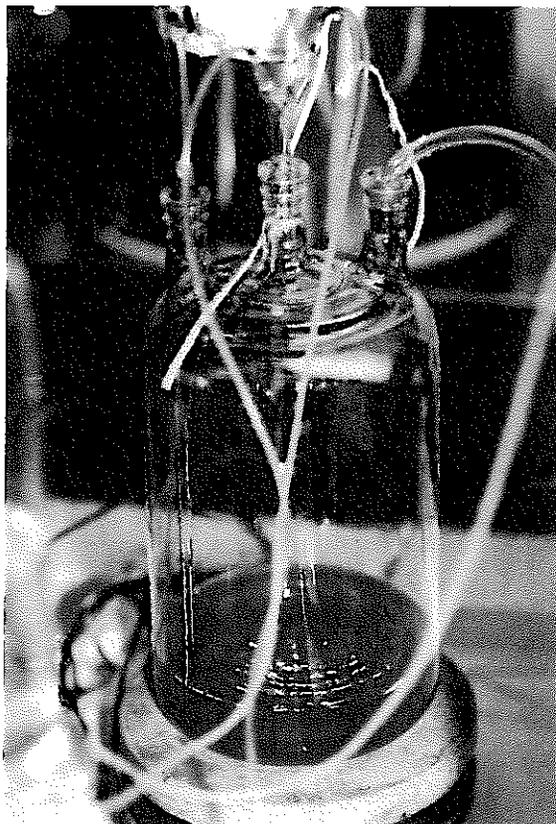


wave length (nm)
without NOx

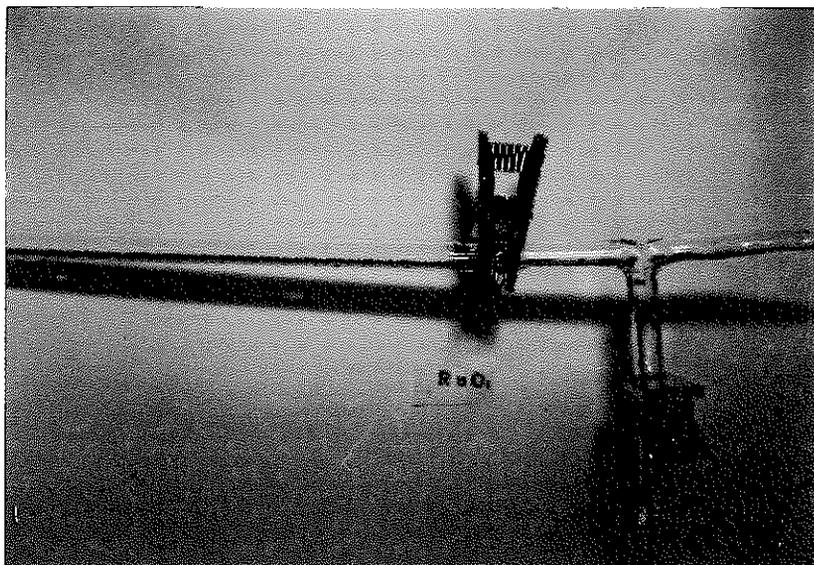


wave length (nm)
Addition of NO

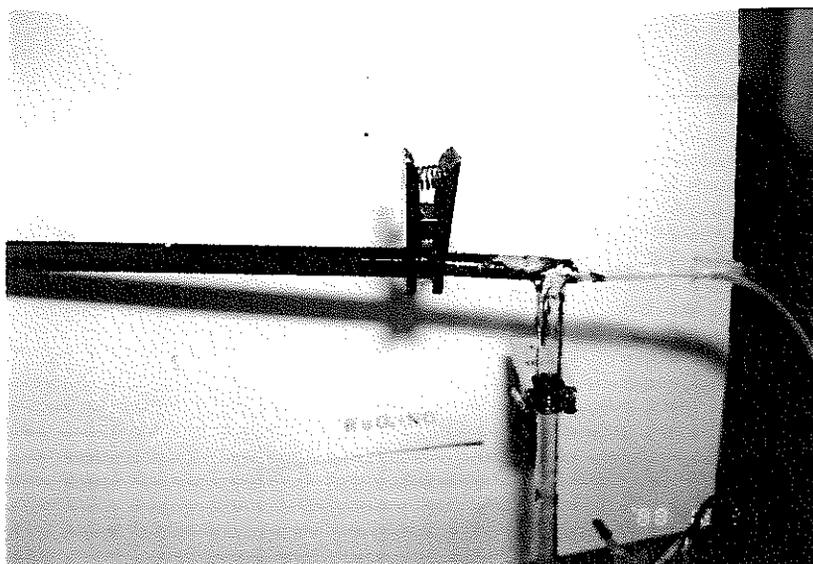
Identification of RuO_4
(absorption spectrum for Ru trapped in CCl_4)



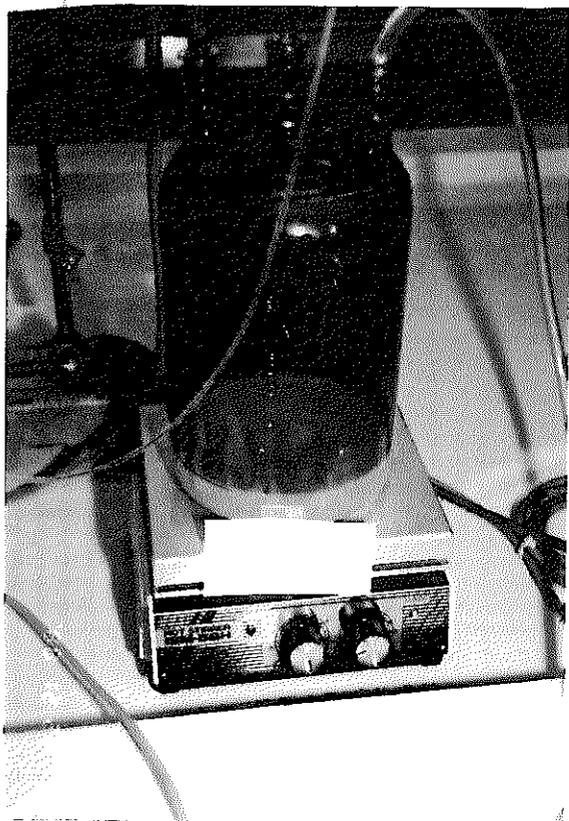
RuO_4 generation



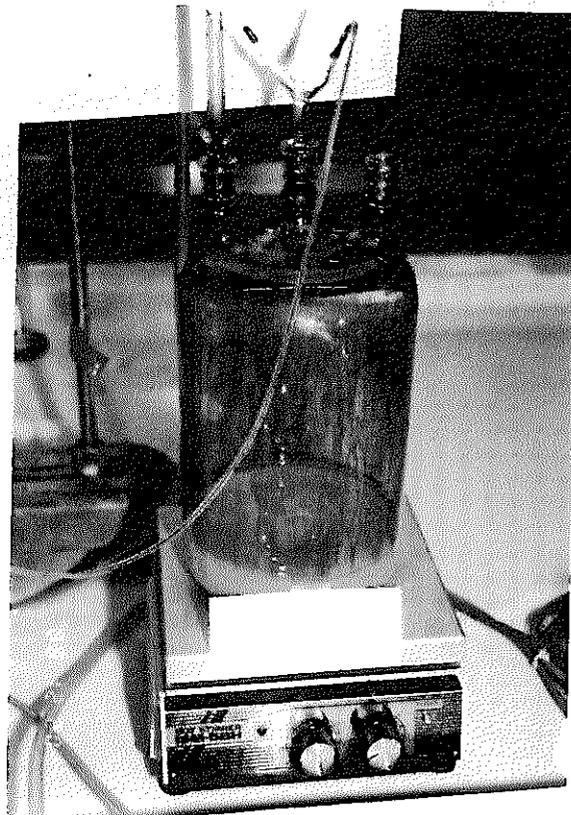
$\text{RuO}_4 + \text{Air}$ through glass pipe



