

## 第12回 PNC /KfK 高レベル廃棄物管理会議 (2 / 2) 別冊資料集

(1993年12月7日～9日，動燃東海事業所にて開催)

### —高放射性廃液固化研究報告—

1994年2月

動力炉・核燃料開発事業団  
東 海 事 業 所

複製又はこの資料の入手については、下記にお問い合わせ下さい。

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東海事業所 技術開発推進部・技術管理室

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動力炉・核燃料開発事業団 (Power Reactor and Nuclear Fuel Development Corporation)

1 9 9 4 年 2 月

第12回 PNC/KfK高レベル廃棄物管理会議 (2/2) -別冊資料集  
(1993年12月7日～9日, 動燃東海事業所にて開催)

報告責任者 大内 仁  
編集者 捧 賢一, 二村 浩尔

要 旨

動燃事業団と独KfKとの間に結ばれている高レベル廃棄物管理における協定の一環として, ガラス固化技術を中心とする廃棄物管理に関する会議が1993年12月7日から9日までの3日間, 東海事業所において開催された。

本資料は, 第12回PNC/KfK高レベル廃棄物管理会議報告書 (1/2) -会議 PNC PN8100 94-002の別冊資料集として, 発表に用いたOHP資料をとりまとめたものである。

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報告者所属 : 環境技術開発部 環境技術第一開発室

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1. 会議日程

1993.12.7

Agenda of  
The 12th Annual meeting between KfK and PNC  
in the field of High-Level Waste Management  
held at PNC Tokai on 7-9 December

**The 1st day, Dec.7, Tuesday**

8:00 Pick-up at the Hotel

8:25 Arrive at the Tokai-works and clearance procedure at the gate

8:40 Welcome and meeting Mr.Miyahara, the Director and Mr.Sasao, the Deputy director of Tokai works

Move to ETF

9:00 Opening remarks and Confirmation of agenda

**TECHNICAL PRESENTATION(1)**

**OVERVIEW**

9:10 Current status of radioactive waste management in JAPAN

(PNC: Zaitu,T.)

9:30 Current status of radioactive waste management in GERMANY

(KfK: Roth,G.)

**VITRIFICATION PLANT AND PROCESS PERFORMANCE**

9:50 Conceptual study for the conditioning of the HAWC-WAK waste and on-site vitrification

(KfK: Roth,G.)

10:10 Plant description and nonradioactive start-up test of TVF

(PNC: Yoshioka,H.)

11:00 Evaluation of the noble metals behavior, ESM feed preparation system

(KfK: Grünwald,W.)

**MELTER AND PROCESS TECHNOLOGY**

11:30 GRAVIS process control and data acquisition system (KfK: Roth,G.)

12:00 Lunch

13:00 Tour of TVF

**TECHNICAL PRESENTATION(Continued)**

15:00 3-dimensional mathematical modeling and physical modeling of melter

(PNC: Ayame,Y.)

15:30 Measurements of DF data for Sr, Cs and Ru for the melter and the off-gas line

(KfK: Weisenburger,S.)

16:00 Inspection technique based on LASER for melter and dismantling of spent melter

(PNC: Kobayashi,H.)

16:30 Melt level detection system recently tested in the K-6' and K-W3 melter  
New overflow heating technique applied in the ESM melter

(KfK: Tobie,W.)

17:00 Move to Hotel

The 2nd day, Dec.8 Wednesday

TECHNICAL PRESENTATION(2)

9:00 FUNDAMENTAL RESEARCH ON THE FUTURE HLLW TREATMENT

- |  |                    |
|--|--------------------|
| New glass melter                         | (PNC: Igarashi,H.) |
| Partitioning of heat generating elements | (PNC: Yonezawa,S.) |
| Higher volume reduction glass melting    | (PNC: Sasage,K.)   |

WASTE CHARACTERIZATION AND PRODUCT QUALITY ASSURANCE

10:00 Characterization and quality assurance for the HAWC-WAK waste glass if produced in the PAMELA and disposed in Germany (KfK: Weisenburger,S.)

10:30 Characterization of glass and HLLW for the quality assurance(PNC)

Characterization of the glass produced in cold test operation of TVF,  
(PNC: Kawamura,T.)

Characterization of the HLLW from the Tokai Reprocessing Plant  
(PNC: Yamashita,T.)

11:00 Break

TECHNICAL DISCUSSION

11:10 Identify subjects to discuss and discussion

12:00 Lunch

13:00 Discussion

17:00 Move to hotel

The 3rd day, Dec.9 Thursday

8:40 Summarize the technical discussion

12:00 Lunch

13:00 Summarize the technical discussion

15:00 Future collaboration

Preparation of the meeting minutes

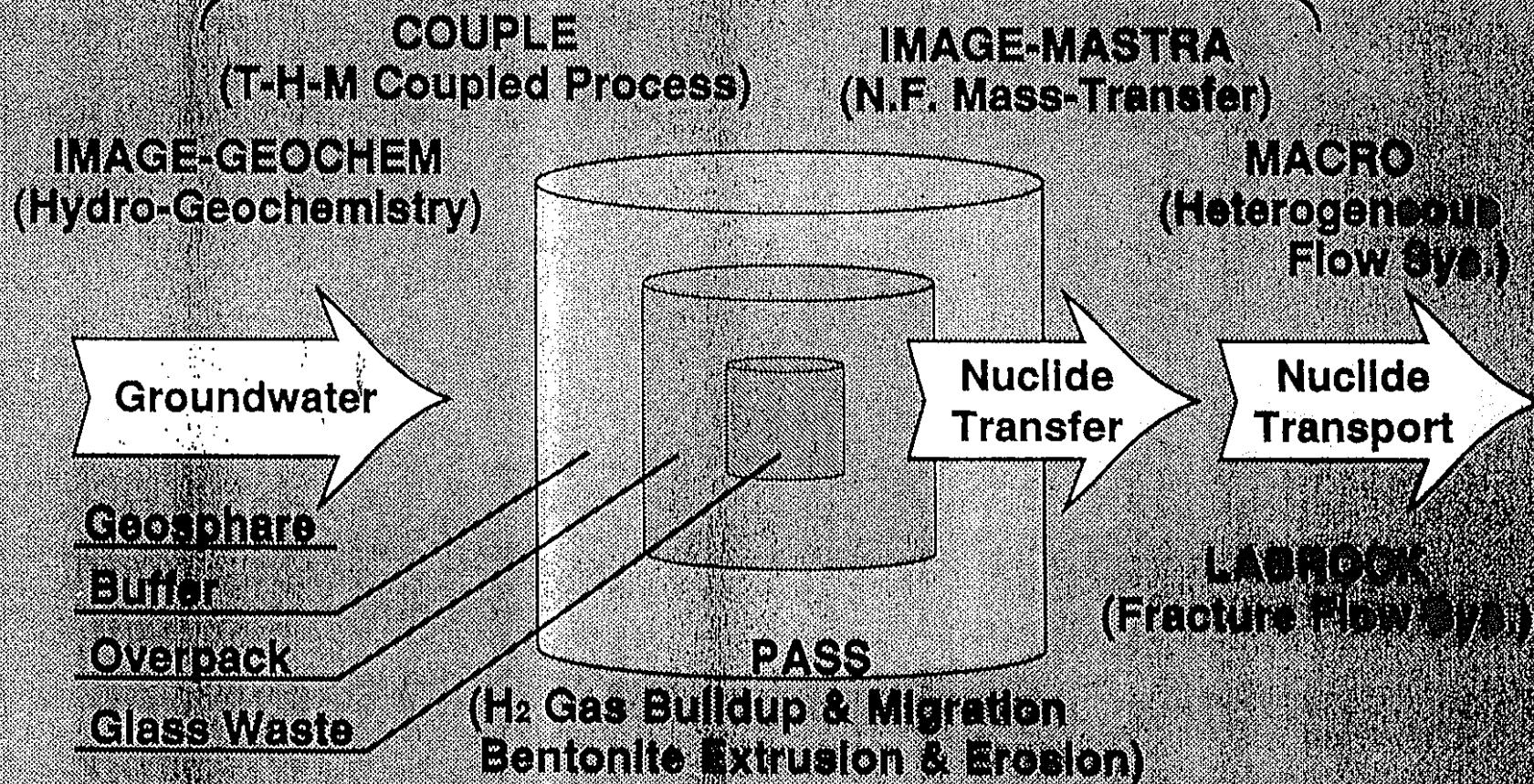
17:00 Move to Hotel

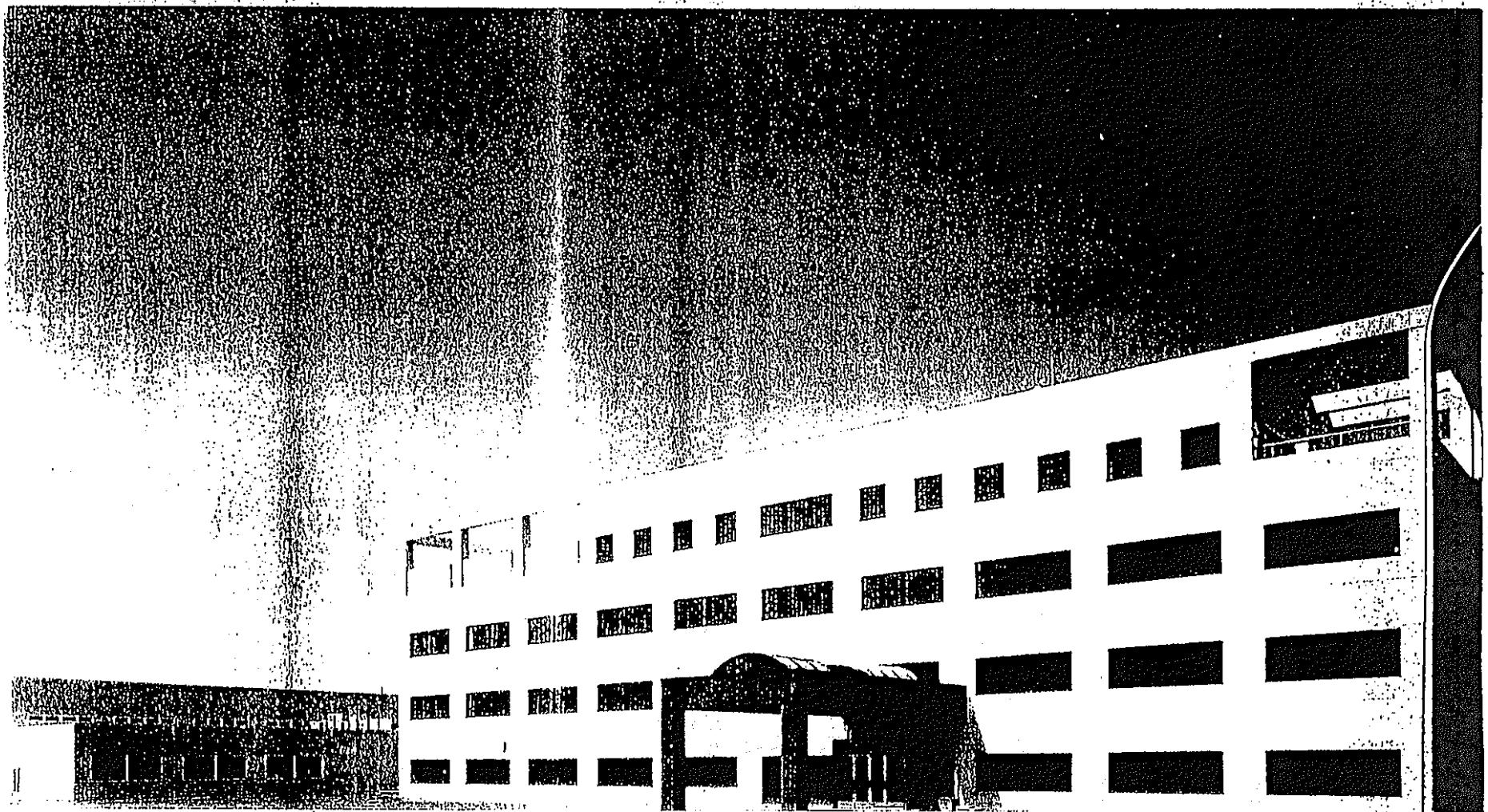
2. PNC 側の発表資料

2.1 Current status of radioactive waste management in JAPAN

# Major Tests in GIRF

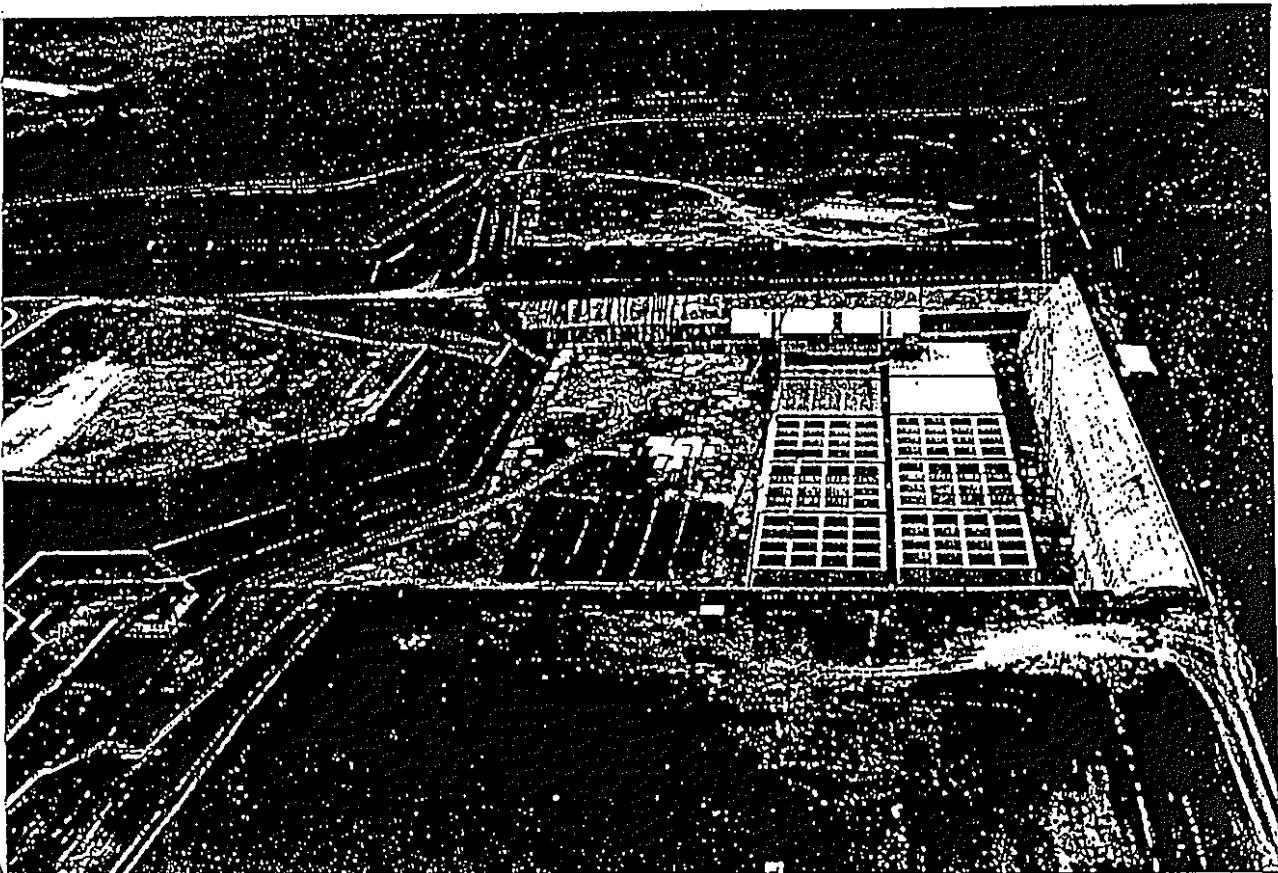
## EDAS (Kinetic & Thermodynamic Data)





Geological Isolation Research Facility (GIRF)