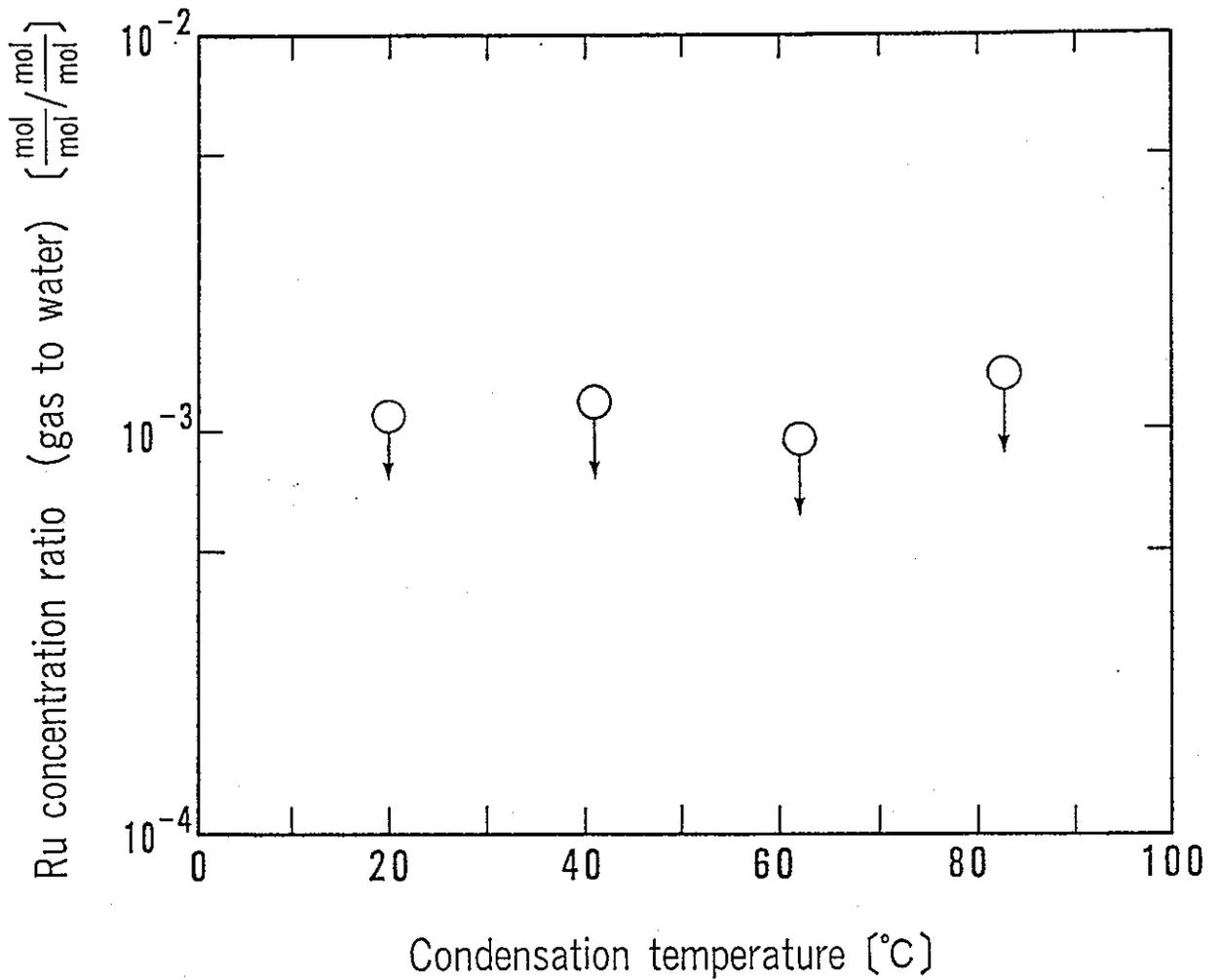
$\text{RuO}_4 + \text{Air} + \text{NO}$ through glass pipe



RuO_4 generation
(addition of NO_2)



RuO_4 generation
(addition of NO)



Effect of condensation temperature on Ru concentration ratio (gas to water) for off gas from calcining simulated HLLW