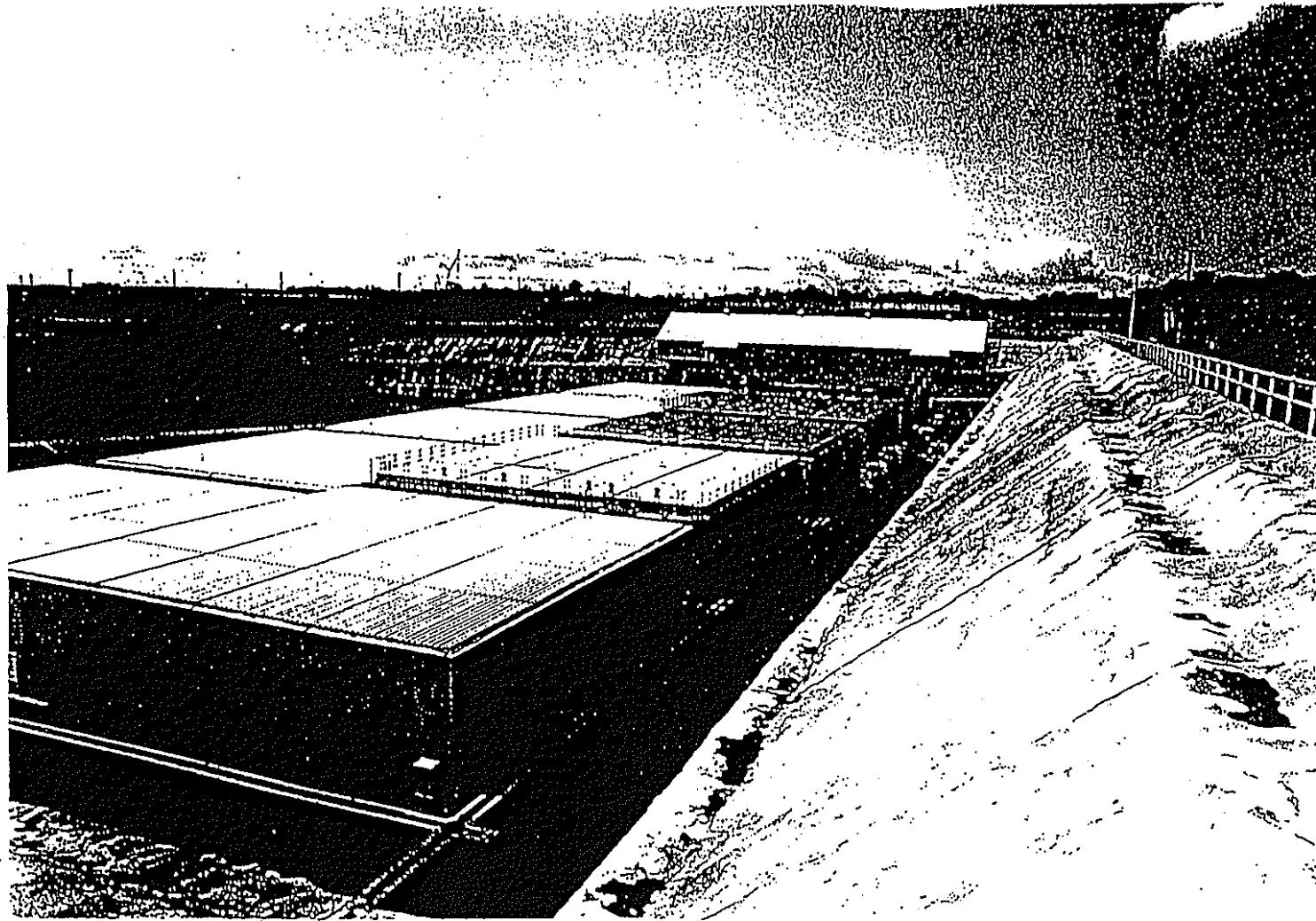
# Management of LLW



Rokkasho

Tokyo

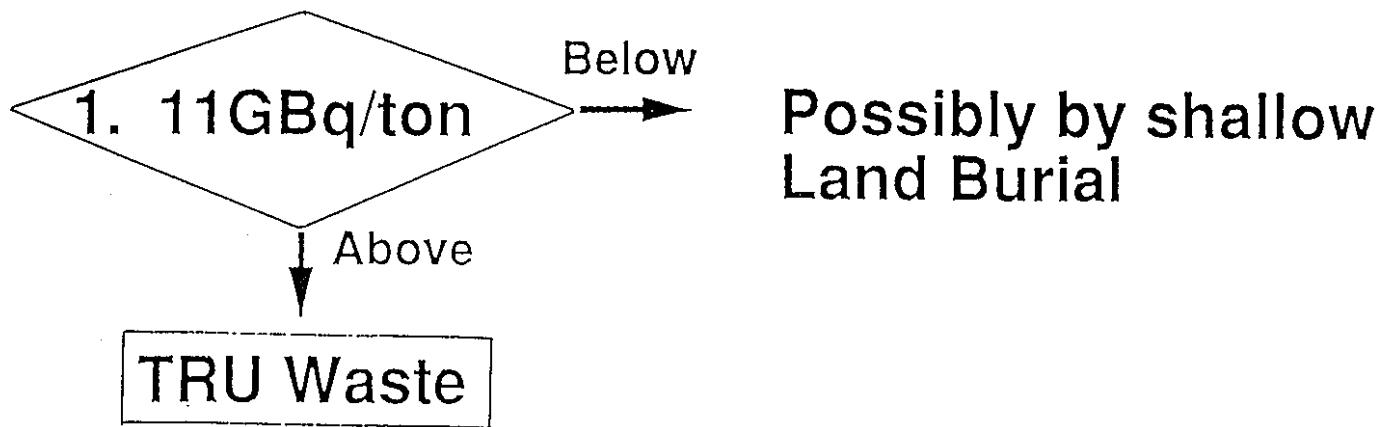
Shallow Land Burial at Rokkasho



## Shallow Land Burial at Rokkasho

## Management of Radioactive Waste Containing TRU Nuclides

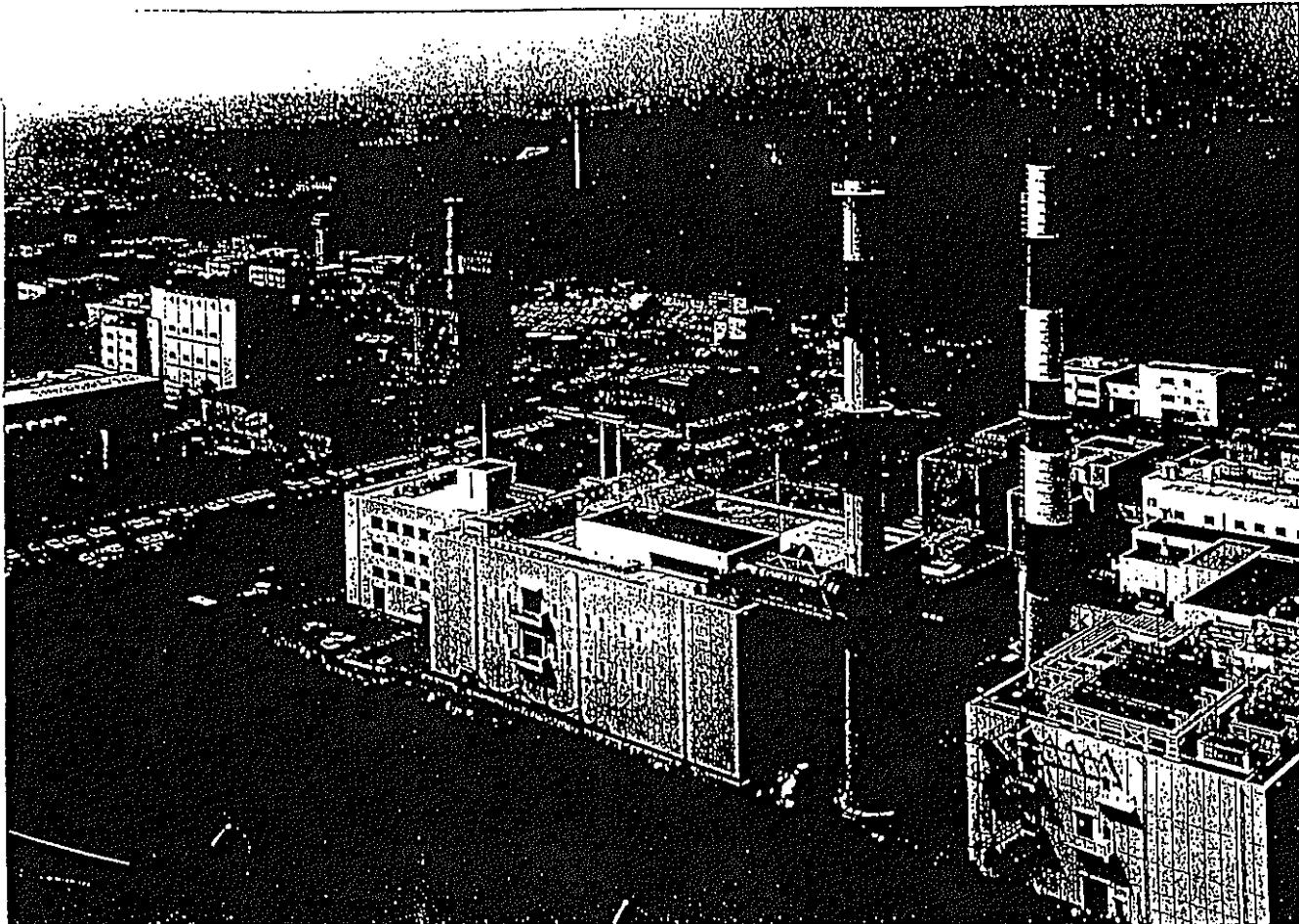
- Minimize the Generation and Develop the More Advanced Technology to Reduce the Volume
- Dispose of :



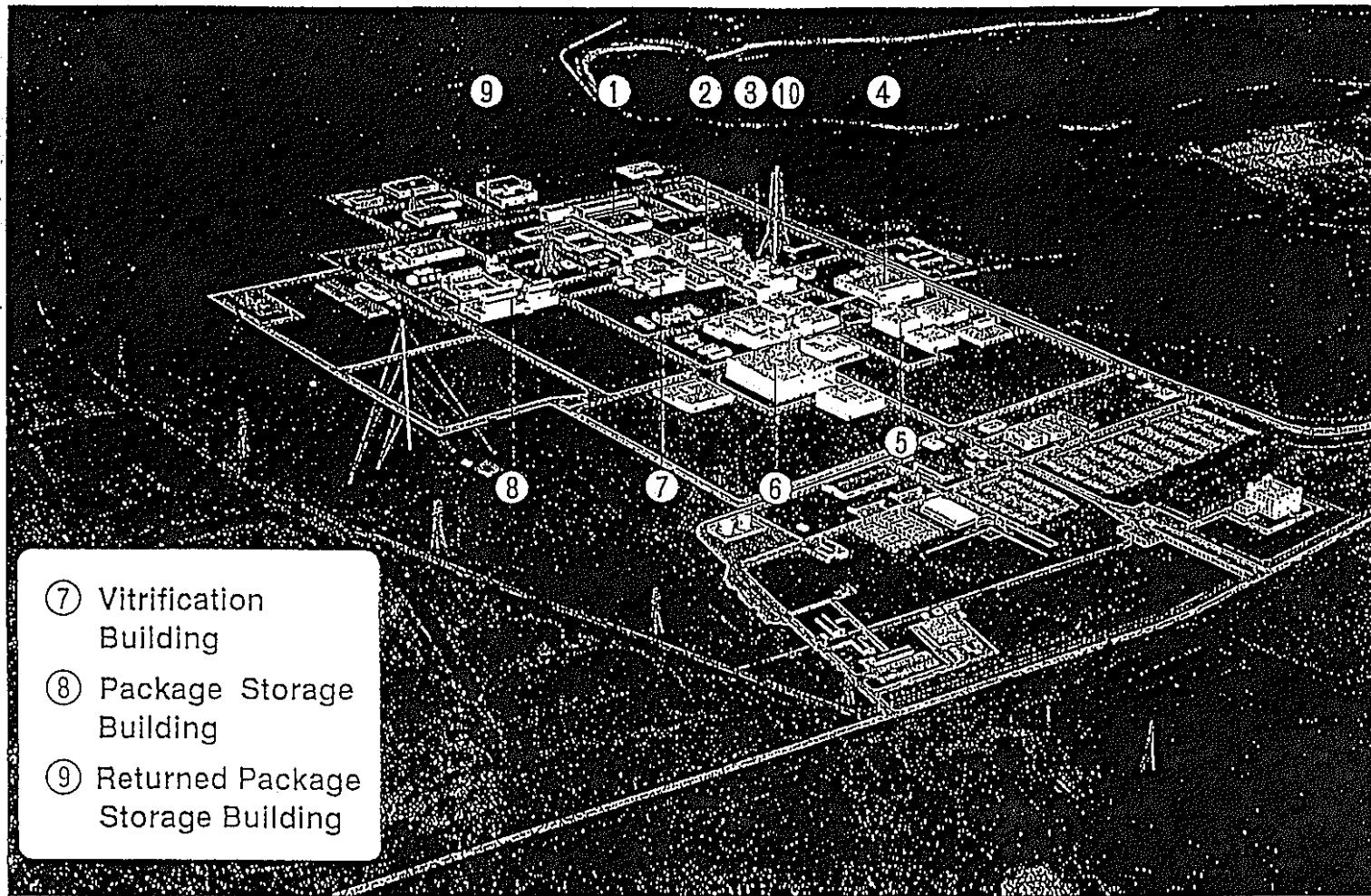
More Adequately in Underground Repository

- Clarify the Feasibility of Disposal Technology by the Late of 1990's

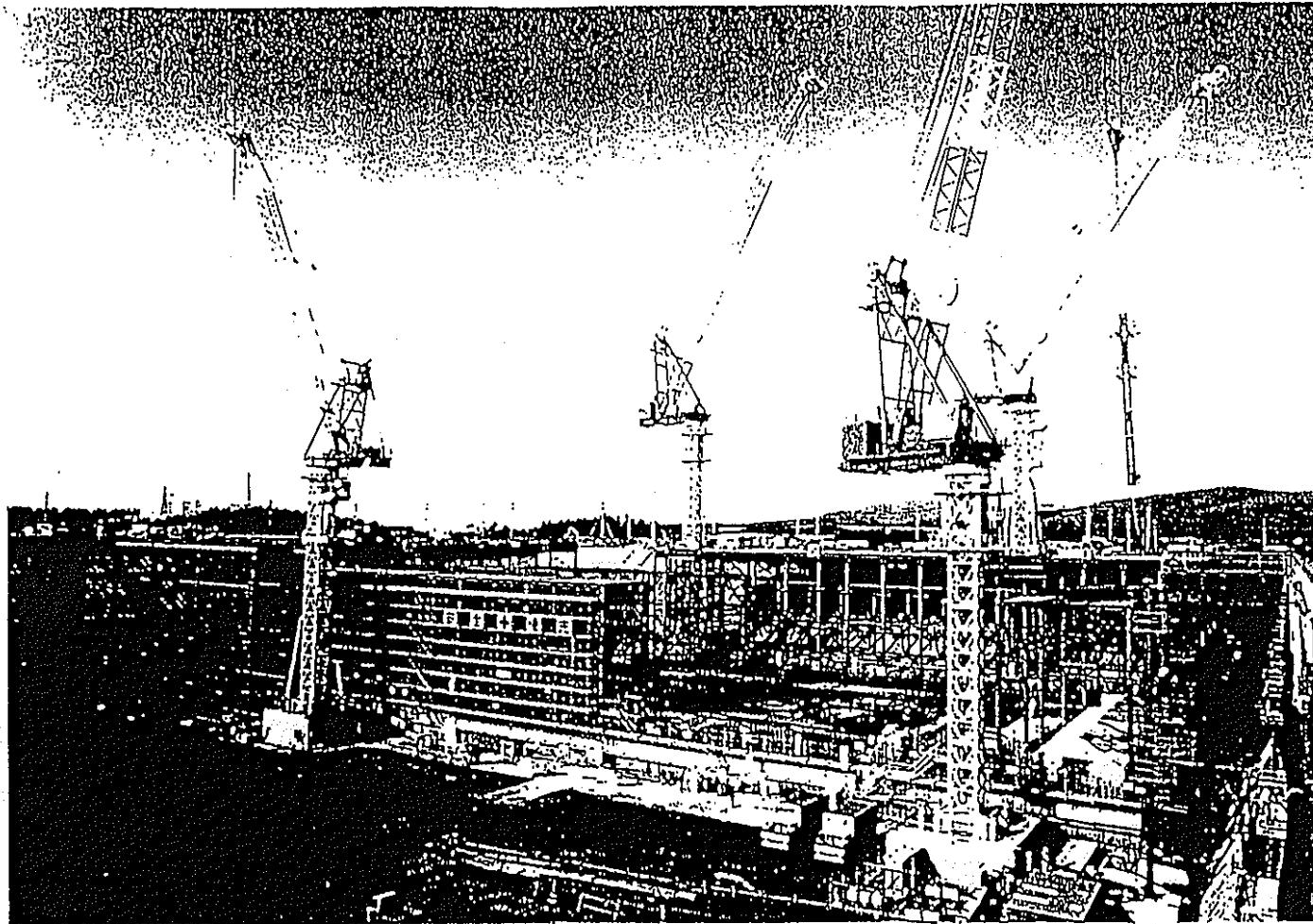
## Management of HLW



Tokai Vitrification Facility



**General Layout of the Reprocessing Plant  
at Rokkasho**



## Construction of Returned Vitrified Package Storage Building



## **HLW Disposal Program should be Based on Careful Consideration**

**It is Considered Highly Important :**

- To Provide a Clear Distinction between Implementation of Disposal and R&D as Independent Processes
- To Increase the Transparency of the Overall Disposal Program by Defining Concrete Schedules and the Roles & Responsibilities of the Organization Involved

# Overall Procedures & Schedule for Implementing Geological Disposal

1993      Steering Committee on High-level Radioactive Waste Project (SHP) was Established on 28th of May

~2000      Implementing Organization will be Established  
- Site Selection with Local Acceptance  
- Site Characterization Studies & Demonstration of Disposal Technology  
- Design of Repository and Application for Licensing

Safety Investigations for Granting License  
Conducted by the Government

2030

Mid of 2040's

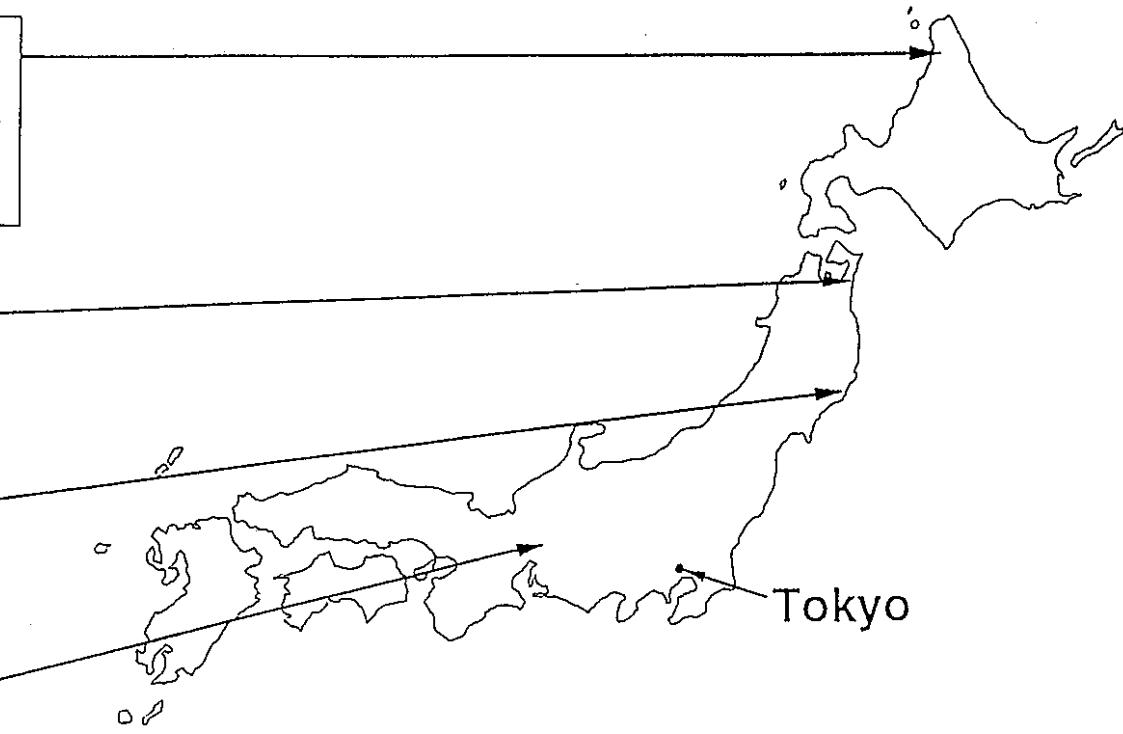
Start of Repository Operation

Horonobe  
Candidate DURF Site  
( Sedimentary Rock )

Rokkasho  
LLW Repository

Kamaishi Mine  
In-situ Study Site

Tono Mine  
In-situ Study Site



?

Candidate DURF Site  
( Crystalline Rock )

?

HLW Repository

In-situ Study Sites where PNC is Conducting Research and Development.  
( DURF means deep underground research facility. )

## **HLW Disposal Program should be Based on Careful Consideration**

- Reviews of the Results of R &D Work should be Carried out by Governmental Authorities at Appropriate Intervals
- It is Important to Ensure that Local Communities' Views and Wishes are Reflected in the Decision-making Process during Each Stage of the Program

## Roles and Responsibilities

### Government

- Overall Responsibility
- Policy and Rule Making

### PNC

- Research and Development
- Geological Environment Study

### Utilities

- Collection of Disposal Cost
- Cooperation in R&D as Waste Producers

## **PNC's First Progress Report on Geological Disposal of HLW**

- Submitted to the Authority in September 1992
- Provided Information for the Public to Find out the Current Status of R&D
- Reviewed and Evaluated by Authority in July 1993
- The Second Progress Report is Scheduled to be Submitted before the Year 2000



## In-situ Experiments at Kamaishi Mine

## 2.2 Cold testing status of Tokai Vitrification Facility

**COLD TESTING STATUS  
OF  
TOKAI DITRIFICATION FACILITY**

**M. YOSHIOKA**

**The 12th Annual KfK/PNC Meeting  
on  
High-level Waste Management**

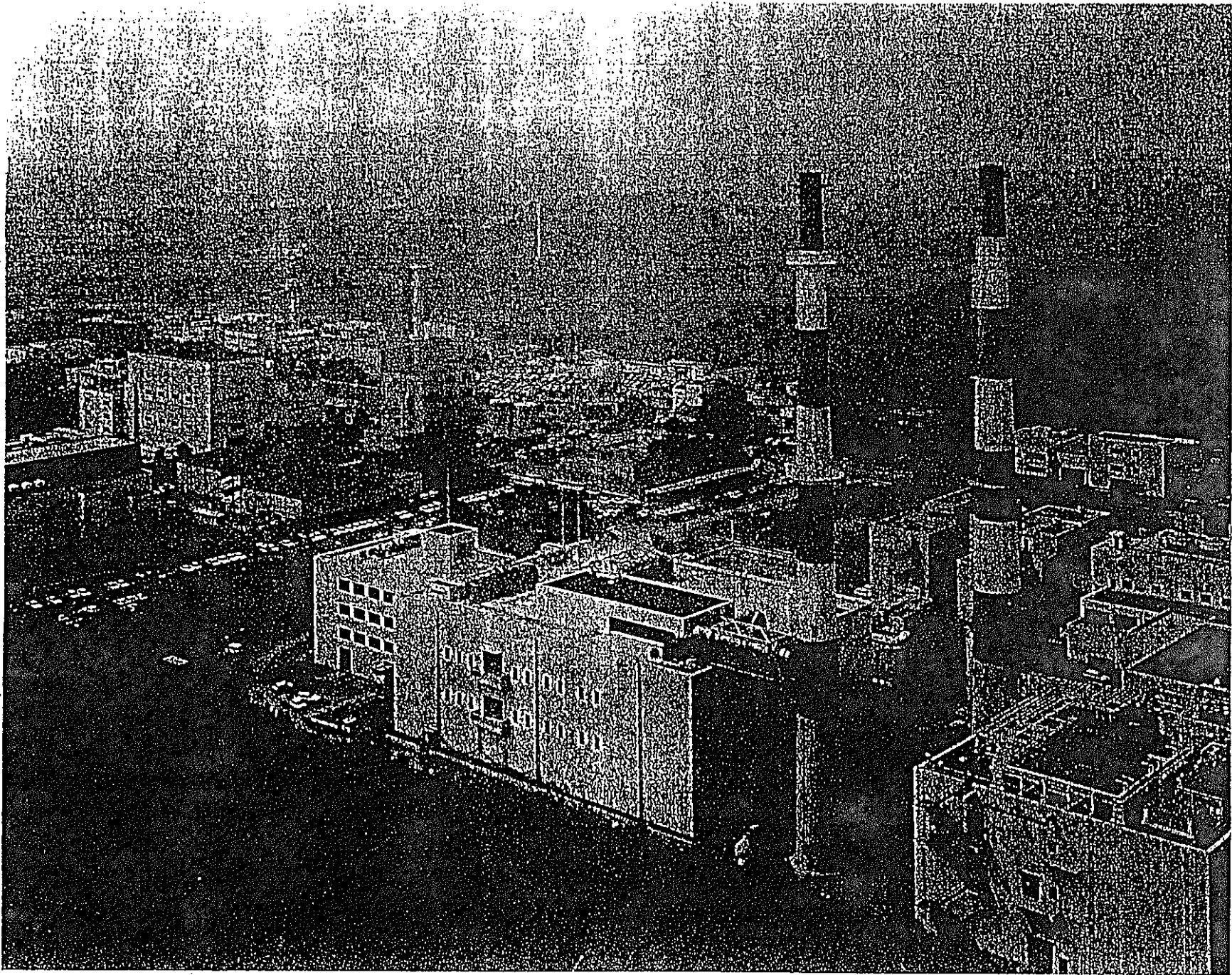
**December 7-9, 1993**

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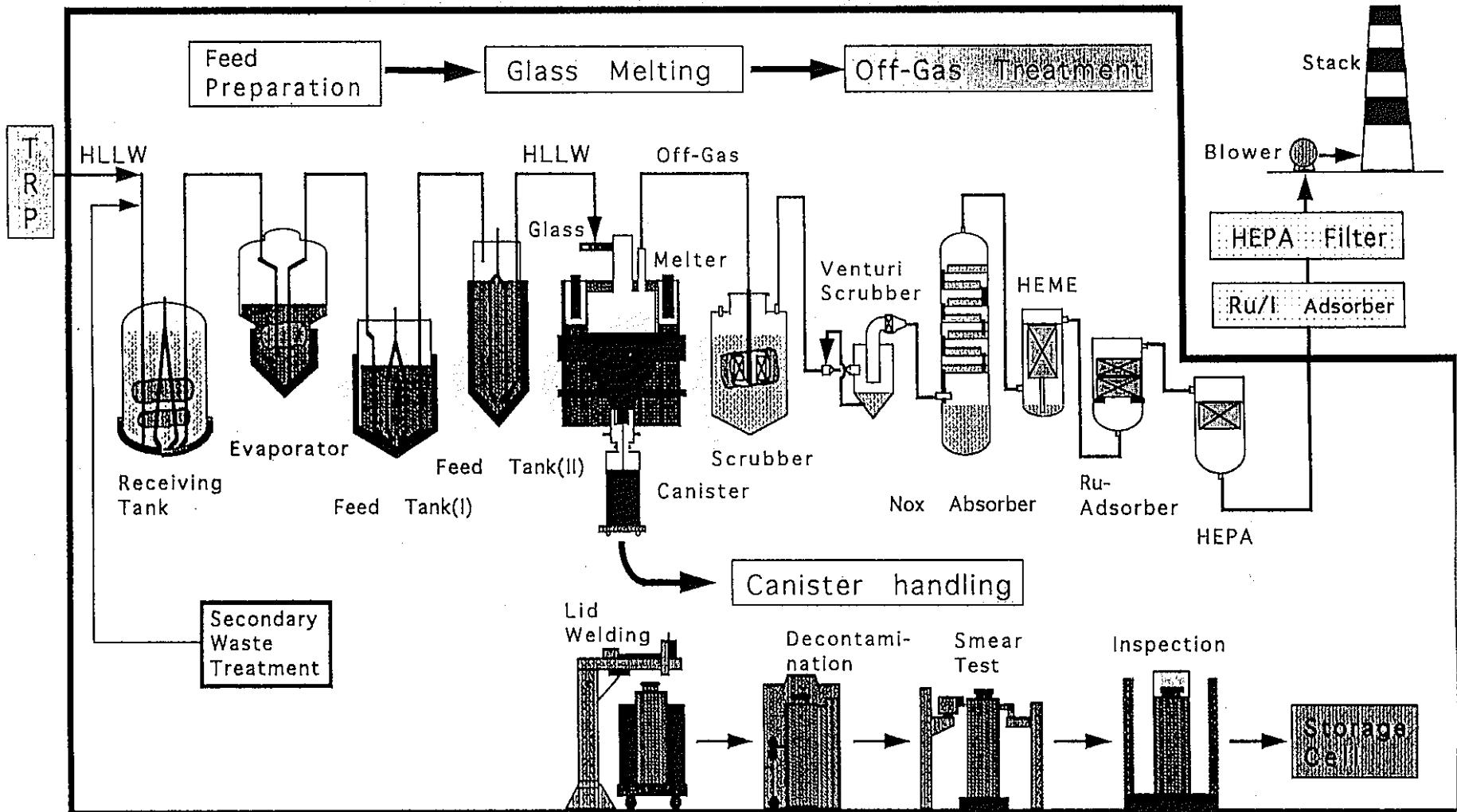
**Power Reactor and Nuclear Fuel  
Development Corporation**

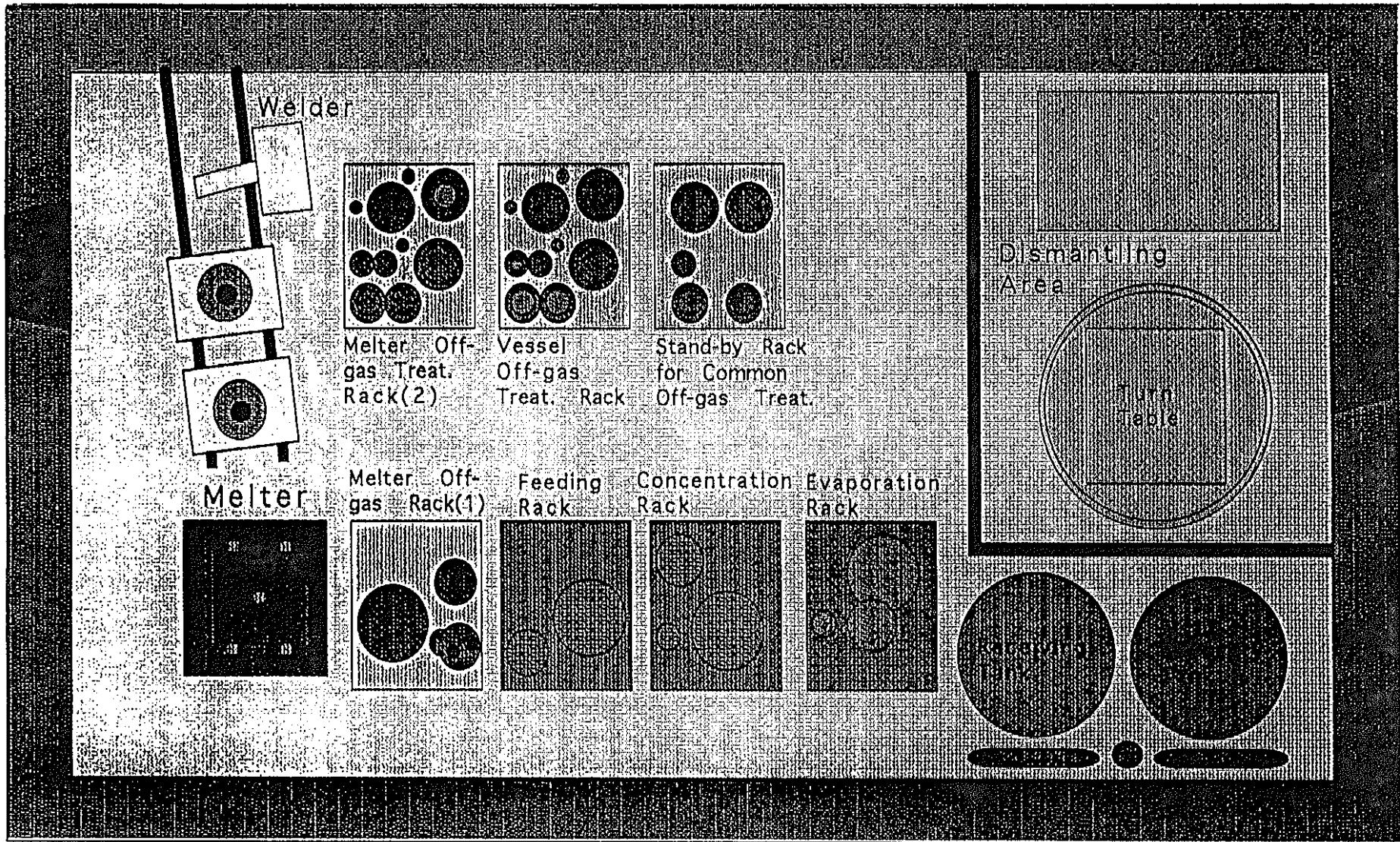


- OUTLINE OF TVF
  - Vitrification Process in TVF
  - Status of Cold Test Operation
- PROCESS CONTROL FOR GLASS PRODUCT
- MELTER OPERATION RESULT
- PERFORMANCE OF OFF-GAS TREATMENT PROCESS



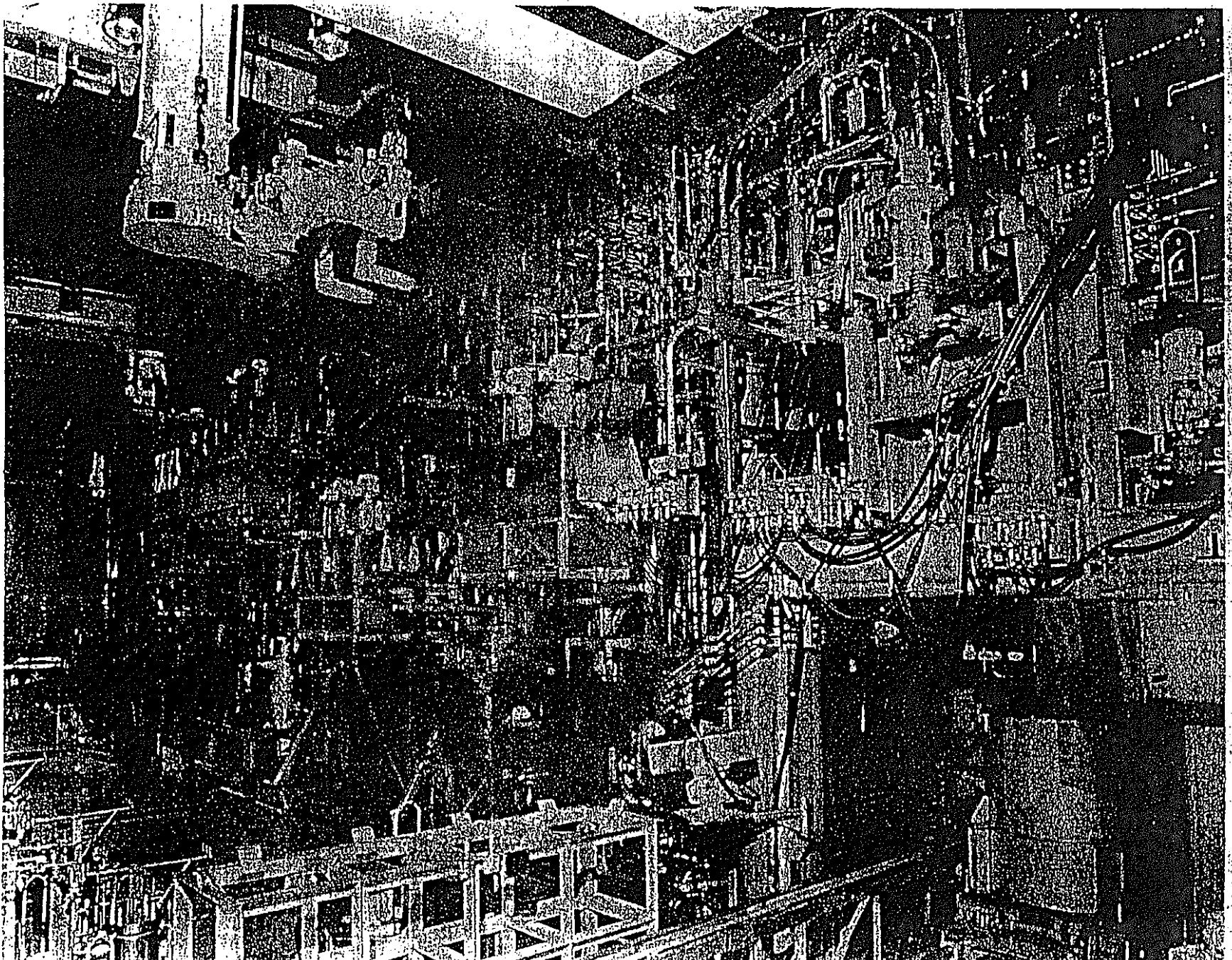
## MAIN PROCESS FLOW IN TVF

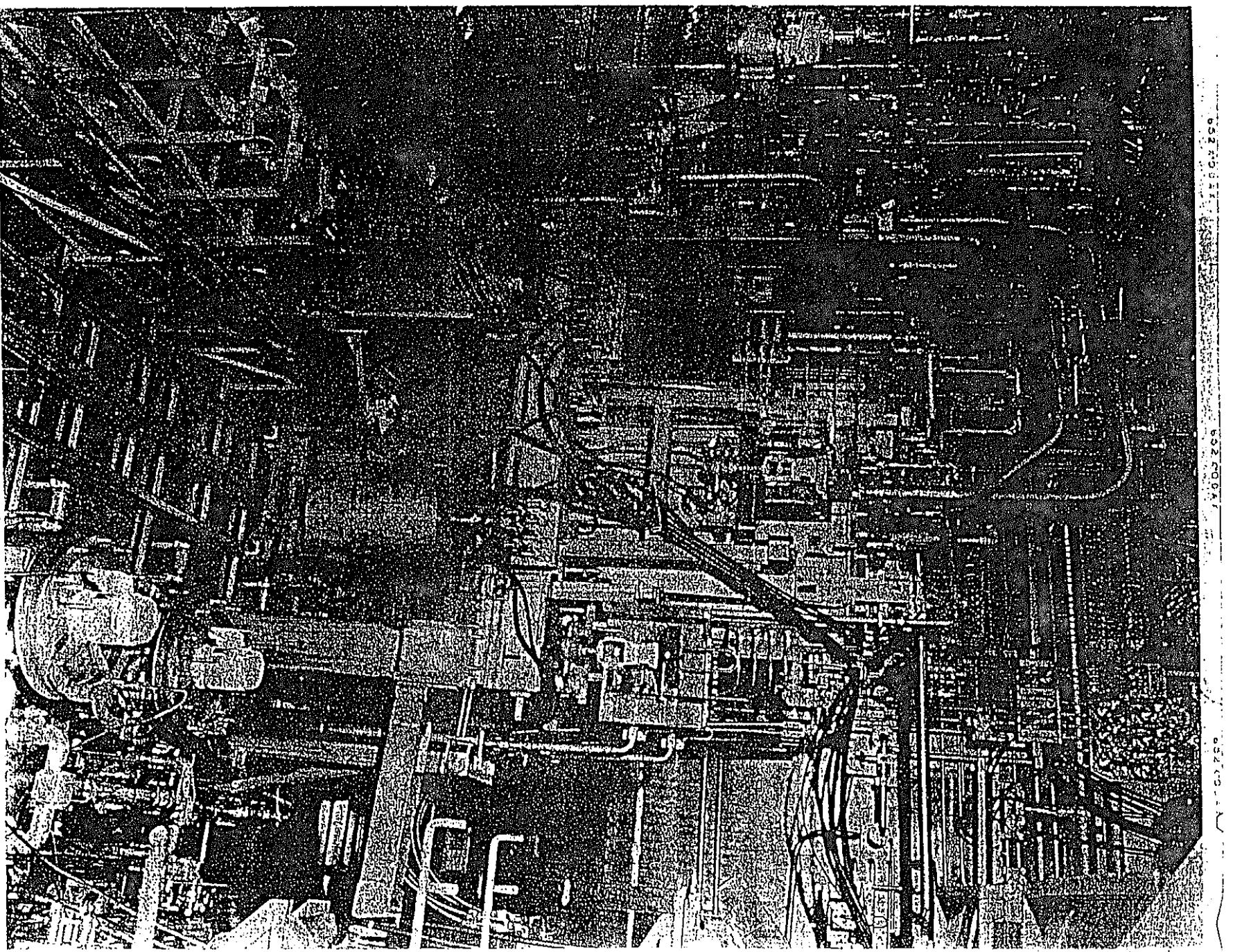




**RACK LAYOUT IN VITRIFICATION CELL**

PNC TN8100 94—003

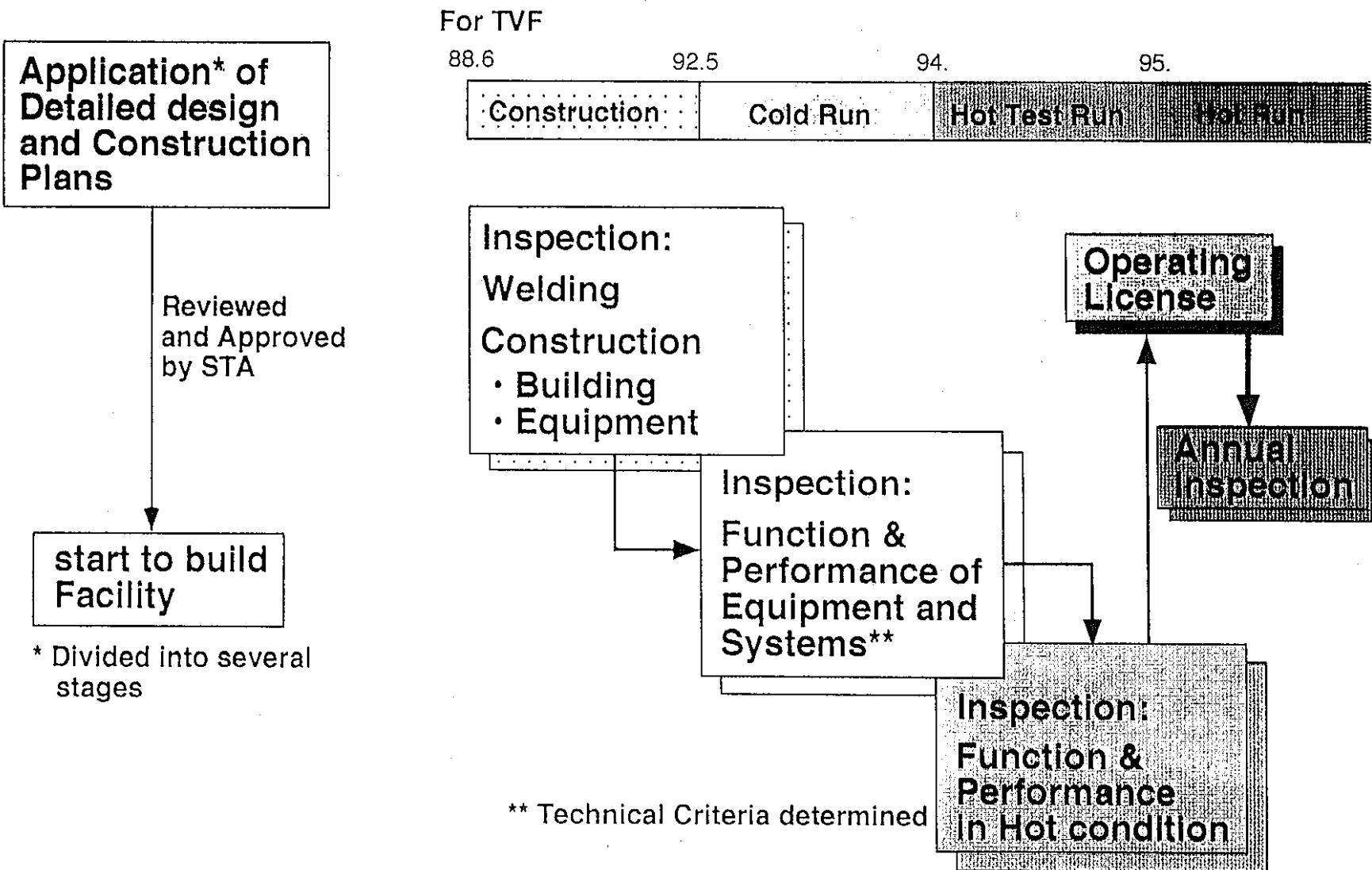




# S C H E D U L E O F T V F

ITEM	1992				1993				1994				1995			
	1	4	7	10	1	4	7	10	1	4	7	10	1	4	7	10
<b>1. Construction Testing</b>																
<b>2. Cold Test</b>																
1) Remote Maintenance					(I)	(II)	(III)	(IV)								
2) Process Equipment						(I) 21can.	(II) 21can. Ru, Rh,P d		(III) 41can.							
<b>3. Hot Test Operation</b> (incl. hot preparation)																
<b>4. Operation</b>																