⊕ denotes under detection limit

Conclusion

1. NO_x in off gas increased the absorption of RuO₄ into condensates

NO/Ru=22 : increase by three orders

NO₂/Ru=189 : increase by two orders

2. Increase in condensation temperature did not decrease the absorption of volatile Ru into condensates

2.5 Current Status of Dismantling Technology for Liquid
Fed Ceramic Melter

Current Status of Dismantling
Technology for Liquid Fed
Ceramic Melter

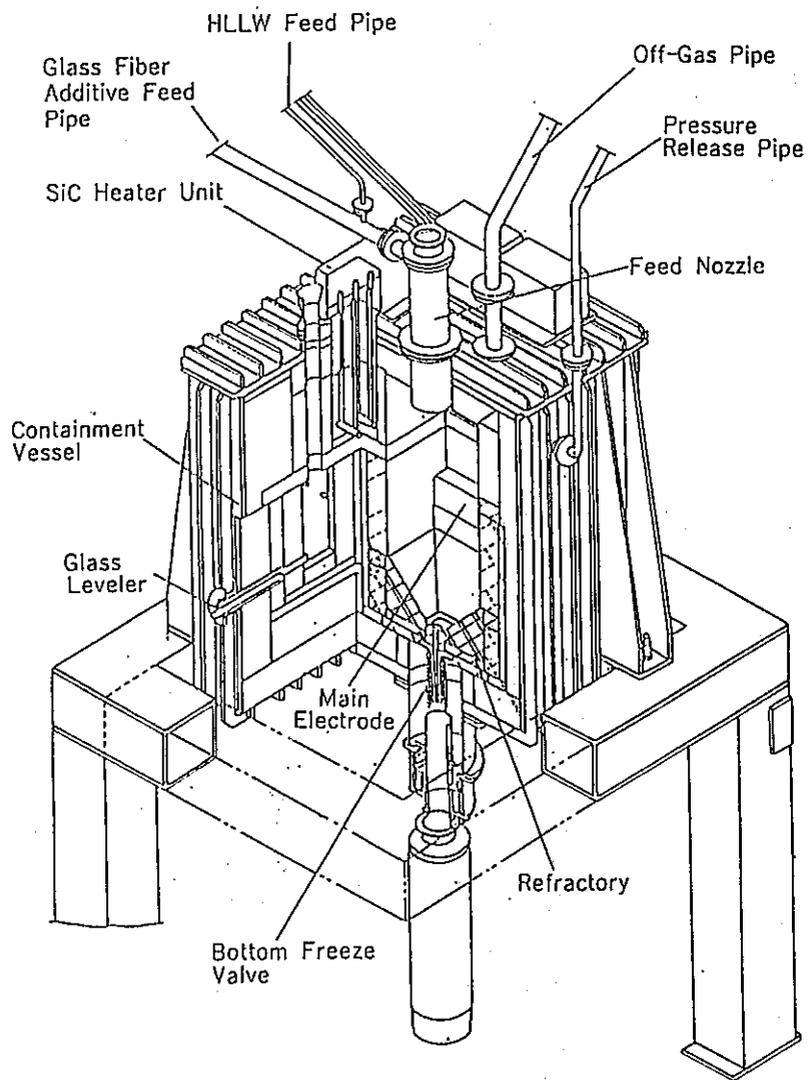
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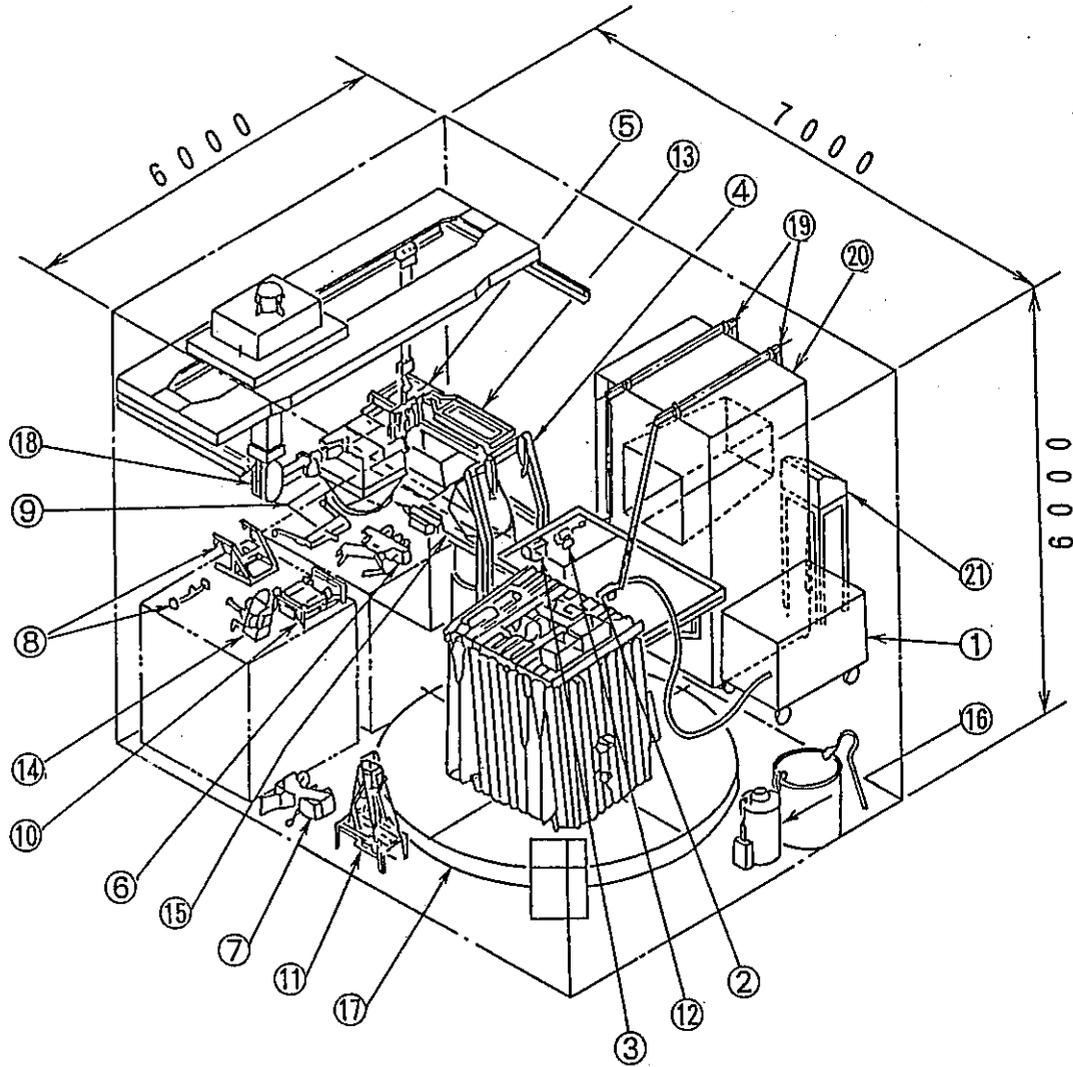
DEVELOPMENT SCHEDULE OF DISMANTLING
 THCHNOLOGY FOR CERAMIC MELTER

F. Y. Item	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994
T V F (Tokai Vitrification Facility)	<p>License Construction Cold Test Hot Operation</p>											
R & D	Basic Study	Screening of dismantling technique		Dismantling test using full scale melter				C & R •Modification of tools •Verification test & Evaluation		Cold test • Full scale		