## CONSTRUCTION AND INSPECTION CONDUCTED FOR OPERATION



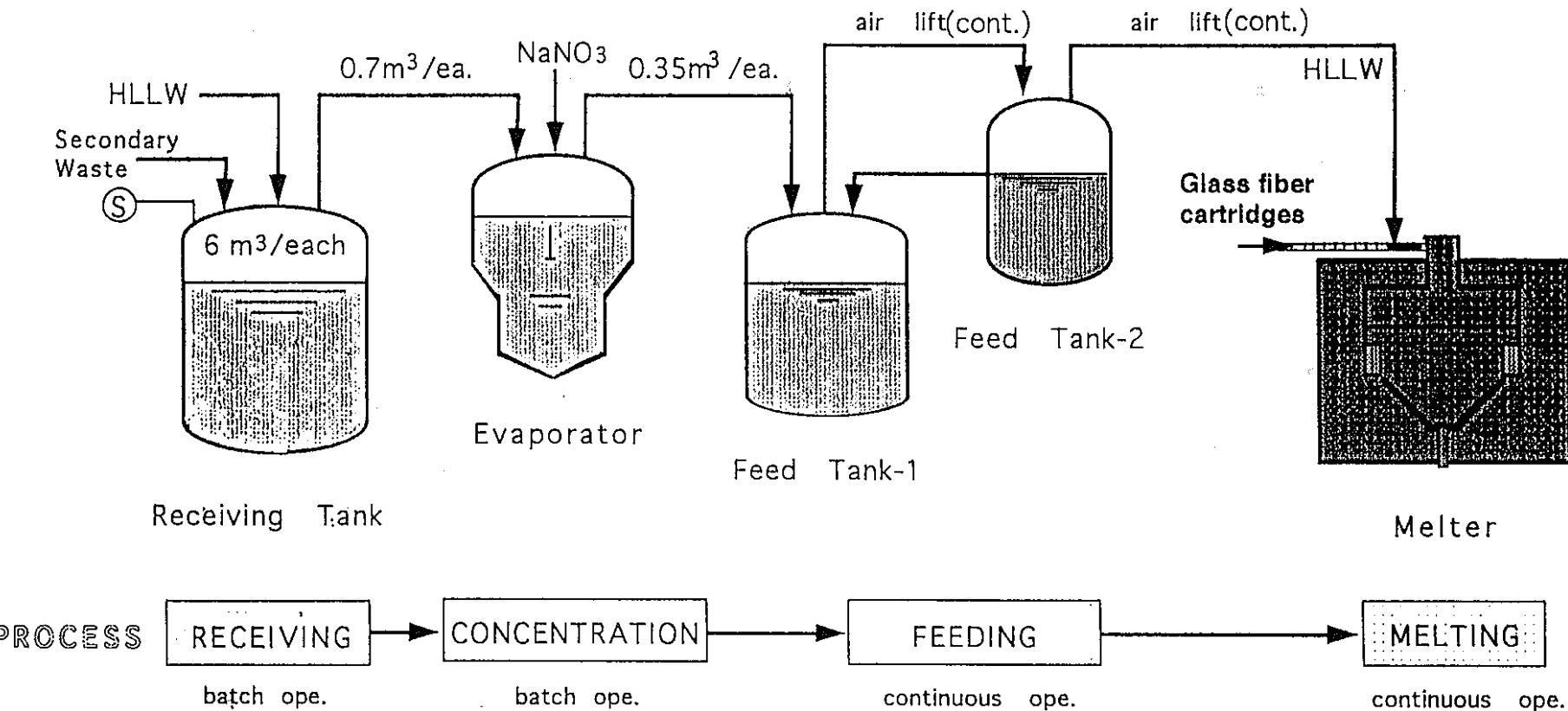
Commissioning Tests

# QUALITY CONTROL OF GLASS PRODUCT

CONTROL VALUE	
<b>Waste Oxide Content:</b>	≤ 30 wt% ( Target 25 wt% )
<b>Sodium Oxide Content:</b>	8~12 wt% ( Target 10wt% )

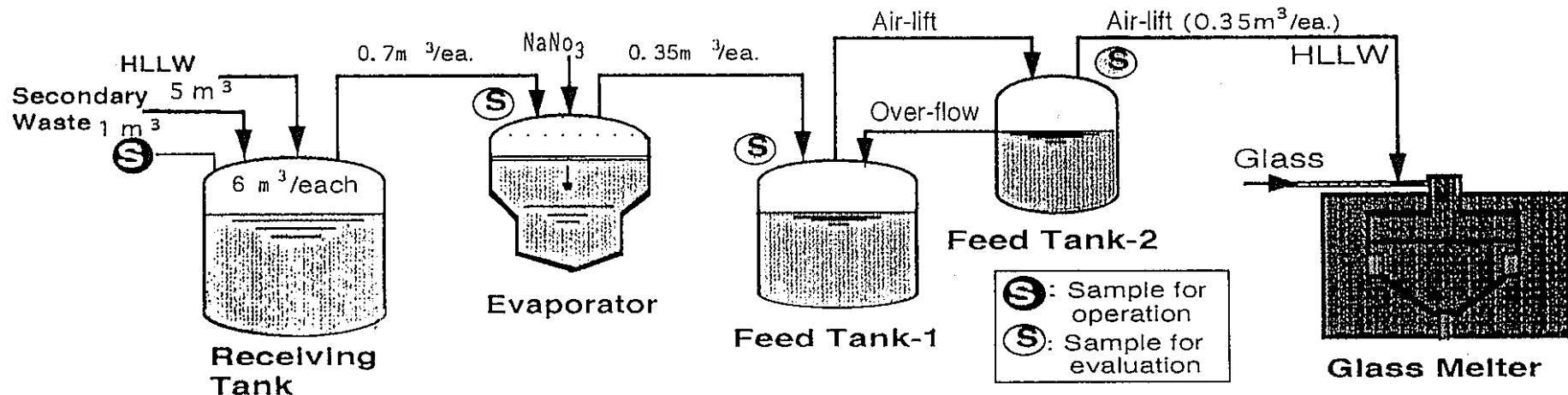
TVF STANDARD GLASS COMPOSITION

Glass elements	Content(%)	
SiO <sub>2</sub>	46.7	
B <sub>2</sub> O <sub>3</sub>	14.3	
Al <sub>2</sub> O <sub>3</sub>	5.0	
Li <sub>2</sub> O	3.0	
CaO	3.0	
ZnO	3.0	
Na <sub>2</sub> O	10.0	75wt%
P <sub>2</sub> O <sub>5</sub>	0.2	
Fe <sub>2</sub> O <sub>3</sub>	1.9	
NiO	0.5	
Cr <sub>2</sub> O <sub>3</sub>	0.5	
FP Oxide	9.6	
Actinide	2.3	
Total	100.0	25wt%



- |                        |  |  |   |  |
|------------------------|--|--|---|--|
| <b>PROCESS CONTROL</b> | <ul style="list-style-type: none"> <li>Analyze Waste Oxide [WO], Na<sub>2</sub>O content (g/l)</li> <li>Start process control based on these data</li> </ul> | <ul style="list-style-type: none"> <li>Adjust Na<sub>2</sub>O content by adding NaNO<sub>3</sub></li> <li>Control the concentration ratio (about 2 times)</li> </ul> | <ul style="list-style-type: none"> <li>Determine HLLW feed rate(L/h), glass feed rate (sec/cartridge) by estimated WO, Na<sub>2</sub>O content</li> <li>Control the feed rates to get 25% of WO, 10% of Na<sub>2</sub>O in glass product</li> </ul> | <ul style="list-style-type: none"> <li>Control Glass Melting</li> <li>Control Glass Pouring (Batch Pouring)</li> </ul> |
|------------------------|--|--|---|--|

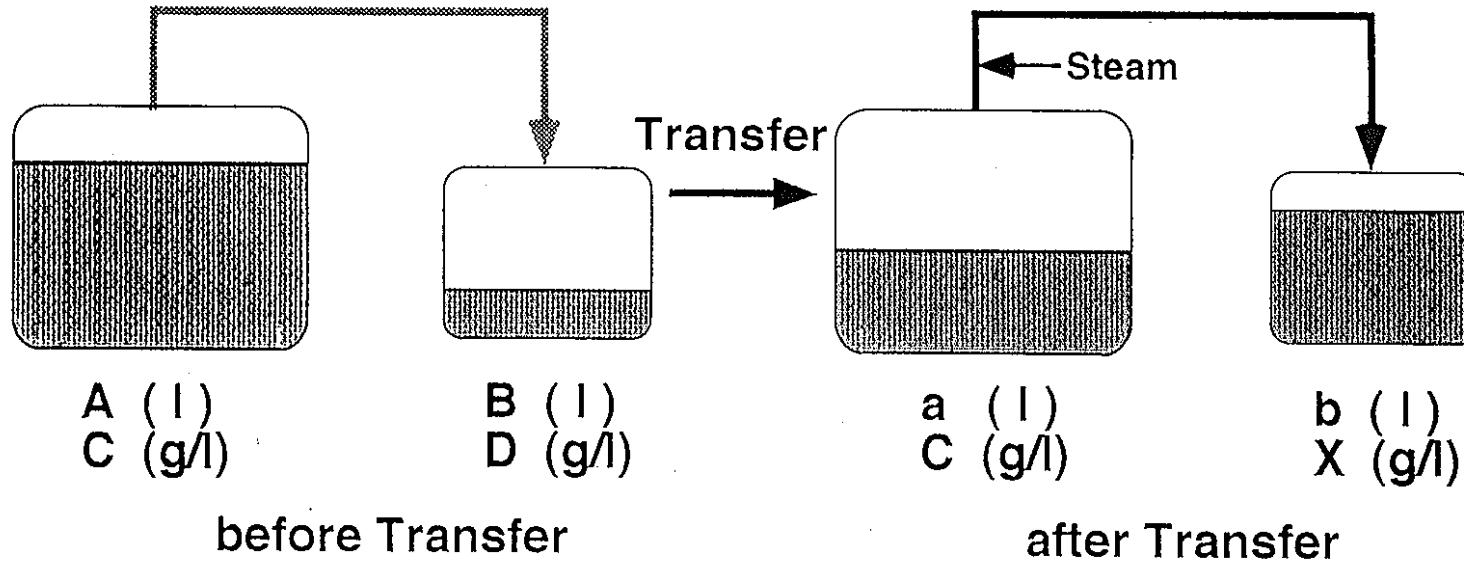
## PROCESS CONTROL FOR GLASS PRODUCT QUALITY



	Receiving Tank	Evaporator	Feed Tank-1 - Tank-2	
Process control	Analysis (Total waste oxide [WO] and sodium oxide [Na <sub>2</sub> O])	<ul style="list-style-type: none"> <li>Estimation of HLLW concen. after transfer</li> <li>Setting of HLLW(l/h) and Glass(sec/each) feed rate</li> <li>Adjustment of Na<sub>2</sub>O, Concentration at 2 times</li> <li>Estimation of concen.</li> </ul>	<ul style="list-style-type: none"> <li>Estimation of HLLW concen. after transfer</li> <li>Setting of HLLW(l/h) and Glass(sec/each) feed rate</li> <li>Adjustment of Na<sub>2</sub>O, Concentration at 2 times</li> <li>Operation value(WO, Na<sub>2</sub>O) = Estimated concen. X HLLW feed / (Estimated concen. X HLLW feed + Glass feed)</li> </ul>	<ul style="list-style-type: none"> <li>Estimation of HLLW concen. after transfer</li> <li>Setting of HLLW(l/h) and Glass(sec/each) feed rate</li> <li>Adjustment of Na<sub>2</sub>O, Concentration at 2 times</li> <li>Operation value(WO, Na<sub>2</sub>O) = Estimated concen. X HLLW feed / (Estimated concen. X HLLW feed + Glass feed)</li> </ul>
TEST ITEM	Estimation method of concen.			Comparison of estimated concentration with analyzed one
	Quantitative feed			Evaluation of feed result (feed accuracy) with target in HLLW and Glass
	Control accuracy of product			<ul style="list-style-type: none"> <li>Comparison of analyzed value (WO, Na<sub>2</sub>O) with Operation value</li> <li>Comparison with solidified glass</li> </ul>

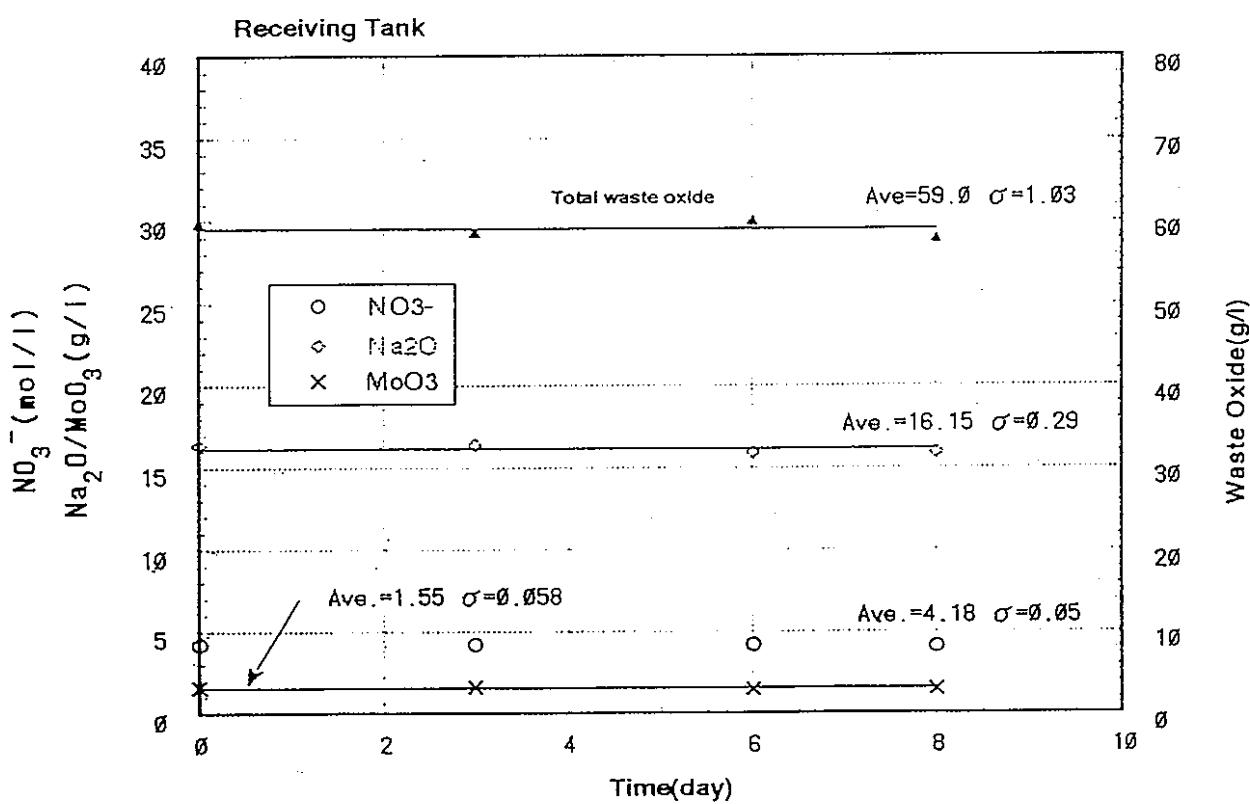
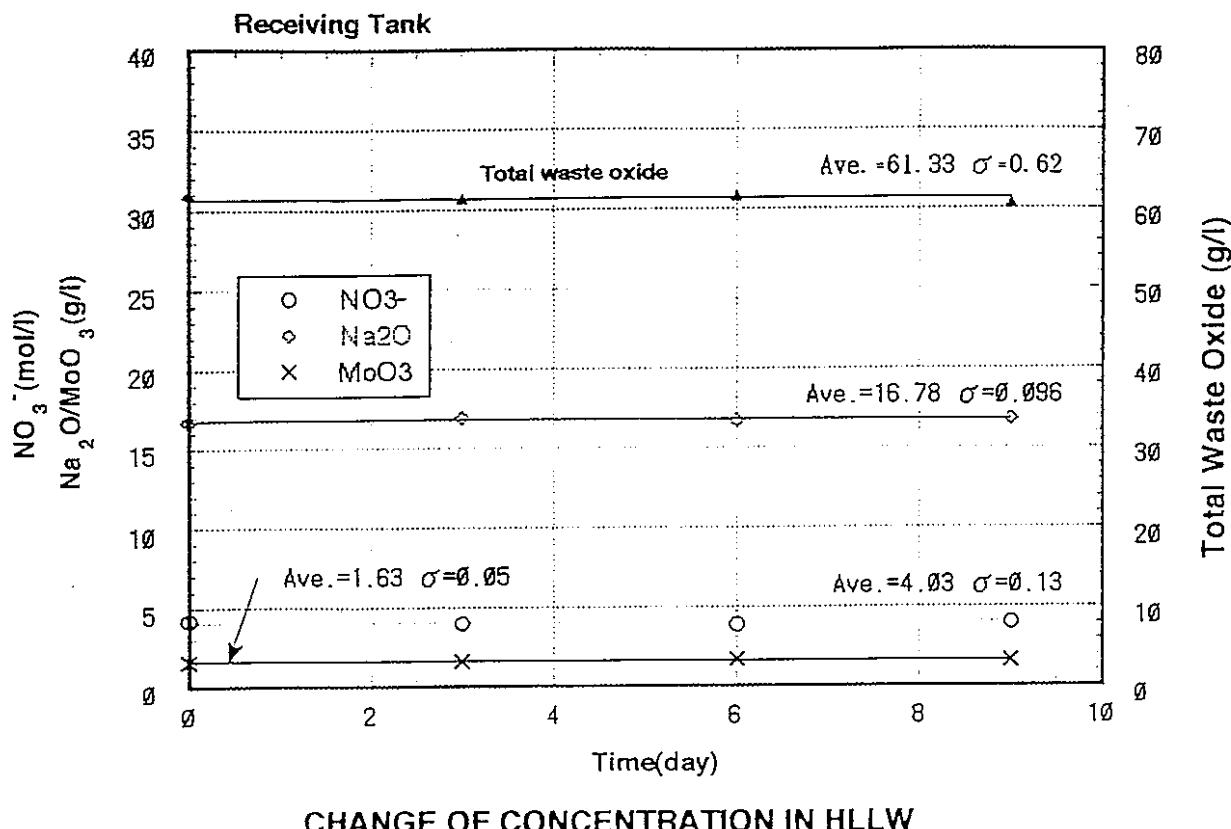
TEST FOR GLASS PRODUCT QUALITY CONTROL