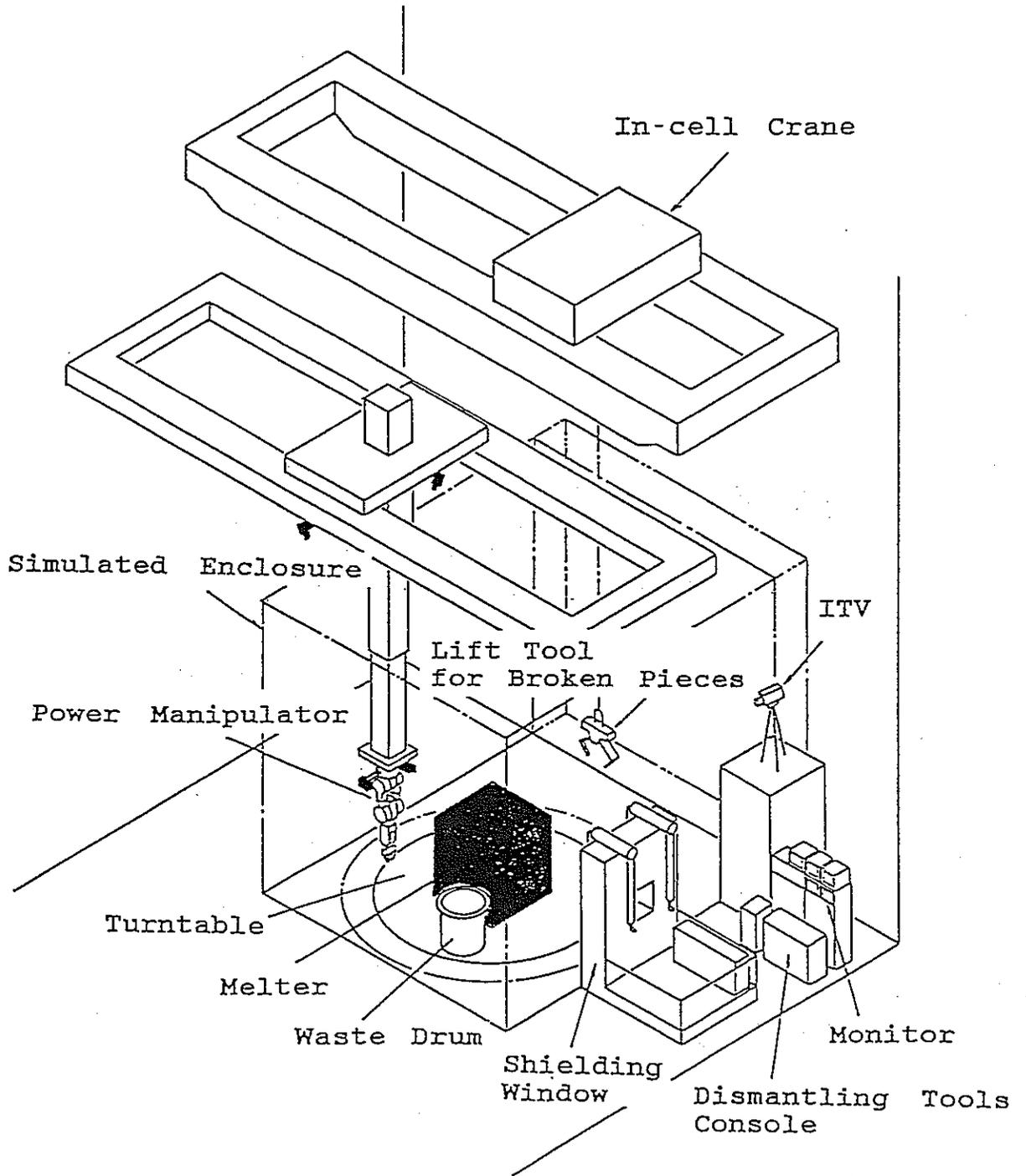
SPCIFICATION OF MELTER (TVF)

Heating	Joule-Heating
Containment Vessel	SUS 304L
Electrode	Inconel 690
External Dimension	W1850×L1870×H2300 (mm)
Glass Contact	K-3 2060kg
Refractory	
Total Weight	15,400Kg

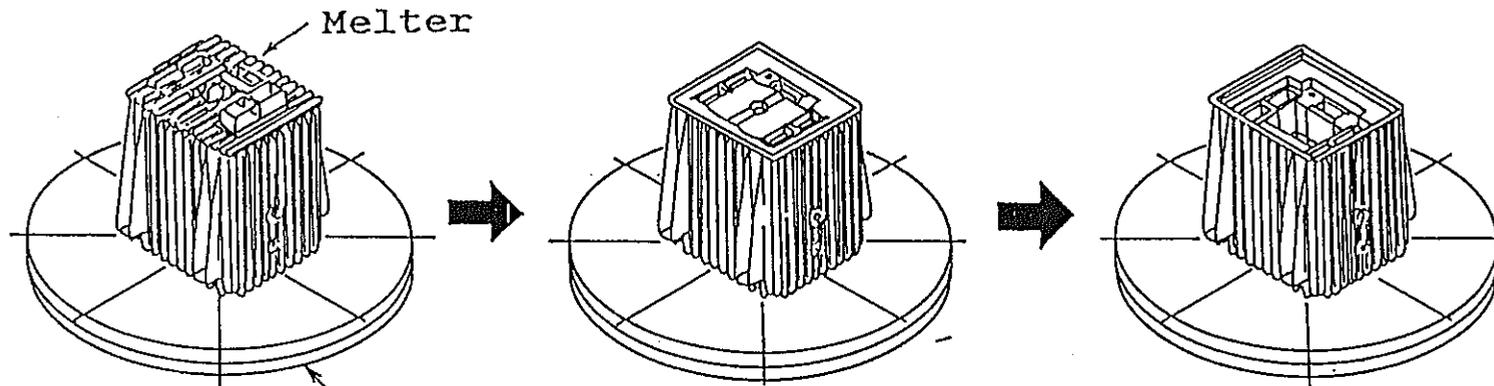


1 ~ 3	Plasma cutter
4	Hacksaw
5	Hoist scale
6 ~ 7	Handling tools of cut piece
8	Handling tools of electrode
9	Receiver
10 ~ 11	Internal dismantling breaker
12	Impact wrench
13 ~ 14	Handling tools of electrode
15	Breaker
16	Vacuum cleaner
17	Turn-table
18	Power manipulator
19	Master/slave manipulator
20	Shielding window
21	Control box

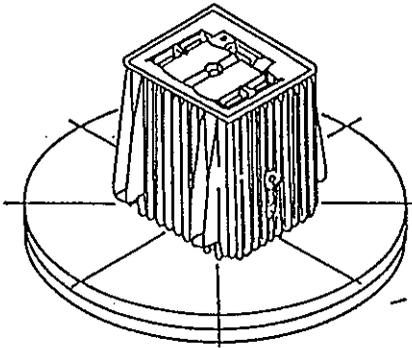
CONCEPT OF THE DISMANTLING AREA (TVF)