## Estimation method of HLLW Concentration(W.O, Na<sub>2</sub>O)



Equation

$$X = \frac{(A - a) \times C + B \cdot D}{b}$$

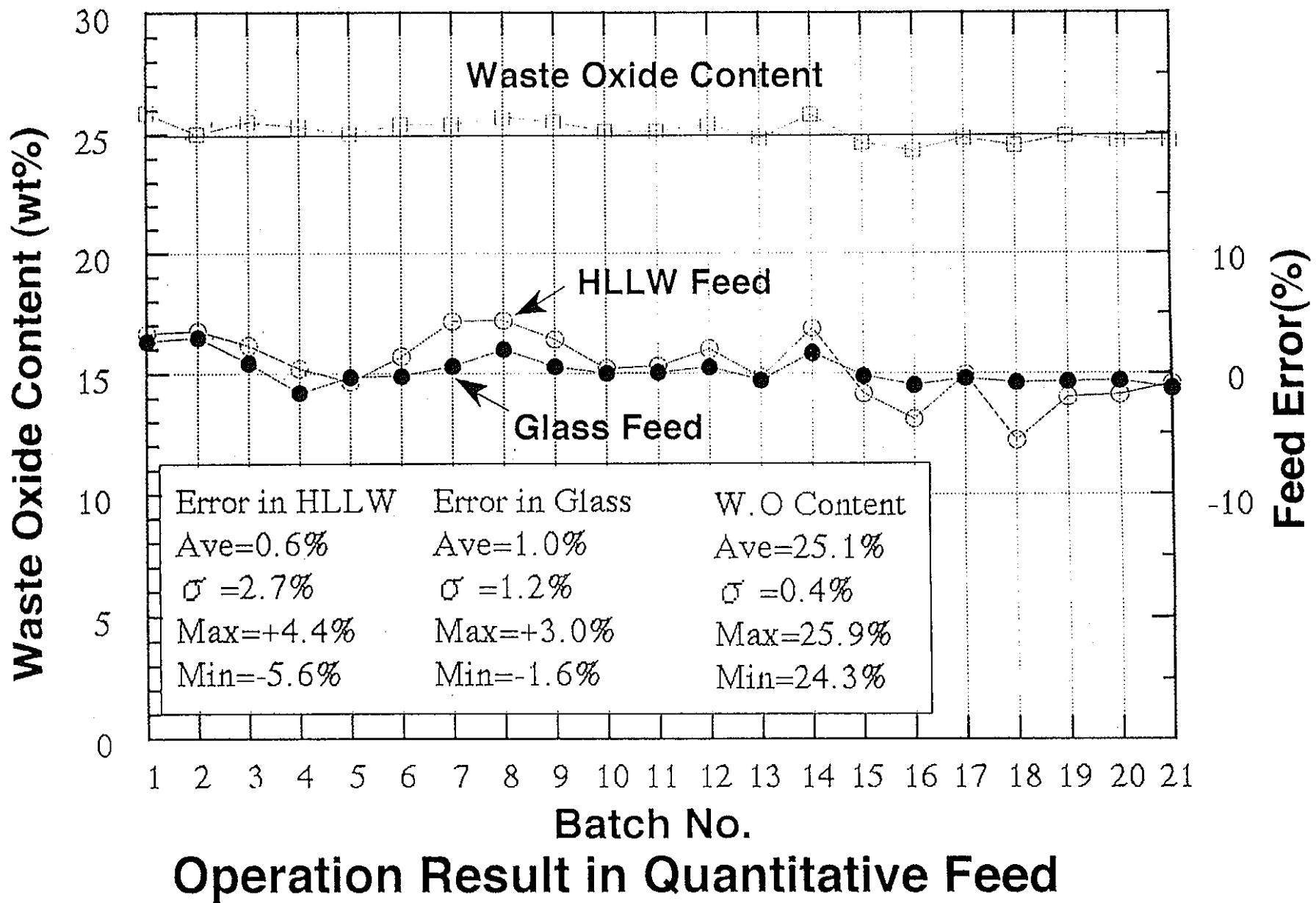
**CHANGE OF CONCENTRATION IN HLLW AND RECOVERY**

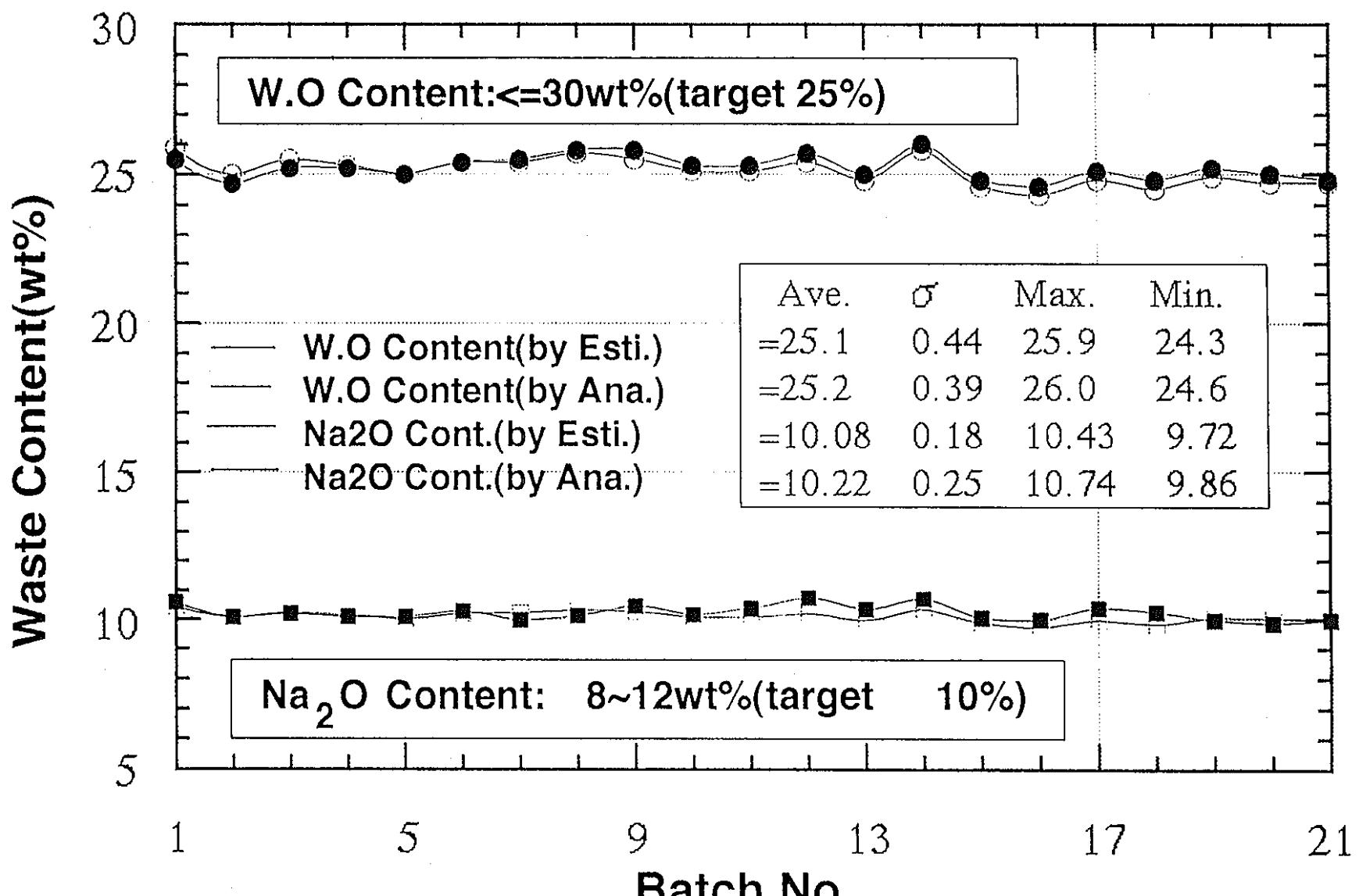
# Evaluation of Estimation Accuracy for HLLW Concentration

Transfer		Receiving Tank --> Evaporator		Evaporator --> Feed Tnk-1	
		Control Item	Content (wt%)	Error (%)	Content (wt%)
Total Waste Oxide	Ave.	24.9	-0.41	25.0	0.04
	$\sigma$	0.65	2.61	0.36	1.45
Sodium Oxide	Ave.	9.98	-0.15	10.18	1.81
	$\sigma$	0.44	4.44	0.32	3.25

Assump.: Feed amount to get the target content by estimated concentration  
 Evalua. : Content by analyzed concentration and feed amount

$$\text{Error(%)} = \frac{(\text{Content by Analysis}) - (\text{Content by Estimation})}{(\text{Content by Analysis})}$$





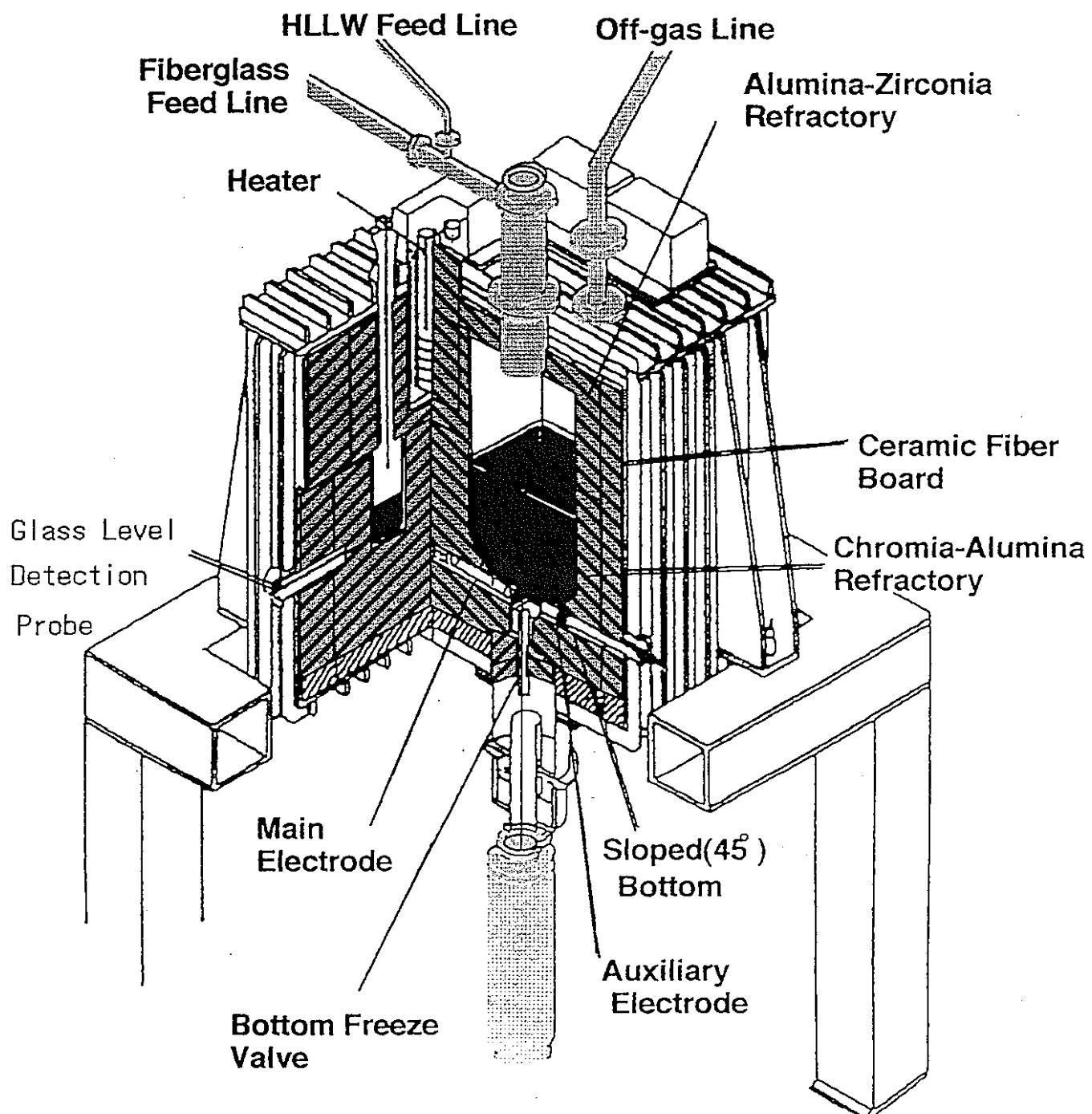
## RESULT OF GLASS PRODUCT CONTROL

# TEST RESULT FOR GLASS PRODUCT QUALITY CONTROL

Evaluation Method	Control Item	Total waste oxide (wt%) (target:25wt%)			Sodium oxide (wt%) (target:10wt%)		
		Max.	Min.	Ave.	Max.	Min.	Ave.
Eval. by Analysis data		26.0	24.6	25.2	10.7	9.9	10.2
Eval. by Estimated data		25.9	24.3	25.1	10.4	9.7	10.1
Eval. by Analysis of Glass		26.3	24.2	25.2	11.0	10.0	10.7

## Statistical Evaluation of Control Limit with 95% Confidence

Total Waste Oxide: Target  $\pm$  1.2 wt%  
 Sodium Oxide : Target  $\pm$  0.8 wt%



## TVF MELTER STRUCTURE

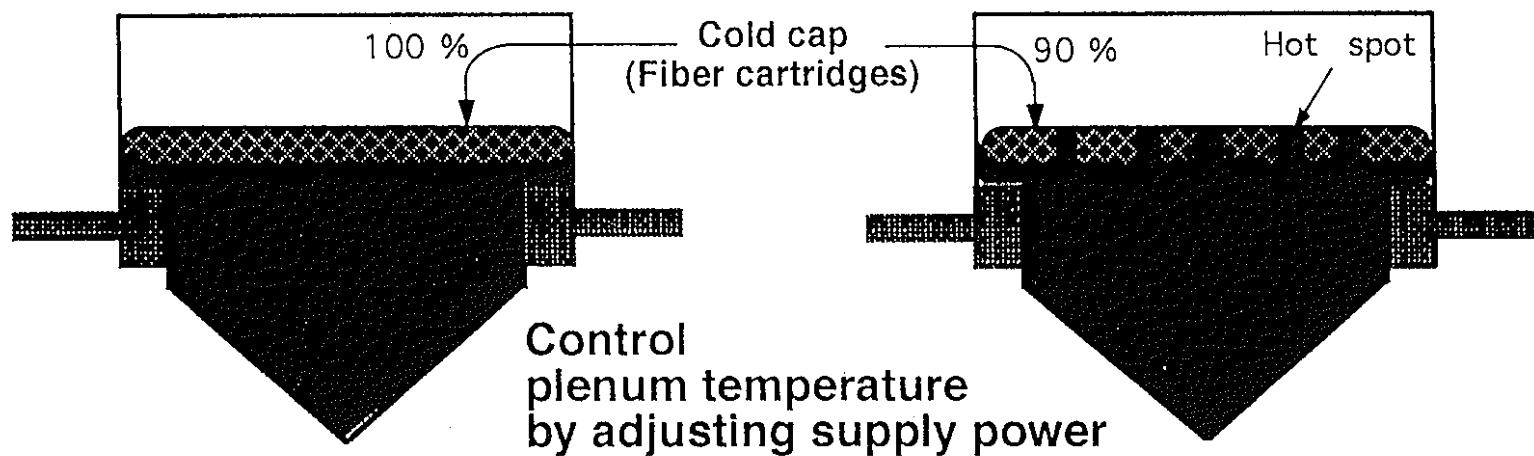
## PROCESS CONTROL FOR THE MELTER OPERATION

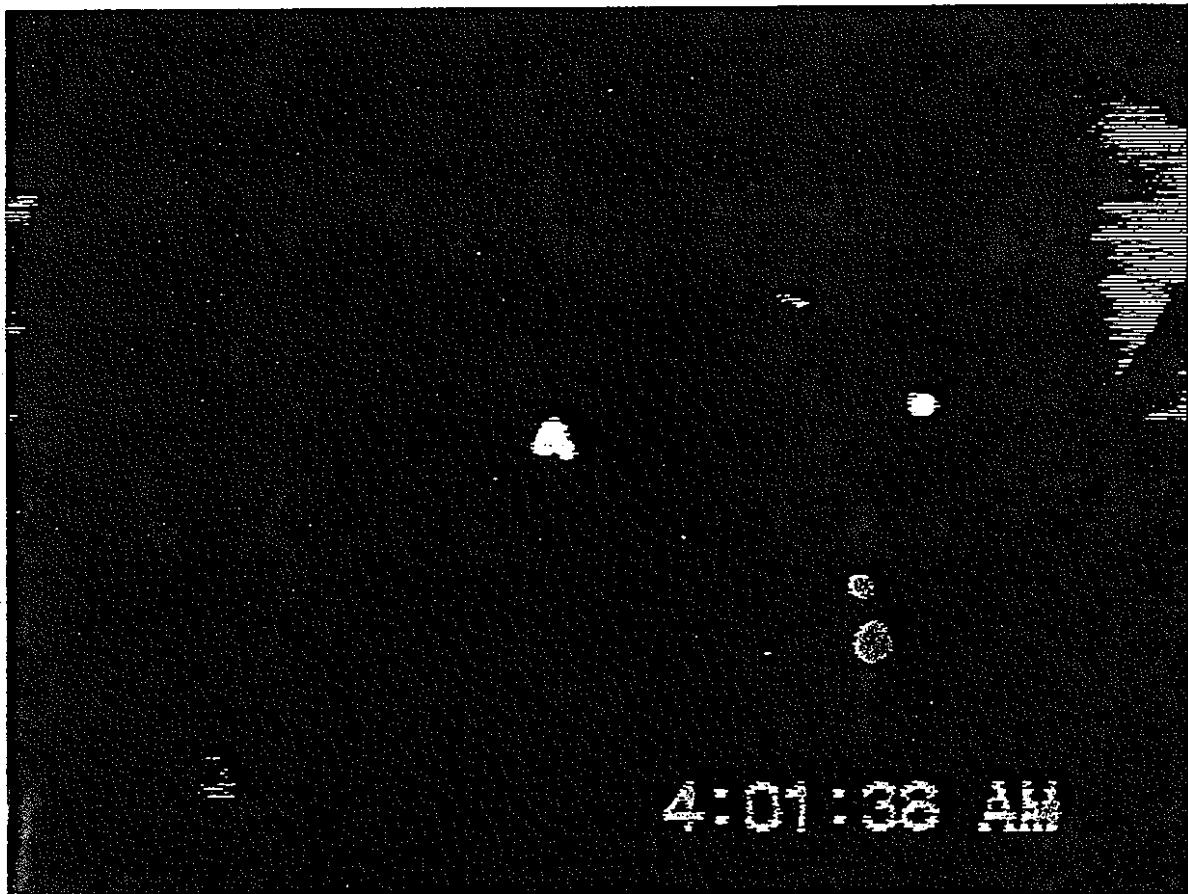
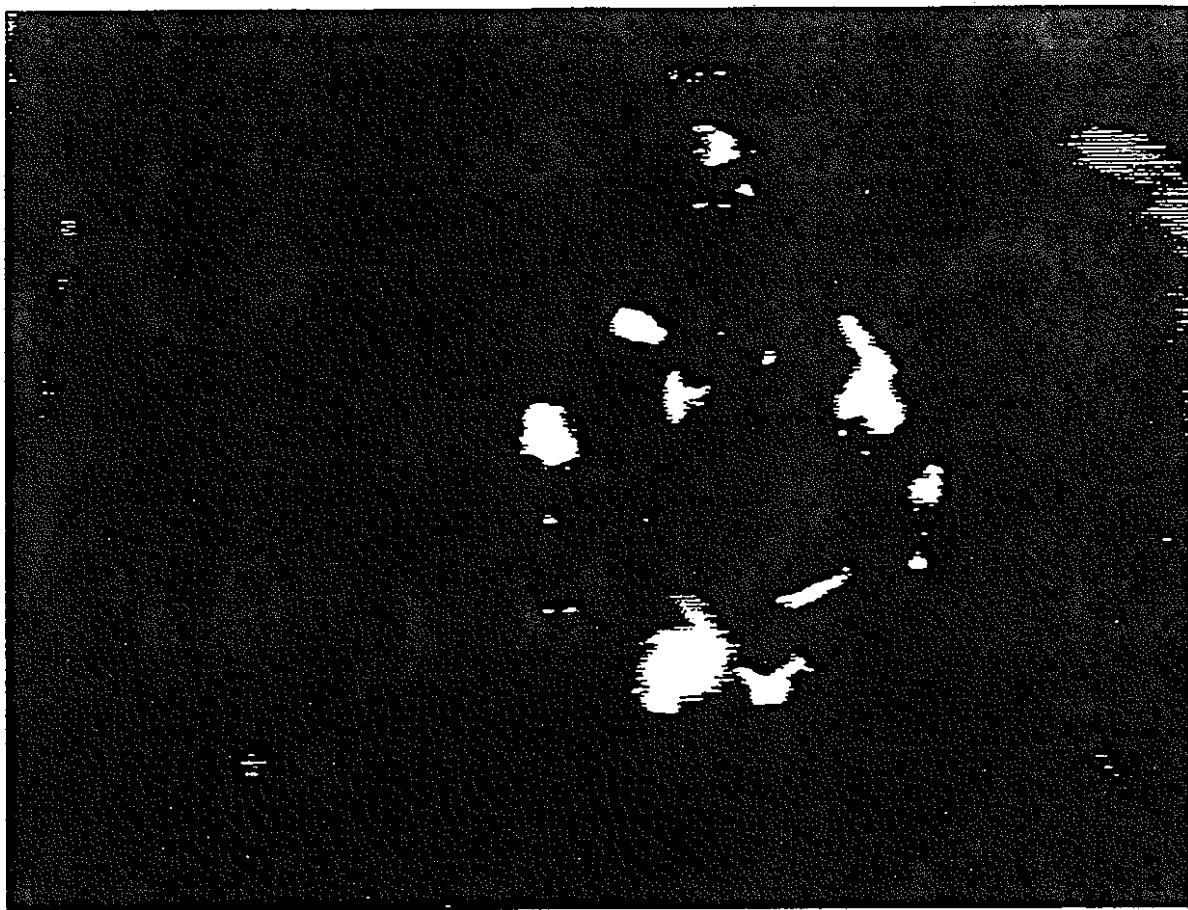
### (1) Low Plenum Temp. Operation

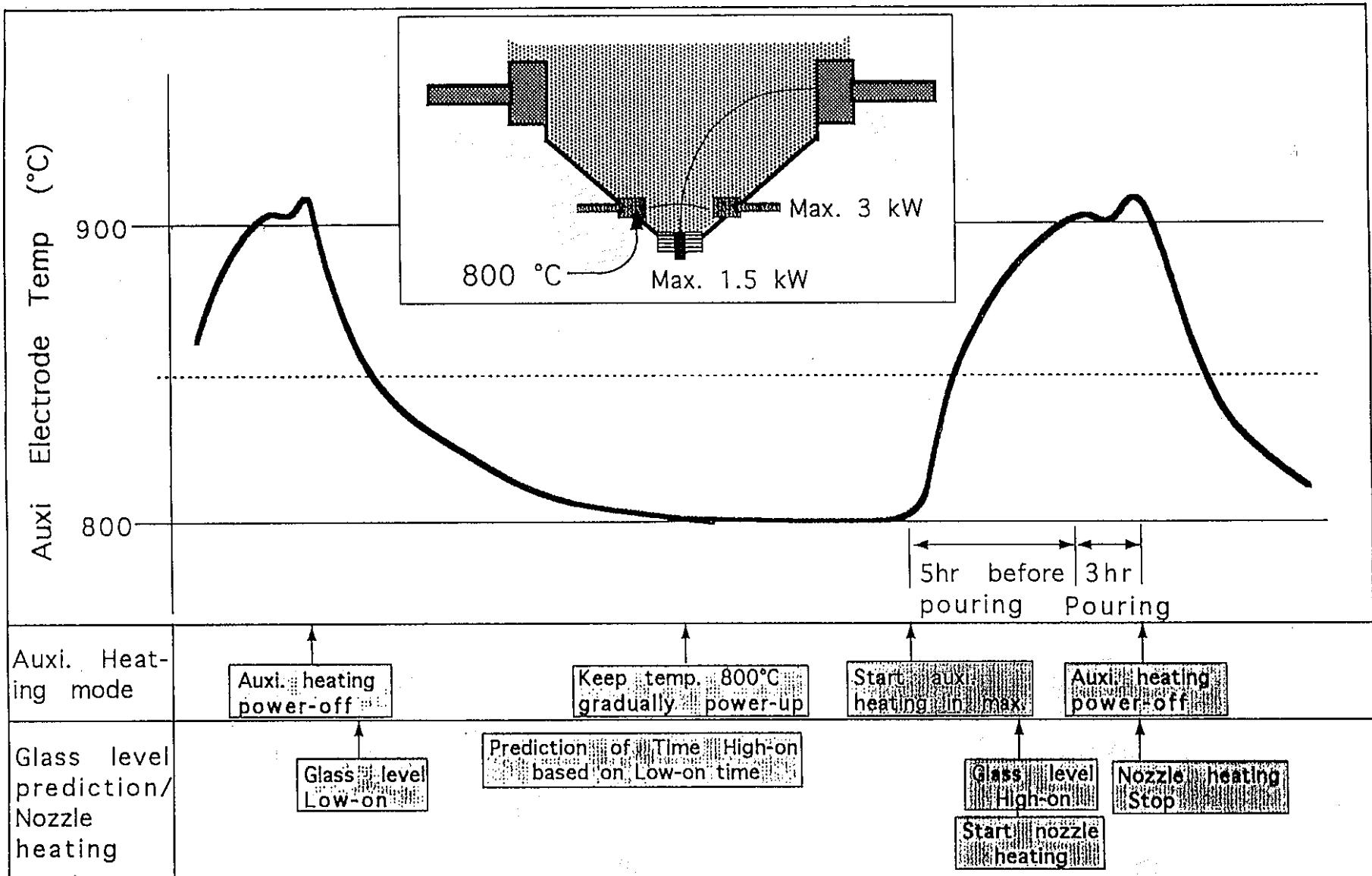
- about 300°C in Plenum space
- No Glass surface (almost 100 % Cold cap coverage)
- No pressure surge (due to Fiber cartridge)

### (2) High Plenum Temp. Operation

- 400-500°C in Plenum space (Exposure of glass surface)
- Expect high Melt rate
- Increment of Particle release
- Difficult to control bottom temp.

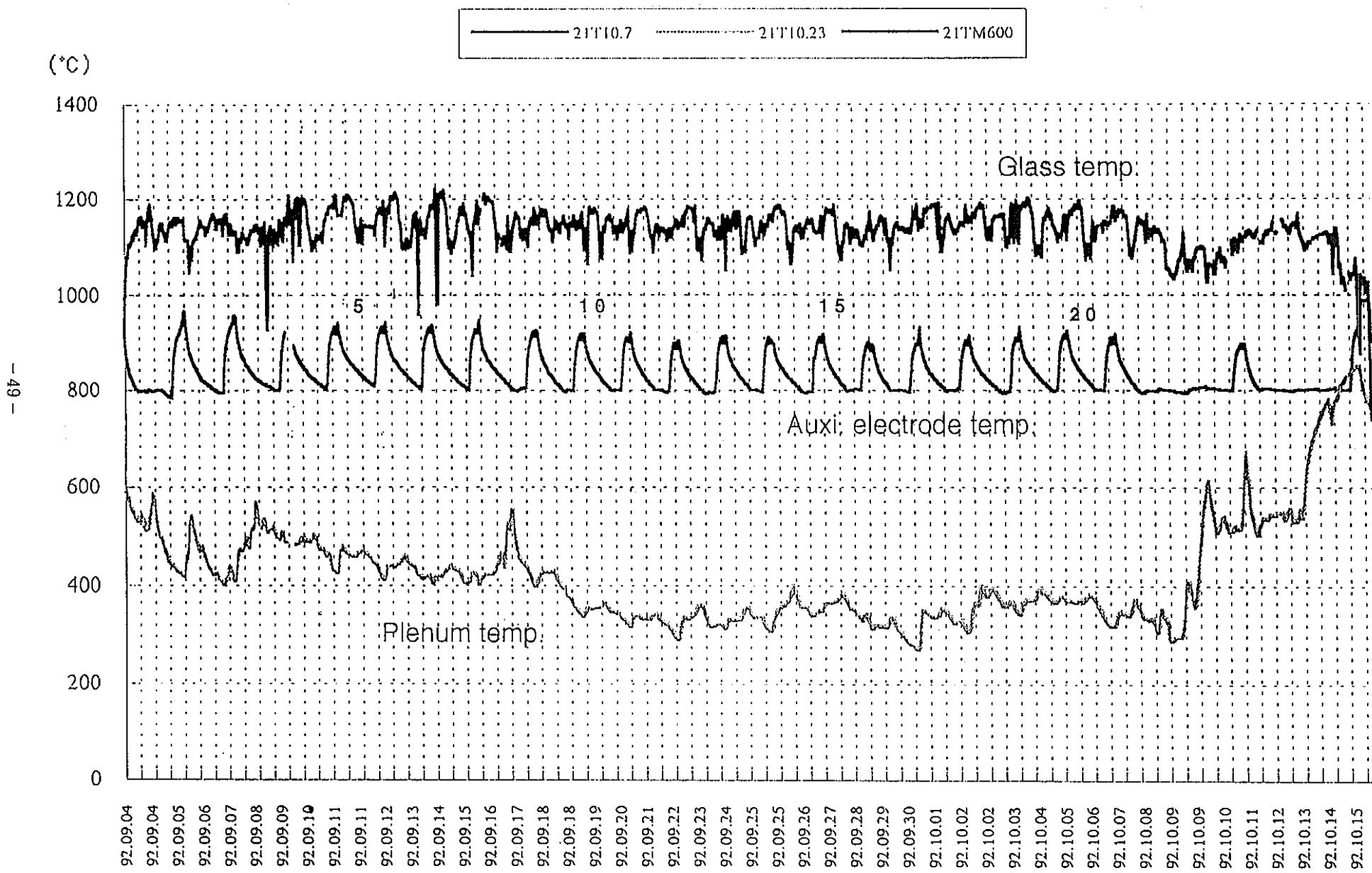




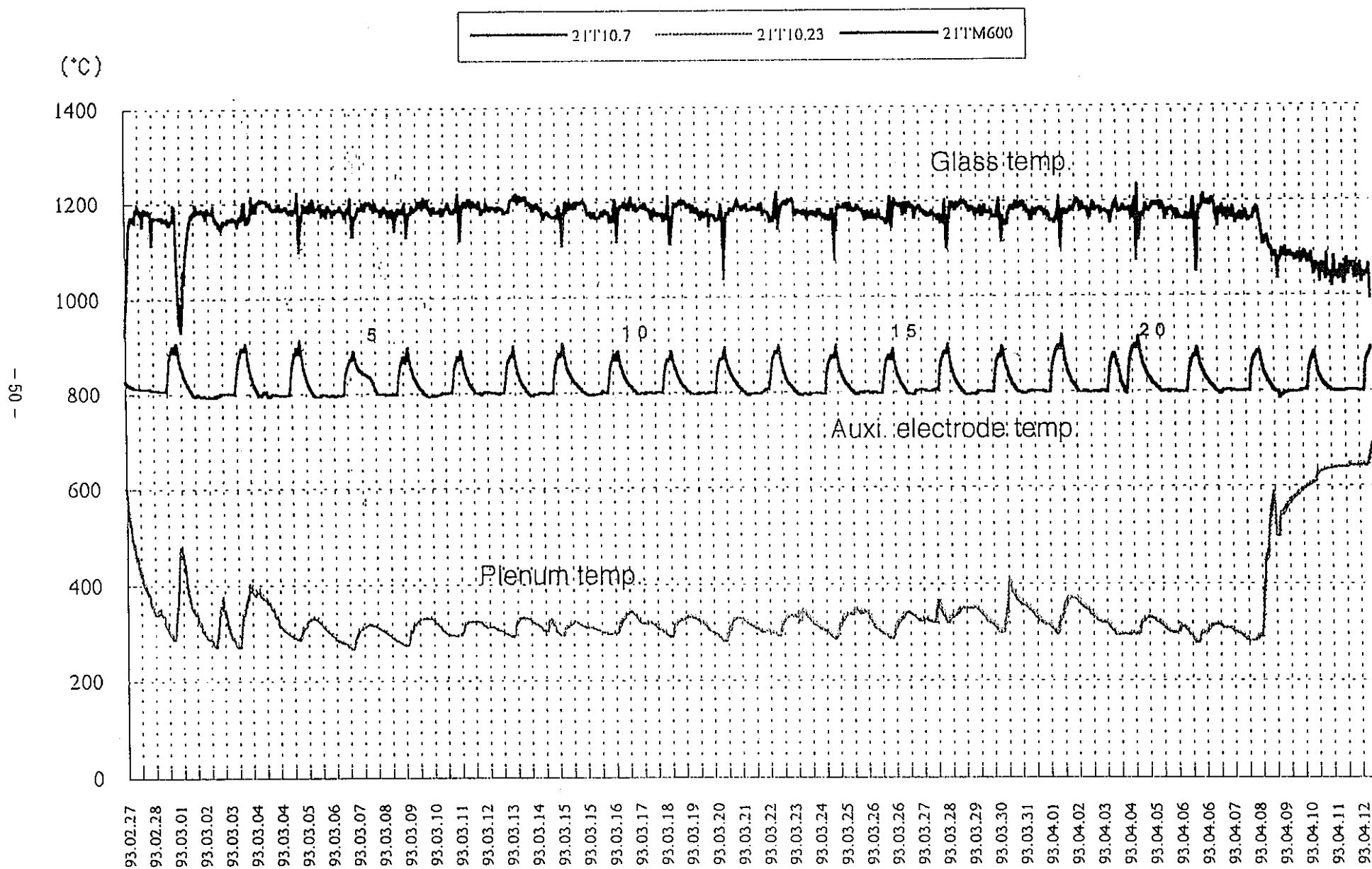


## MELTER OPERATION BY BOTTOM TEMP. CONTROL MODE

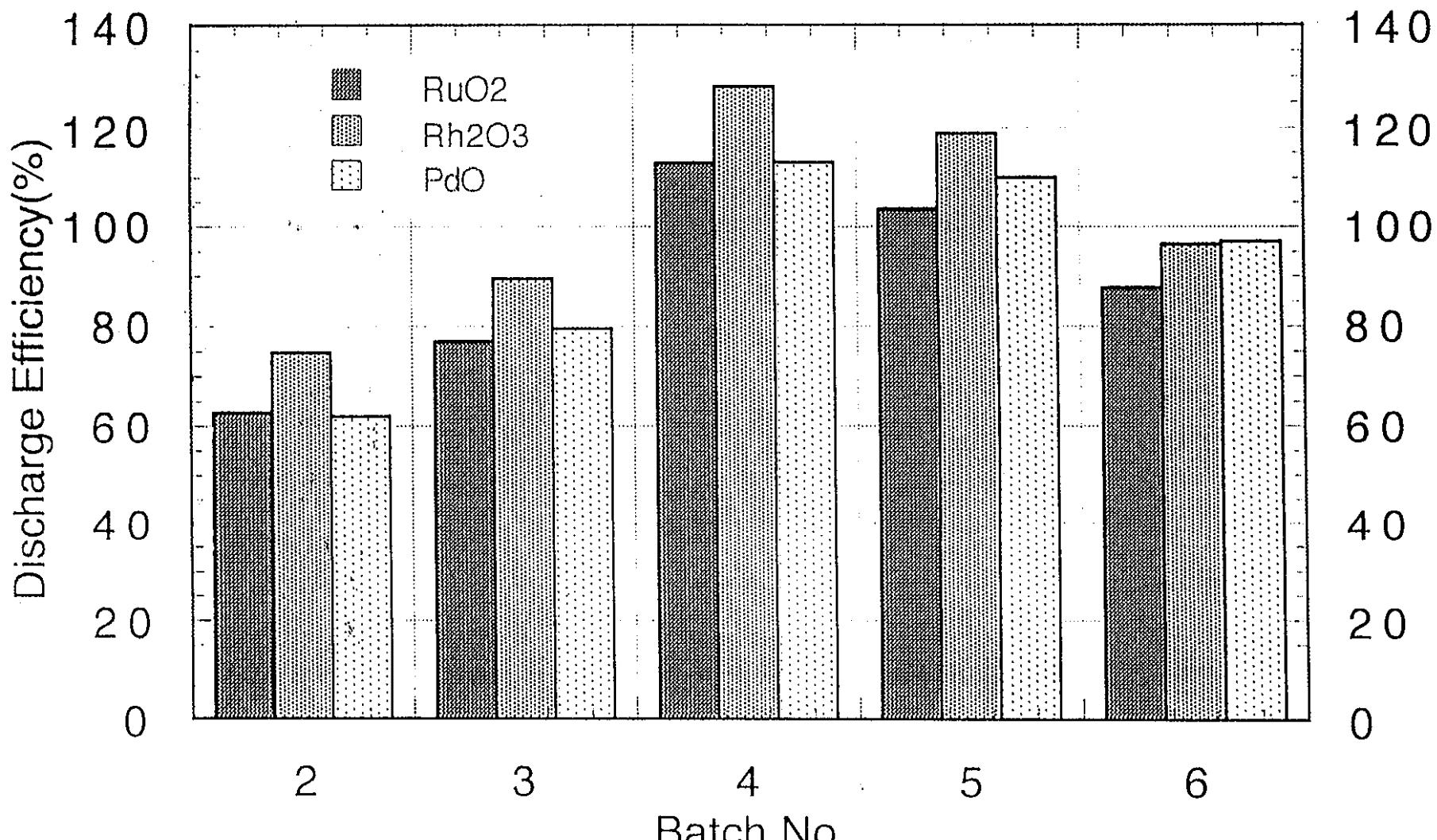
## MELTER OPERATION RESULT(1ST RUN)