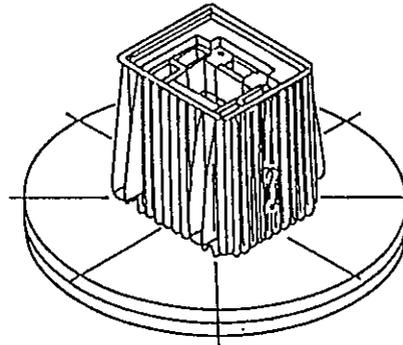
Outline of Dismantling Test Equipments



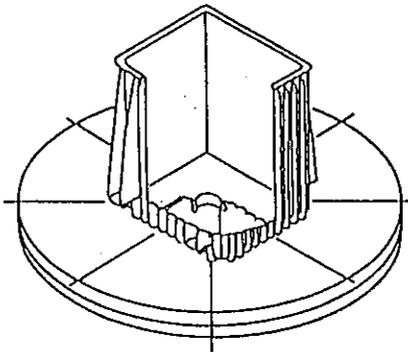
Melter
Turntable



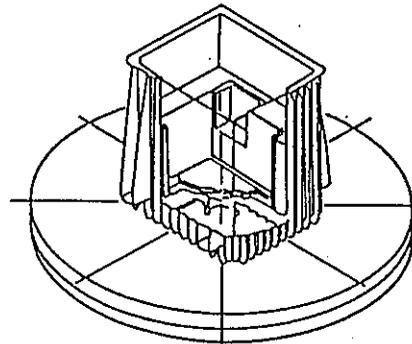
Cut of Casing at
Melter Lid



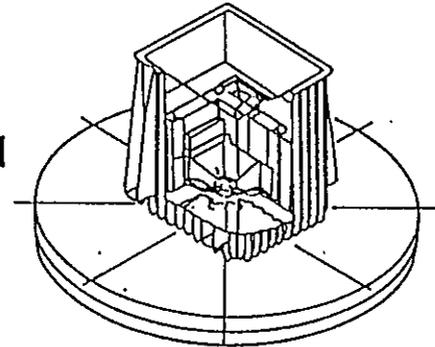
Breakdown of
Melter Lid
Refractory



Cut of Whole Casing

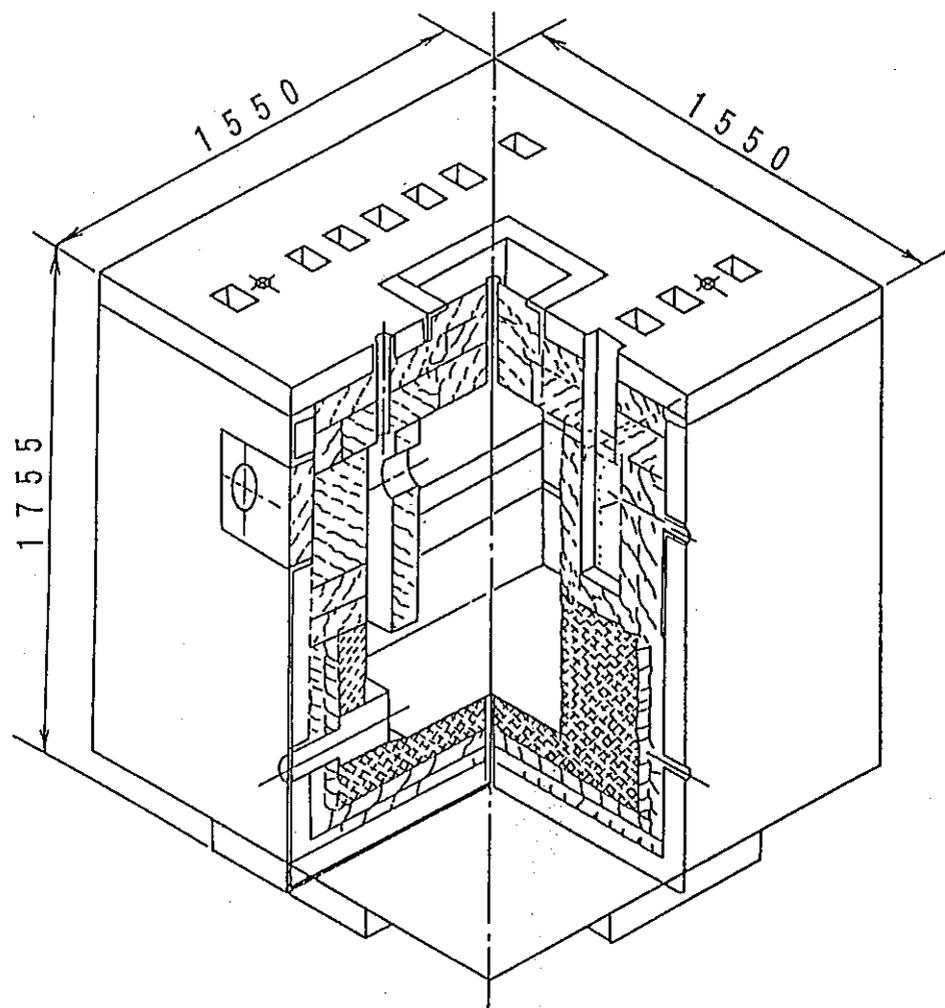


Breakdown of Glass-
contact Refractory



Breakdown of Refractory
in Superstructure

PROCEDURE OF DISMANTLING FOR MELTER

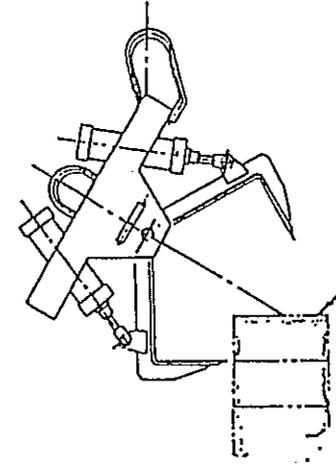
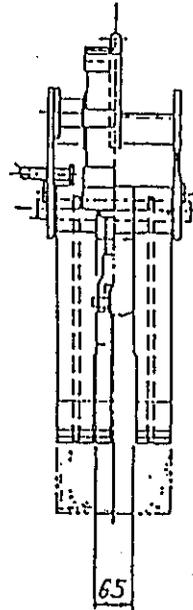
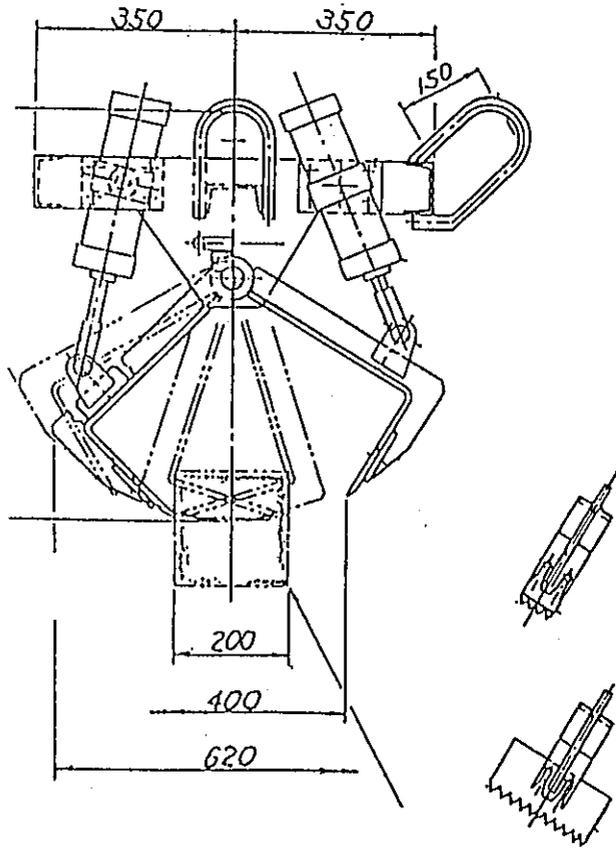


Heating	Joule-Heating
Containment Vessel	SUS 304
Electrode	Inconel 690
External Dimension	W1550 x L 1550 x H 1755 (mm)
Glass Contact	CS-5 1970kg
Refractory	K-3 230kg
Total Weight	11,000kg

SPECIFICATION OF M/U-1 MELTER

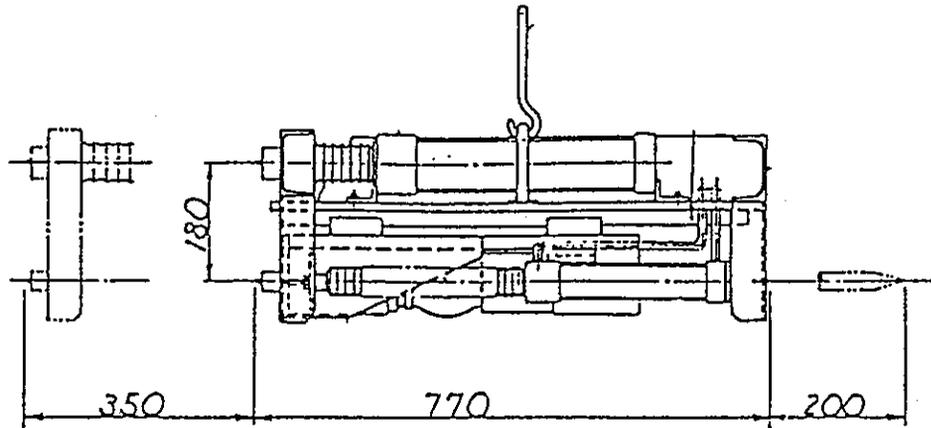
SUMMARY OF DISMANTLING METHOD

Object of Dismantling	Dismantling Method	Tools
Containment vessel	Plasma cut	Plasma cutting machine
Electrode	Cut	Hacksaw
Refractory	Direct grapple	Handling tool of cut piece
	Blow	Breaker



Driving force
: Compressed air
(6 kg/cm)
Handling capacity
: 2000 kg
weight : 60 kg