## MELTER OPERATION RESULT (2ND RUN)

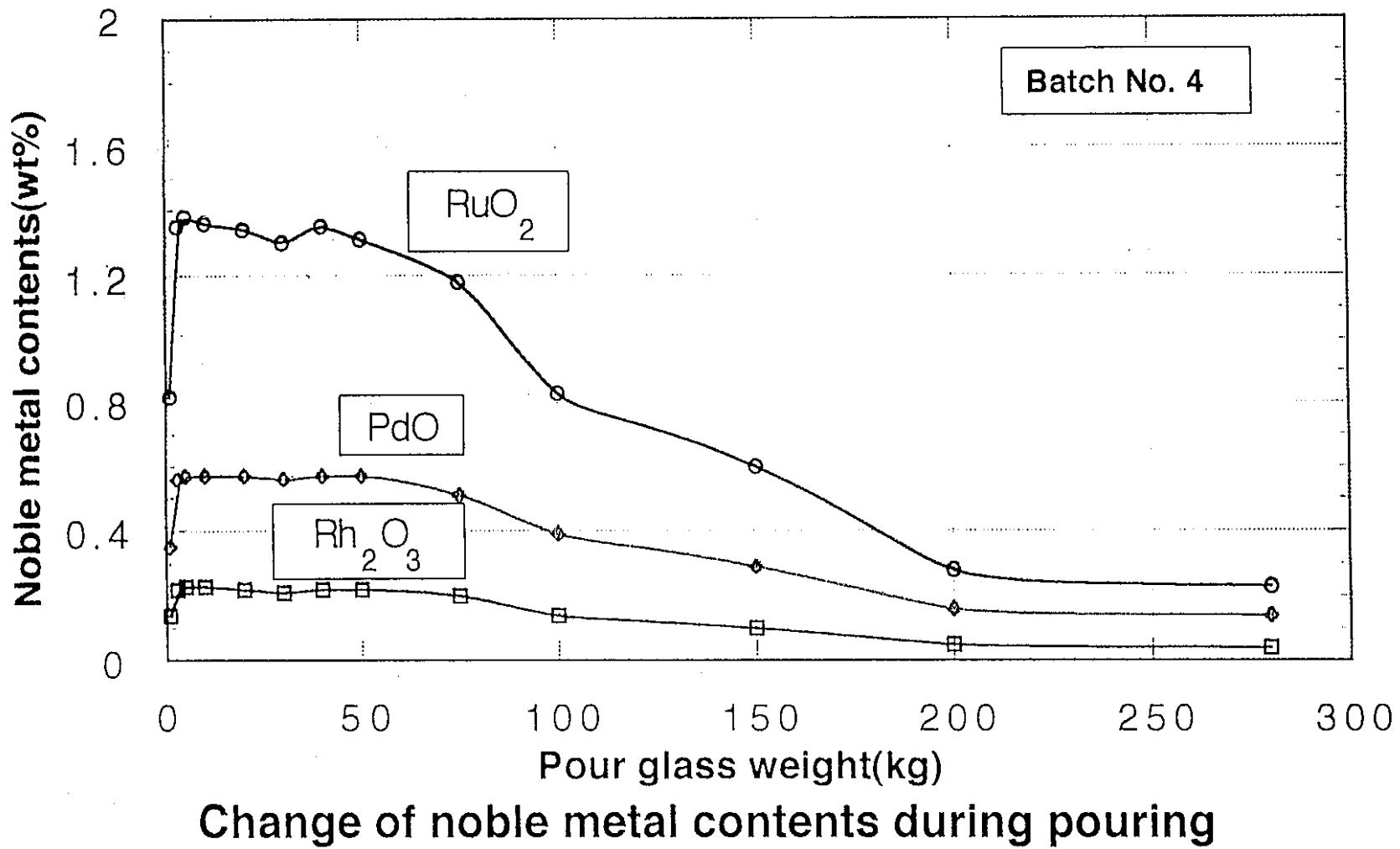


- 15 -

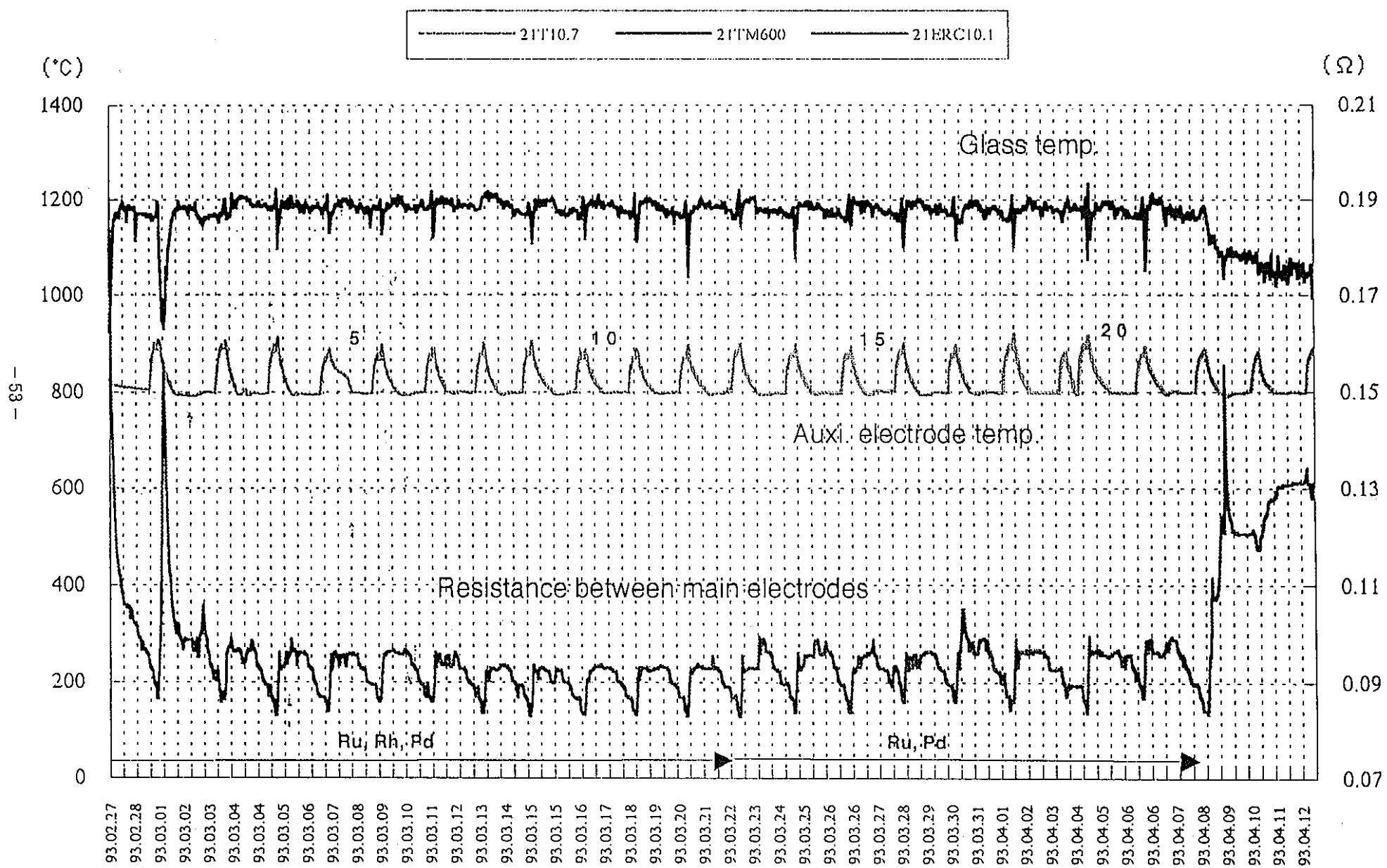


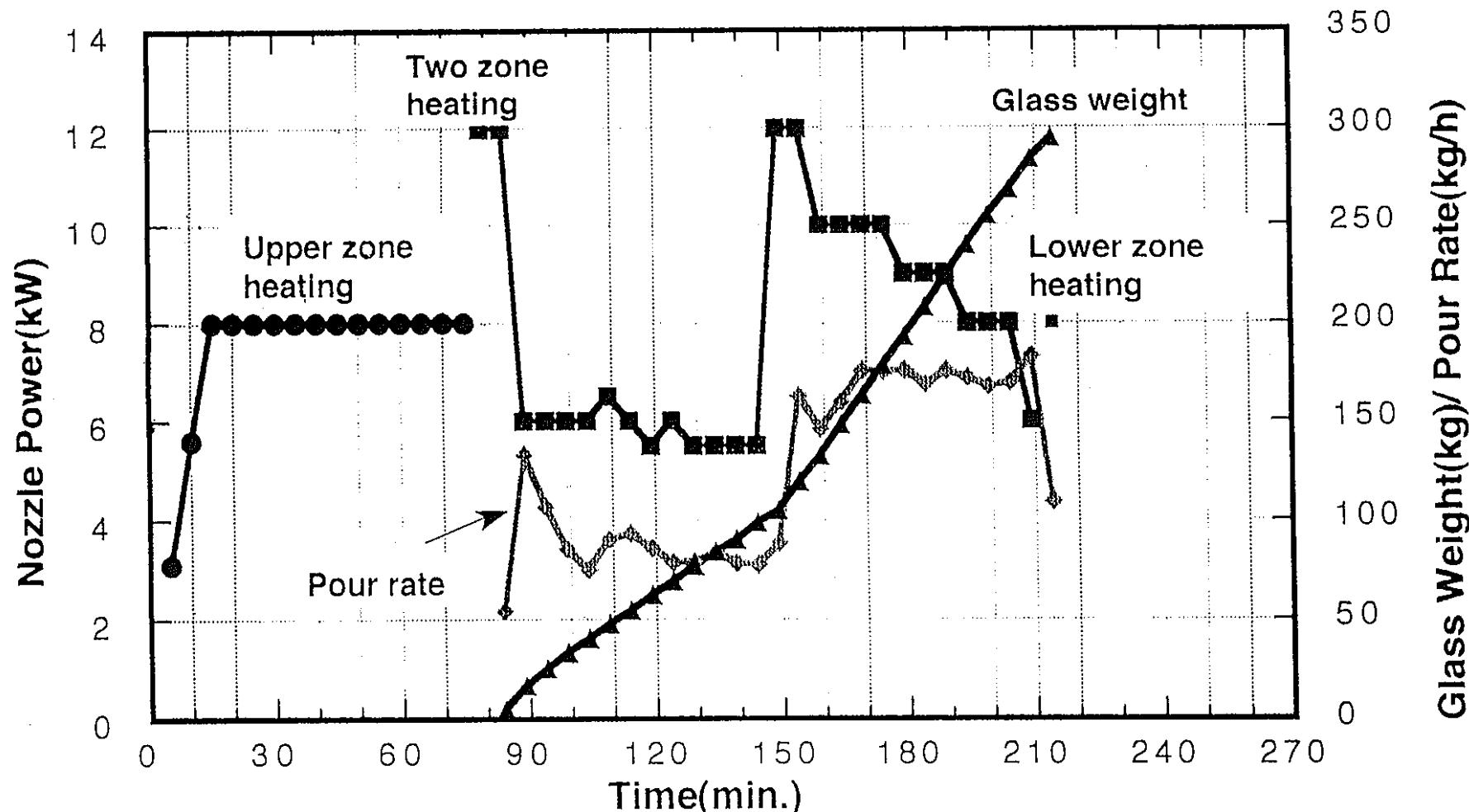
## DISCHARGE OF NOBLE METALS IN MELTER OPERATION

- 52 -

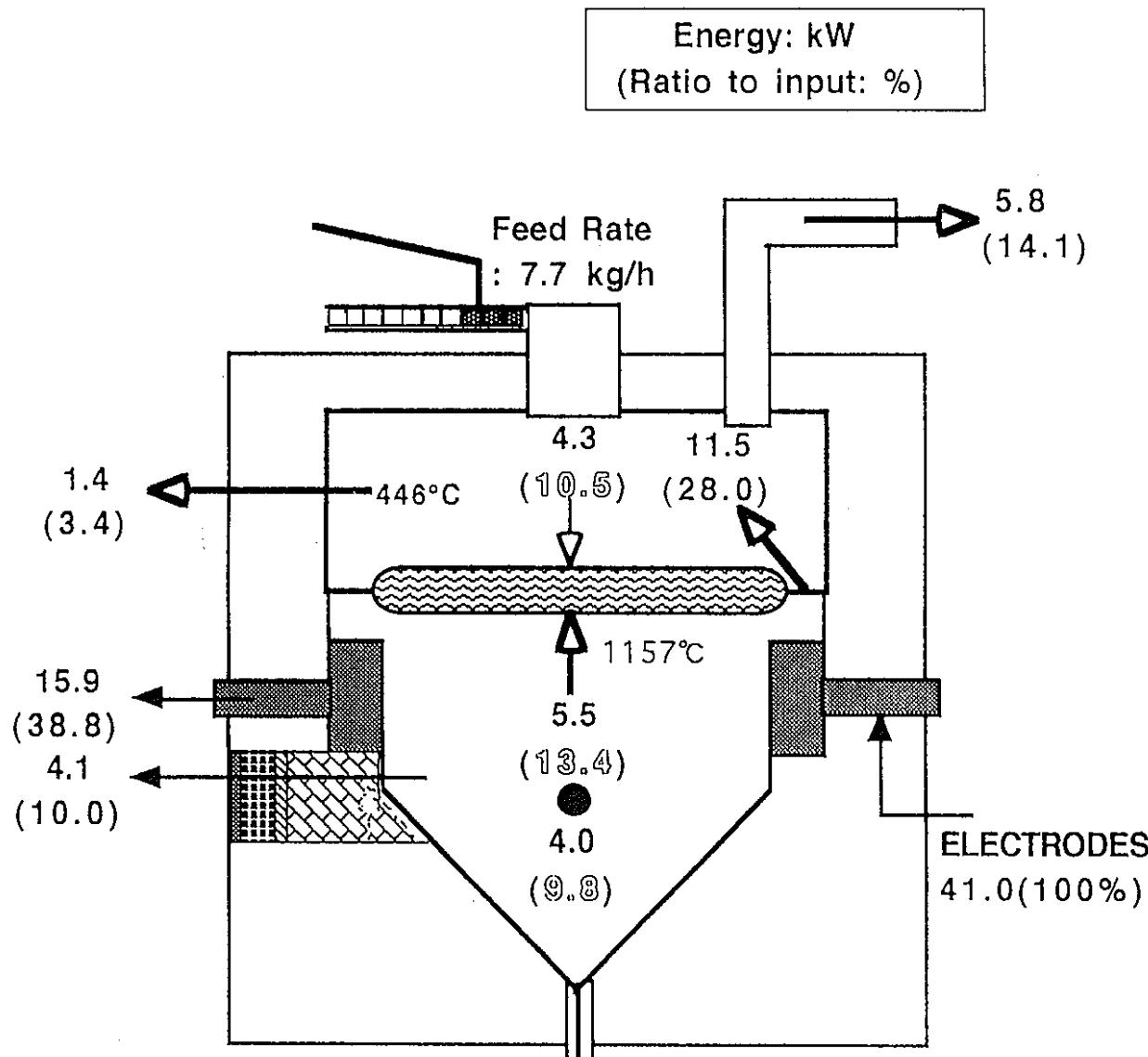


## CHANGE OF RESISTANCE (2ND MELTER RUN)



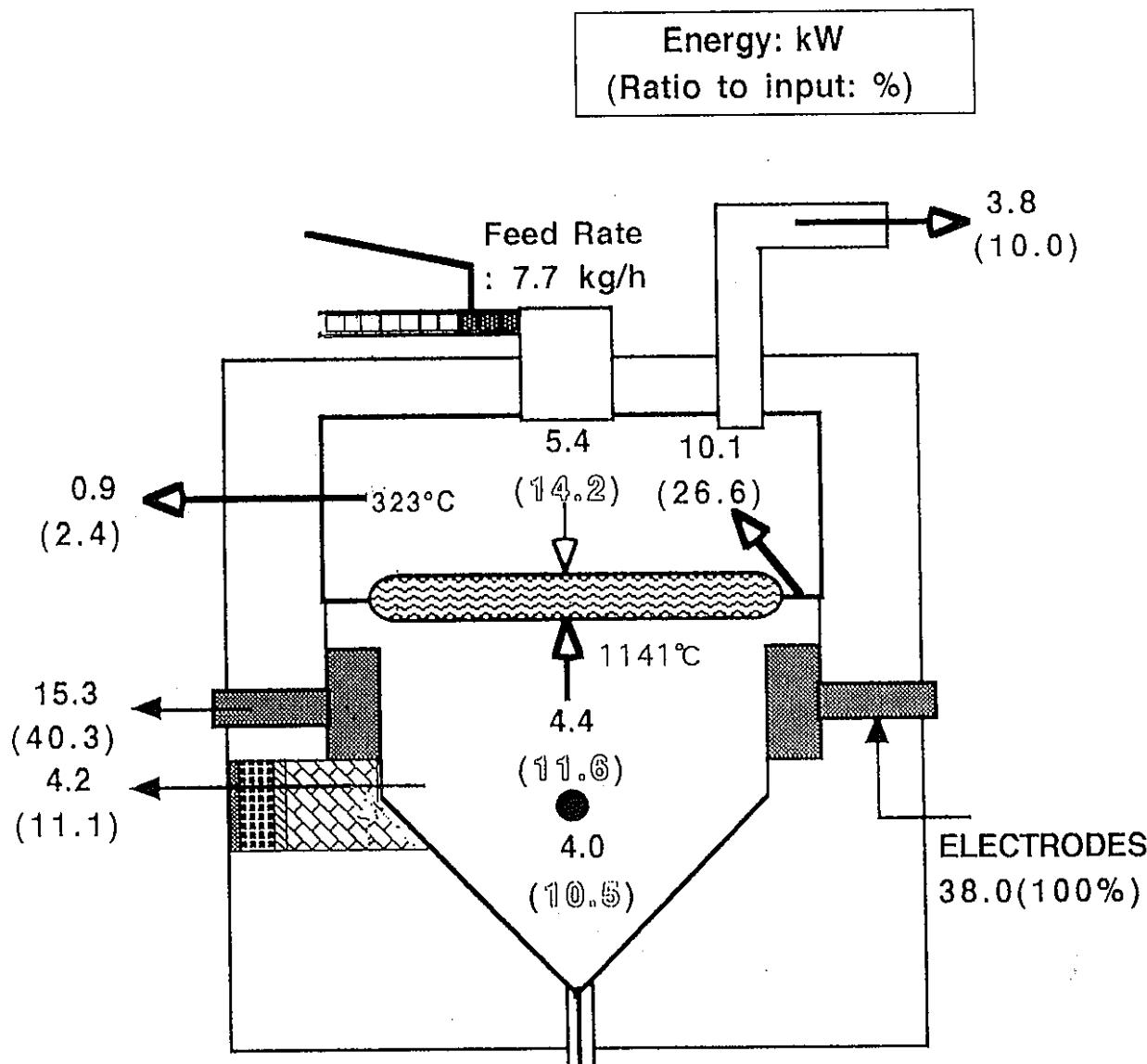


**GLASS POURING RESULT IN 2ND MELTER RUN**



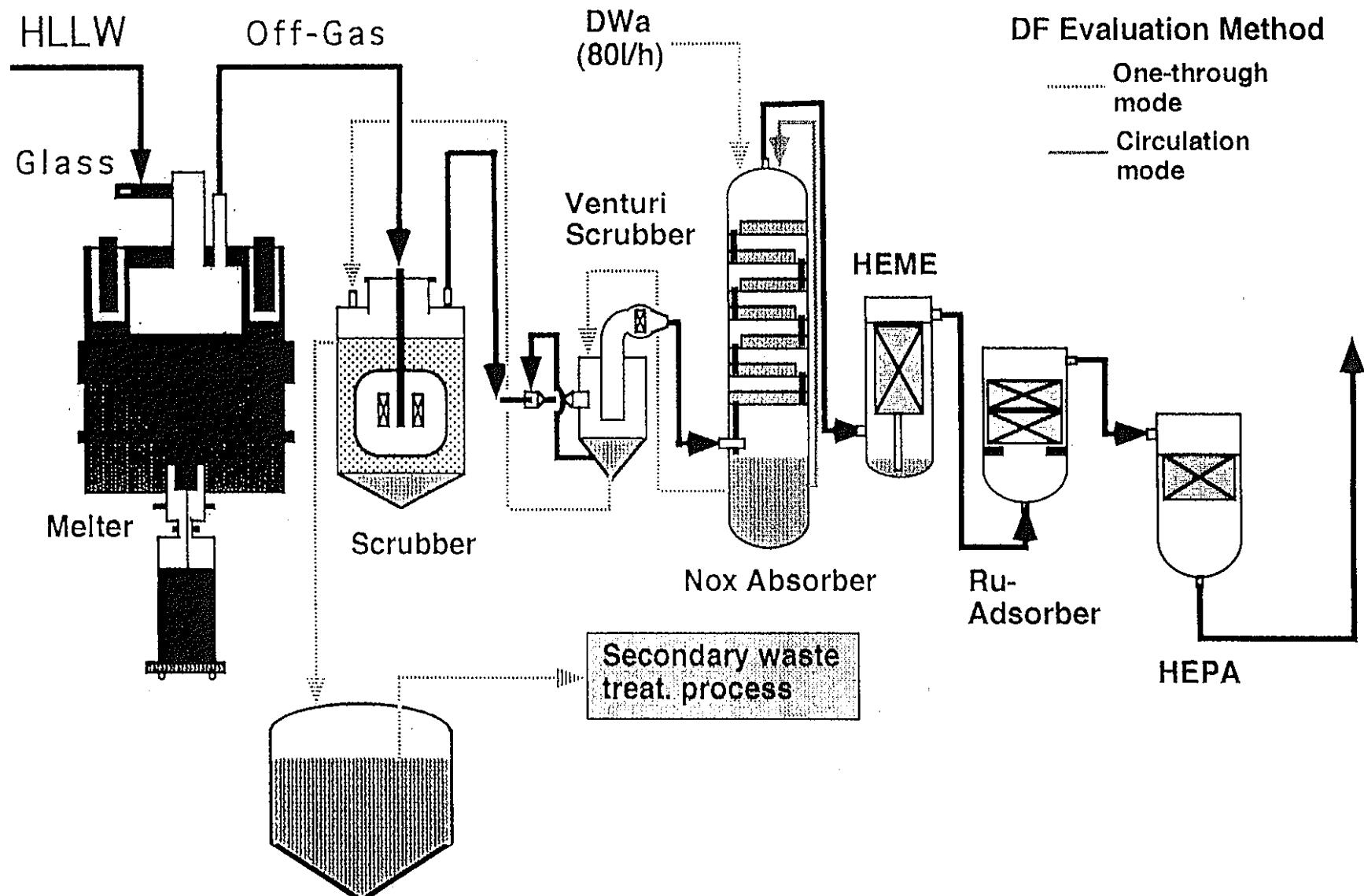
<u>SUPPLY (kW)[Result]</u>	
ELECTRODES	41.0
<u>DEMAND (kW)[Estim.]</u>	
COLD CAP	9.8
by N. Convection	5.5
by Radiation	4.3
GLASS MELTING	4.0
MASS FLOW	5.8
Steam	2.7
Gas	3.1
CONDUCTIVE	
HEAT LOSS	5.5
Plenum space	1.4
Melt pool	4.1
HEAT LOSS BY ELECTRODES COOLING	15.9
TOTAL	41.0