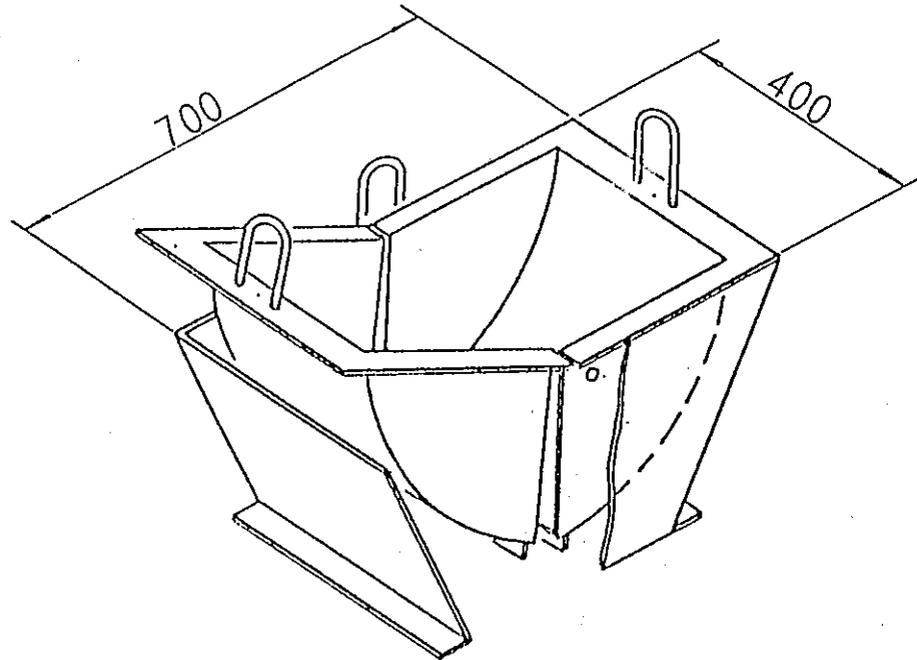
Handling tools
of cut piece



Driving force : Compressed air

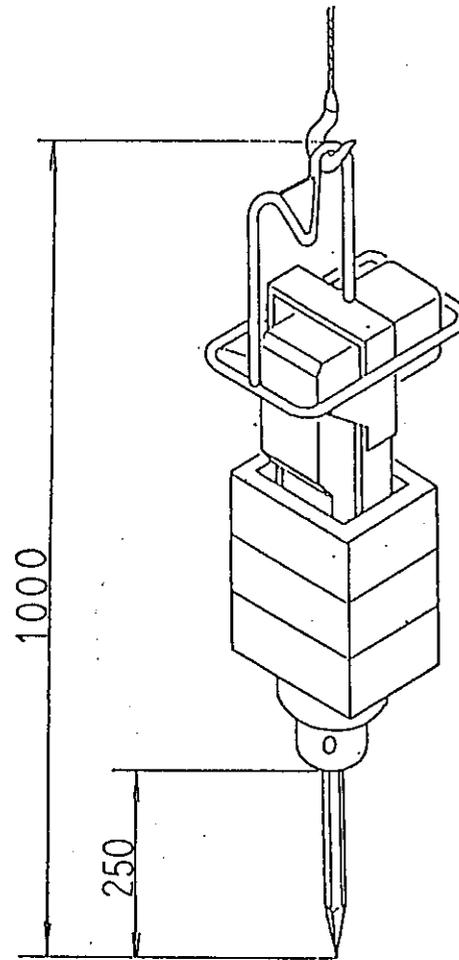
weight : 250kg

Internal dismantling breaker



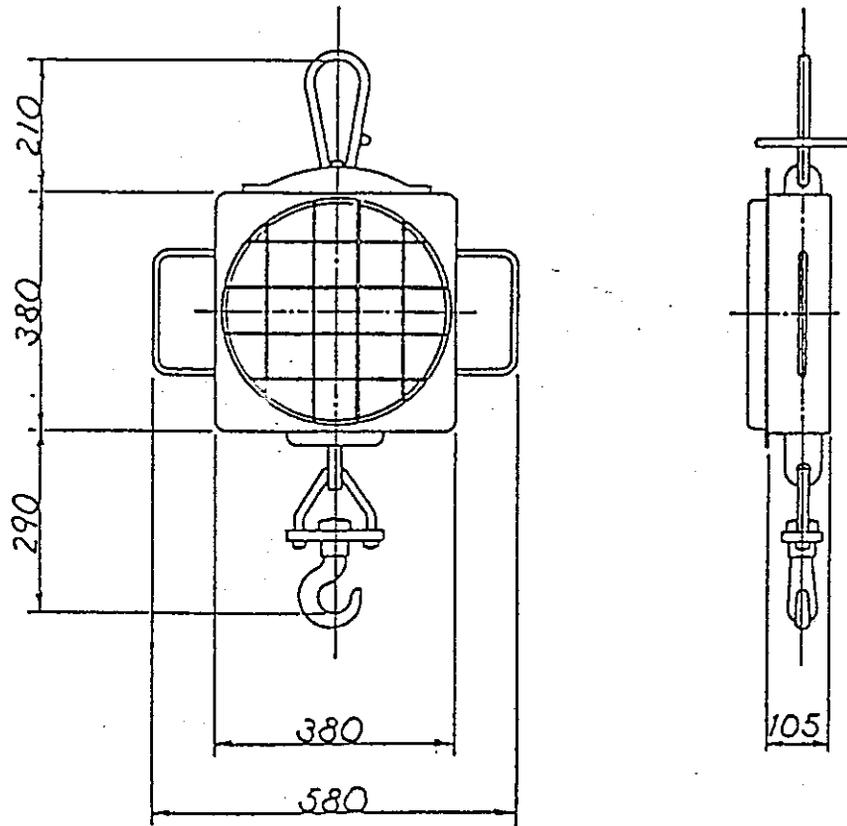
weight : 50kg

Receiver



weight
: 70kg
Driving force
: Compressed air

Breaker

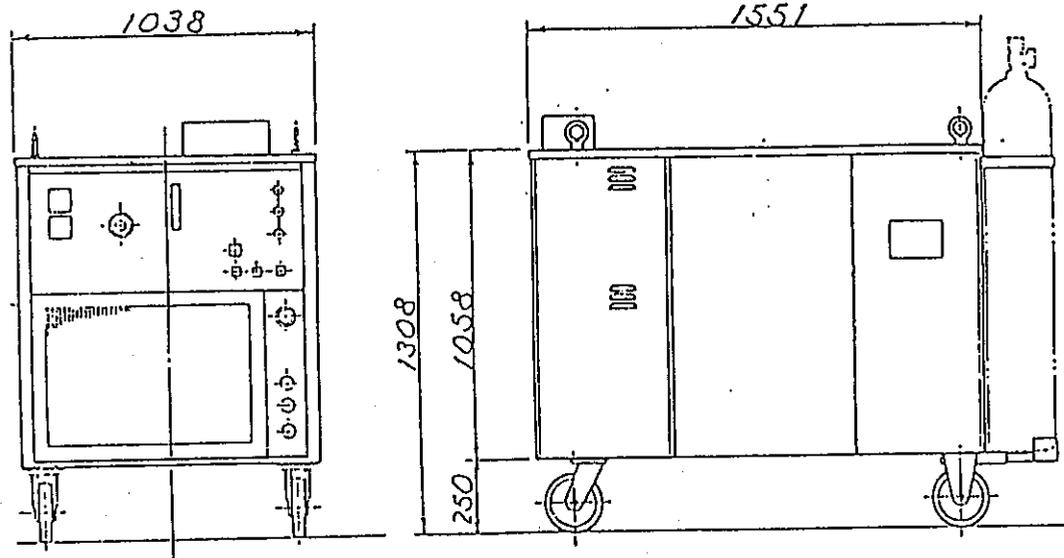


Weighing capacity: 2000kg

Minimum scale : 10kg

weight : 15kg

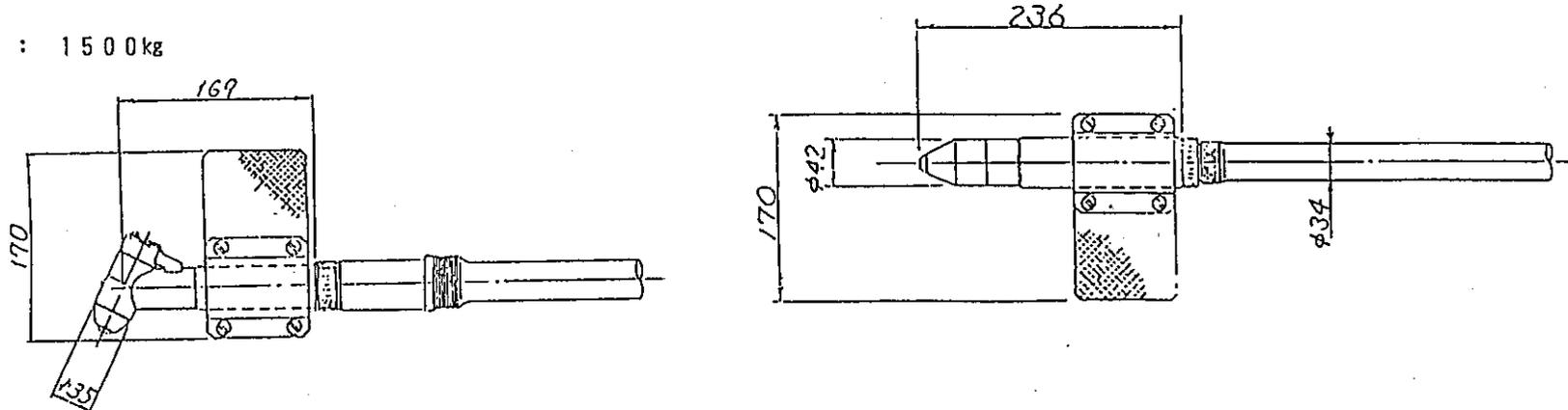
Hoist scale



Rated current
: 250A

Plasma gas: Ar gas
weight

: 1500kg



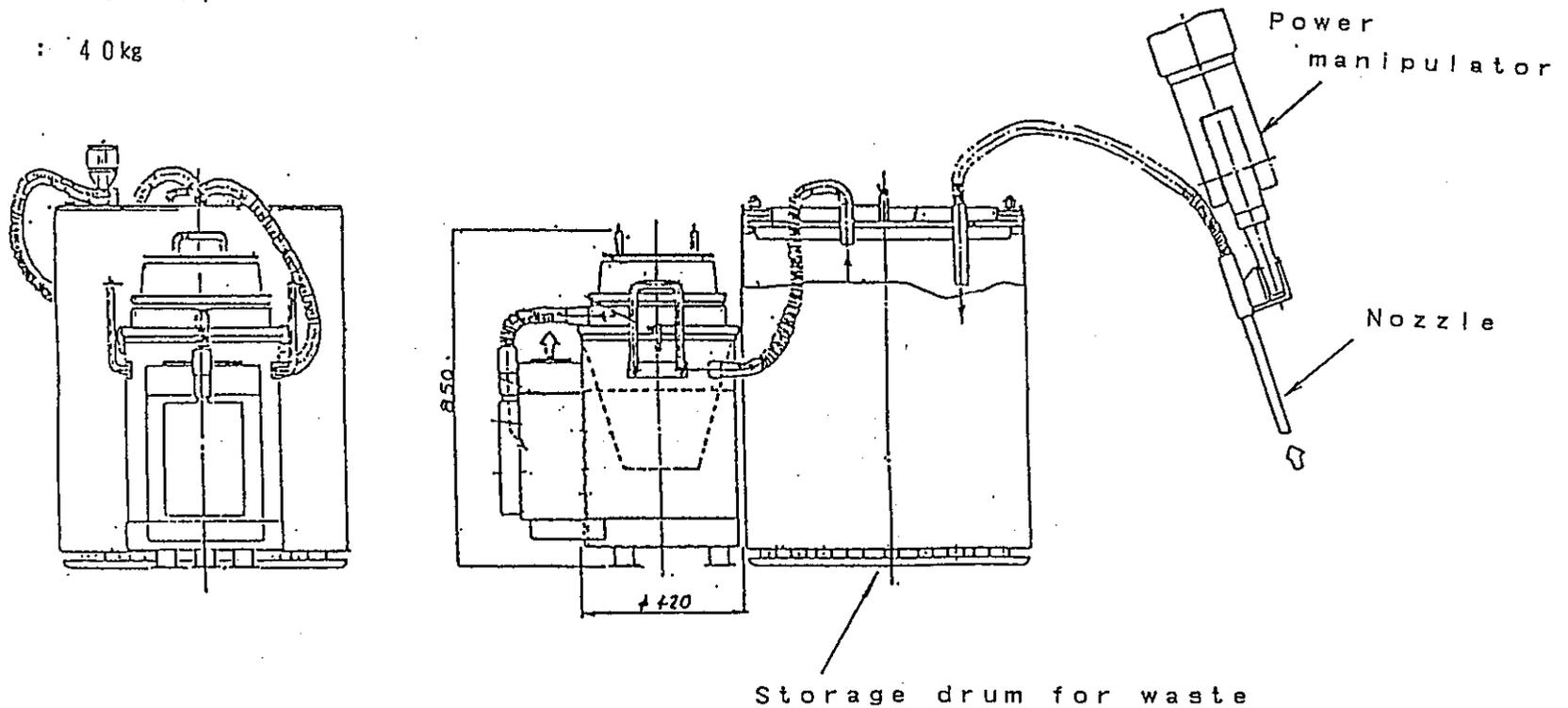
Cutting machine and Plasma torch

Airflow : 9 m³/min

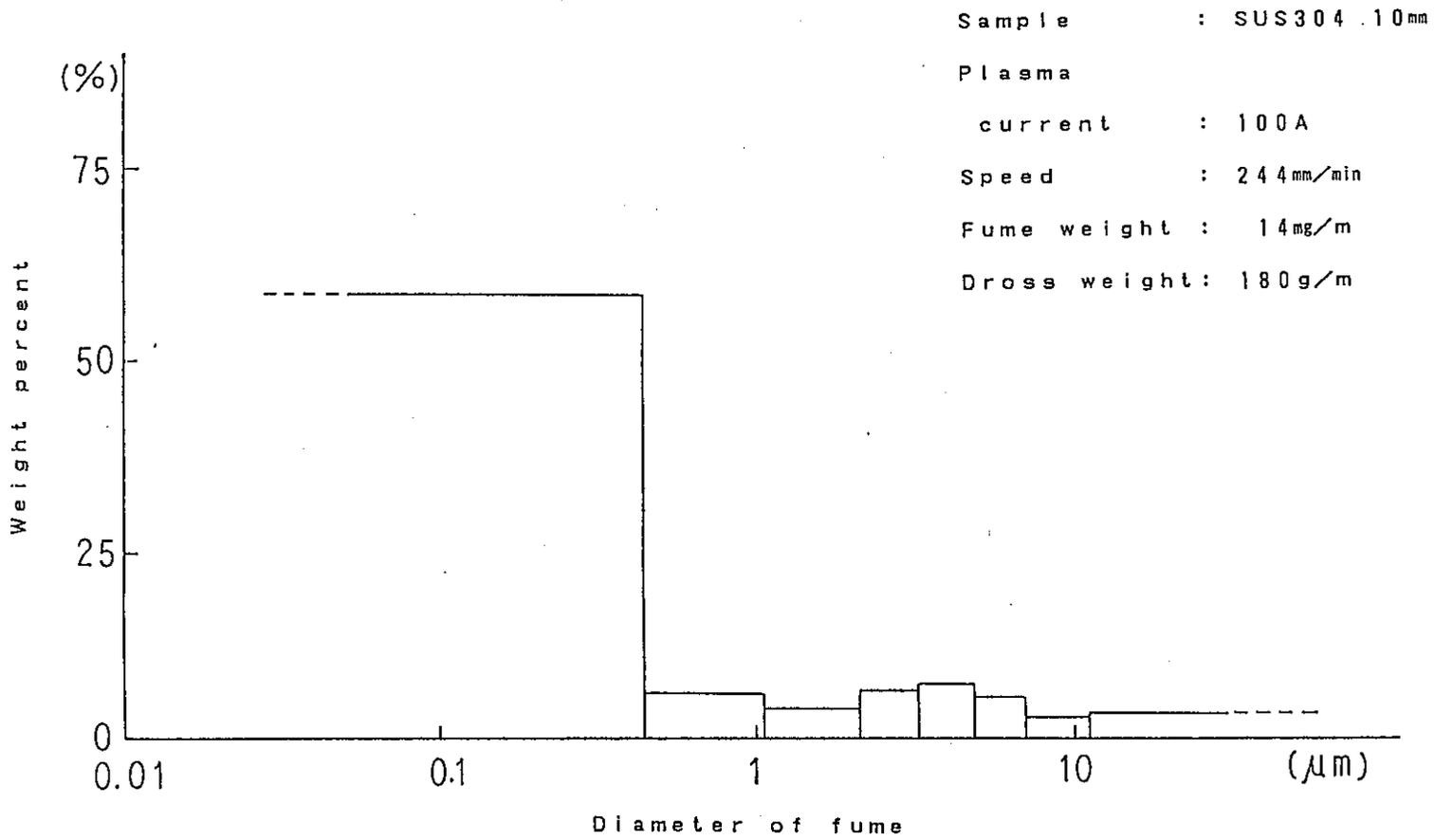
Static pressure

: -2800 mmAq

weight : 40kg



Vacuum cleaner



DISTRIBUTION OF DIAMETER FOR METAL FUME

ESTIMATED WASTE VOL. OF
DISMANTLED MELTER

	Refractory		Metal	
	K-3	Other refractory	Electrode (Incone1690)	Vessel (SUS304)
Total vol. (ℓ)	8 0 0	3 0 0 0	9 0	5 7 0
Total weight (kg)	3 0 0 0	5 0 0 0	7 6 0	4 5 0 0
Number of drum (3 4 0 ℓ) (Max 6 0 0 kg)	6	1 8	9	

ESTIMATED WORK TIME OF
DISMANTLING TEST

Name of Refractory	Total vol. (m ³)	Dismantling time (min)	Handling time (min)	Total (min)
L N - 1 3 5	2. 2 8	1 1 8 6	1 5 5 0	2 7 3 6
C - 1	0. 5 7	4 3 3	4 0 5	8 3 8
D C N	0. 0 8	1 8 1	7 1	2 5 2
Fiber board	0. 4 7	3 5 7	3 3 4	6 9 1
A Z G	0. 3 3	3 5 0	1 9 1	5 4 1
M R T - 7 0 K	0. 4 7	5 0 8	4 6 1	9 6 9
K - 3	0. 7 8	2 5 9 7	1 8 4 1	4 4 3 8
Total amount				1 7 4 H

Dismantling Test of Radioactive Spent Ceramic Melter in CPF

CPF:Chemical Processing Facility

Procedures	1988					1989			
	Aug.	Sep.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	
1. Cold mock-up test	[Bar]								
2. Install dismantling equipments	Turn table, Braker, Dust filter, Tools								
3. Dismantling of evaporator and mixing tank		[Bar]							
4. Dismantling of ceramic melter		Cut by plasma							
		Piping, Uppercasing, Refractory, Lower casing							
5. Take out dismantling wastes from cell		2 cans [Bar]	2 cans [Bar]	4 cans [Bar]	2 cans [Bar]				
6. Install new melter (The 3rd melter)					[Bar]				
7. Performance test of new melter							Test [Bar]	Operation [Dashed Bar]	
8. Inspection by authority						[Bar]			