THE ENERGY BALANCE OF THE TVF MELTER(PC-92-1)  
DURING THE FEEDING OF BATCH NO. 6

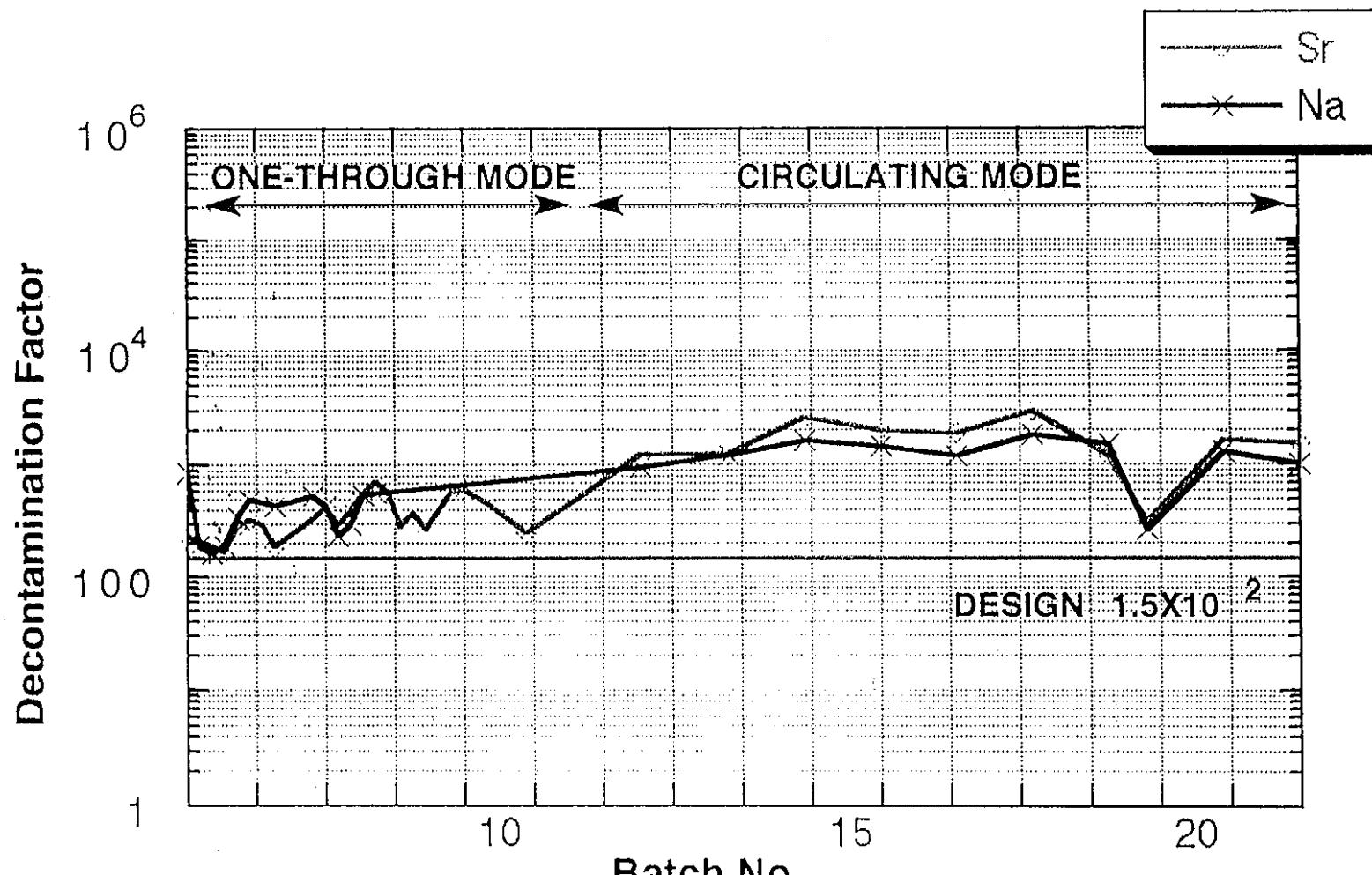


<u>SUPPLY (kW)[Result]</u>	
ELECTRODES	38.0
<hr/>	
<u>DEMAND (kW)[Estim.]</u>	
COLD CAP	9.8
by N. Convection	4.4
by Radiation	5.4
GLASS MELTING	4.0
MASS FLOW	3.8
Steam	1.7
Gas	2.1
CONDUCTIVE	
HEAT LOSS	5.1
Plenum space	0.9
Melt pool	4.2
HEAT LOSS BY ELECTRODES COOLING	15.3
TOTAL	38.0

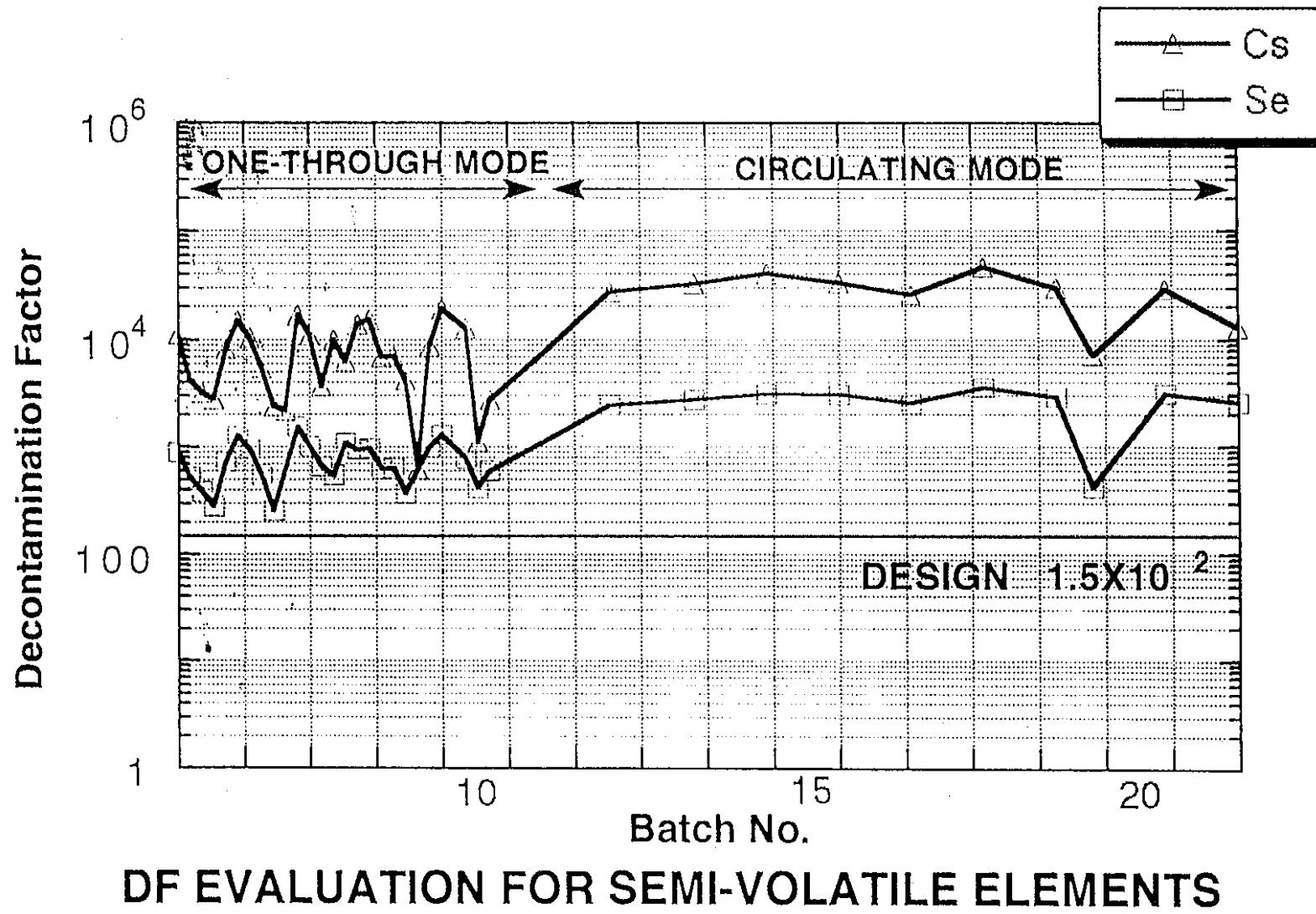
THE ENERGY BALANCE OF THE TVF MELTER(PC-92-1)  
DURING THE FEEDING OF BATCH NO. 12

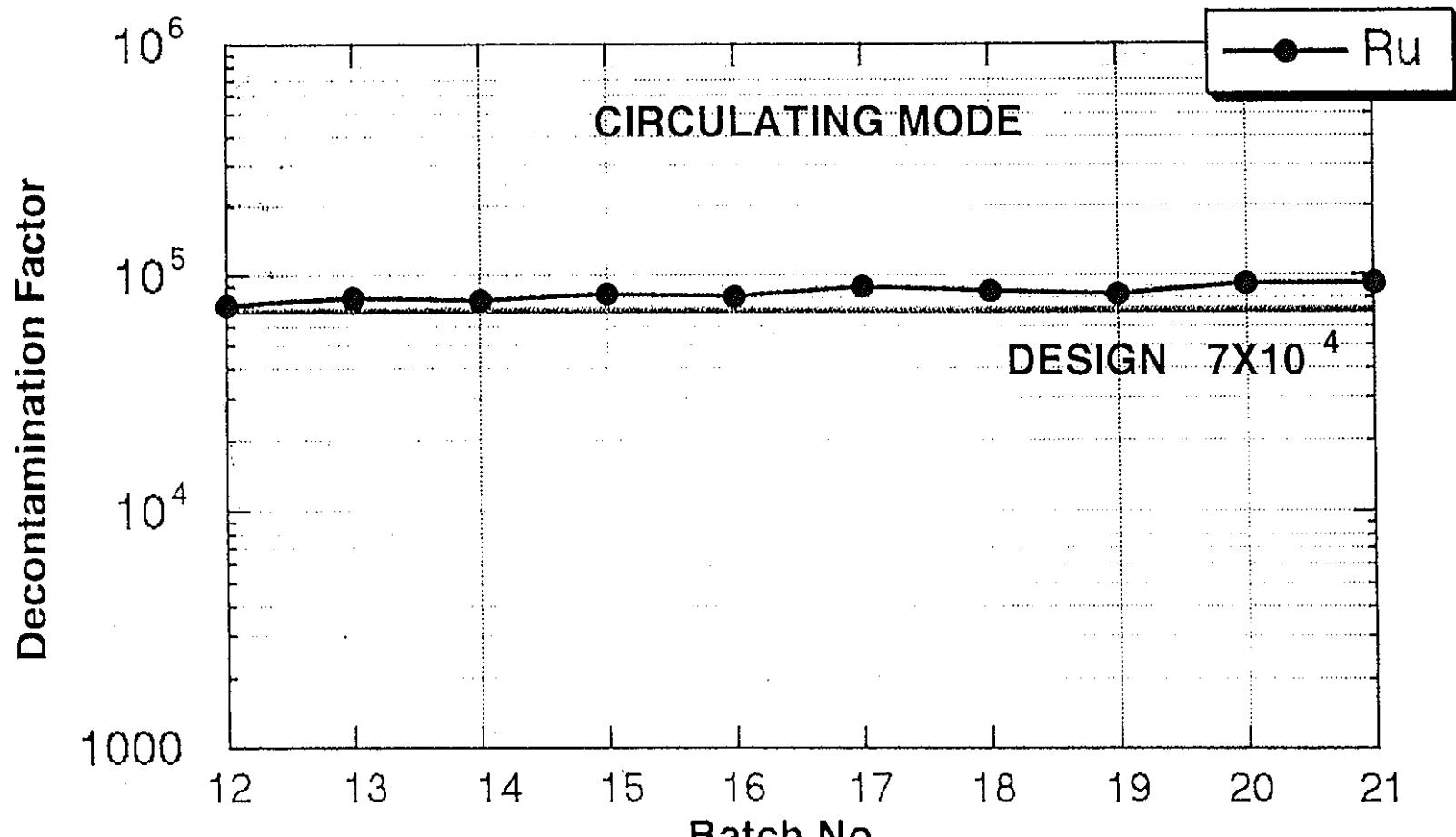


## MELTER OFF-GAS TREATMENT PROCESS



## DF EVALUATION FOR NON-VOLATILE ELEMENTS





DF EVALUATION FOR VOLATILE ELEMENT

# TEST ITEMS AND RESULTS IN COLD TEST RUN

TEST ITEM	Product Quality Control	Safety of Process	Melter Operation Mode	Function of Process Equipment
PROCESS	<ul style="list-style-type: none"> <li>• Pretreatment of HAW</li> <li>• Glass Feed</li> <li>• Glass Melting</li> <li>• Canister Handling</li> </ul>	<ul style="list-style-type: none"> <li>• All Equipment</li> <li>• Off-Gas Treatment</li> </ul>	<ul style="list-style-type: none"> <li>• Glass Melter</li> <li>• Glass Pouring</li> </ul>	<ul style="list-style-type: none"> <li>• All Equipment especially Glass Melter Evaporator</li> </ul>
CONFIRMATION ITEM	<ul style="list-style-type: none"> <li>• Quantitative feed of HAW and glass</li> <li>• Estimation method of HAW concentration</li> <li>• Accuracy of Quality control (glass composition)</li> <li>• Other operating condition</li> </ul>	<ul style="list-style-type: none"> <li>• Decontamination factor in Melter off-gas treat. process</li> <li>• Safety function and control method of equipment</li> </ul>	<ul style="list-style-type: none"> <li>• Melter operation (control) method correspond to noble metal</li> <li>• Glass pouring mode (bottom heating mode)</li> <li>• Glass pouring control</li> </ul>	<ul style="list-style-type: none"> <li>• Function of equipment in change of operating condition</li> <li>• Synthetic function of facility</li> </ul>
TEST RESULT	<ul style="list-style-type: none"> <li>• Quantitative feed has adequate accuracy to satisfy product quality</li> <li>• HAW concentration can be estimated with adequate accuracy</li> <li>• Waste content in product can be controlled in limit with enough accuracy</li> </ul>	<ul style="list-style-type: none"> <li>• DF can satisfy design in both Melter off-gas treat. process and evaporator</li> <li>• Equipment has safety function as expected in design</li> </ul>	<ul style="list-style-type: none"> <li>• Possible to make stable melter operation</li> <li>• Possible to treat noble metal by using low bottom temperature mode</li> <li>• Possible to start stable pouring by heating mode and to control pouring</li> </ul>	<ul style="list-style-type: none"> <li>• Confirm effect of change in operating condition on melter, evaporator</li> <li>• Confirm stable operation of TVF process</li> </ul>

2.3 Three dimensional thermal-hydraulic analyses by physical  
and mathematical modelings

# **Three dimensional thermal-hydraulic analyses by physical and mathematical modelings**

- 65 -

Prepared for  
The 12th Annual KfK-PNC Meeting on  
High-Level Waste Management

by AYAME, Y.

December 7-9, 1993

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Power Reactor and Nuclear Fuel  
Development Corporation



## Background of study

The Relation between Requirement for Design of the Melter and an Item of Thermal-Hydraulic Analysis in the Melter

### Requirement for Design of Melter

- Containment of Molten Glass in the Melter Structure
- Corrosion Resistance of the Electrode and the Refractory Structure
- Melting rate of the Melter
- Quality of the Glass Product

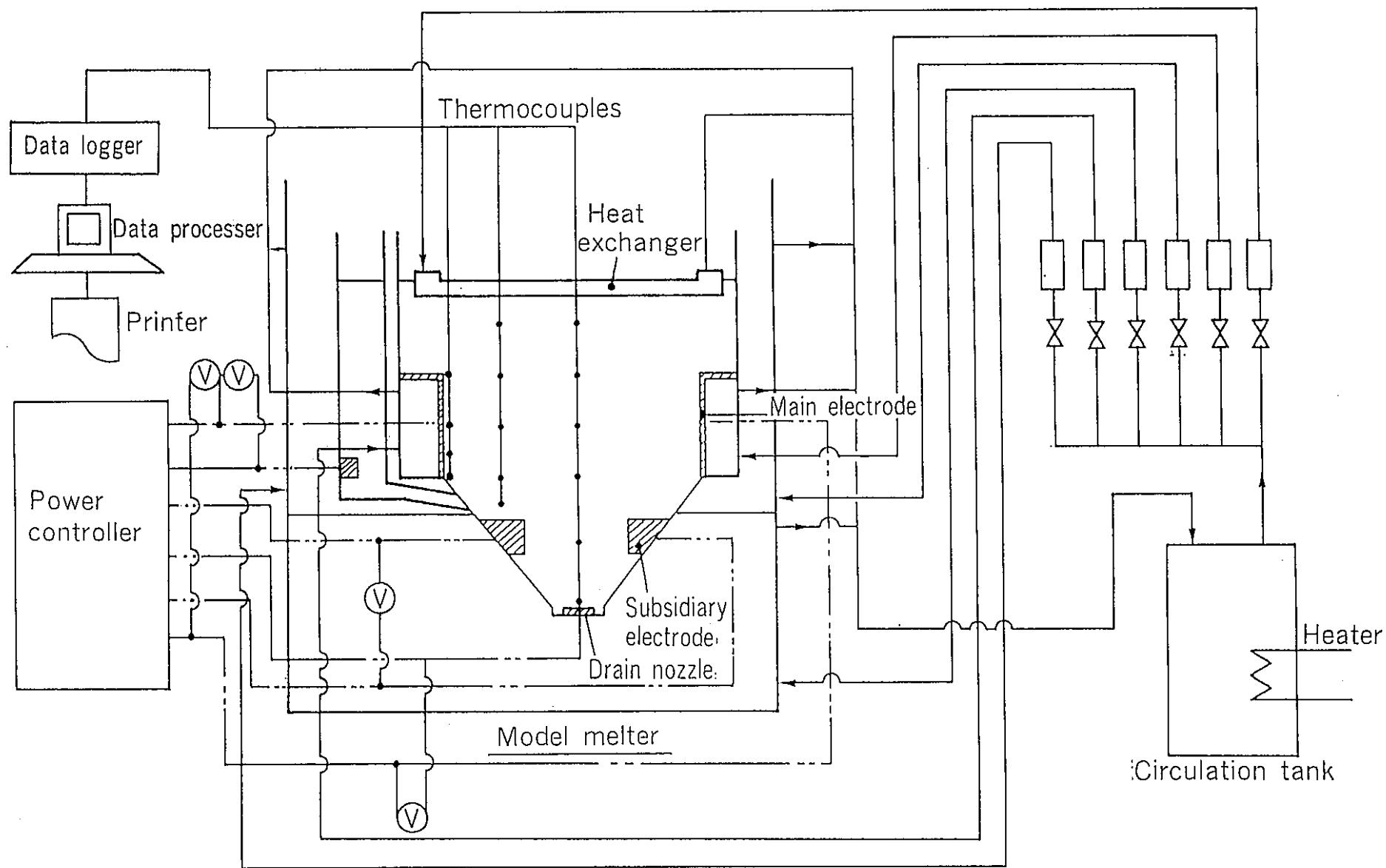
### Item of Thermal-Hydraulic Analyses

- Temperature Distribution in the Melting Cavity
- Temperature Gradient in the Electrode and the Refractory Structure
- Electrical Aspect in the Melting Cavity
- Flow Pattern in the Melting Cavity

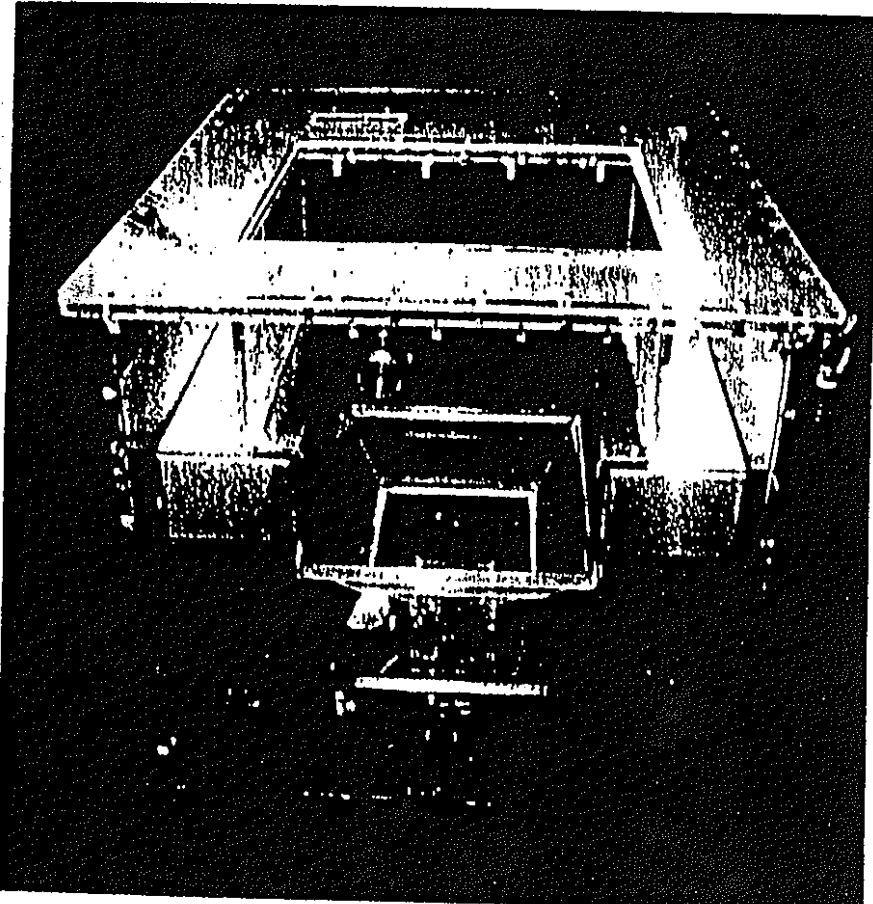
## Scope of study

The Physical and mathematical modeling is useful means to obtain the detailed information on thermal-hydraulic behavior in the melter.

- Validation of temperature and flow pattern in the melter by means of physical and mathematical modeling
- Validation of mathematical modeling by physical modeling



Physical model system



## Specification of physical model

Dimension, internal : L267×W270×H350mm  
external : L515×W425×H410mm

Floor slope :  $45^\circ$  and  $54.5^\circ$

Material, Wall : Polymethylmethacrylate  
(PMMA)

electrode : copper plate

Fluid : Glycerin with 7.5 wt% LiCl

Analogy : one-third scale of TVF melter  
equivalent modified Rayleigh  
number

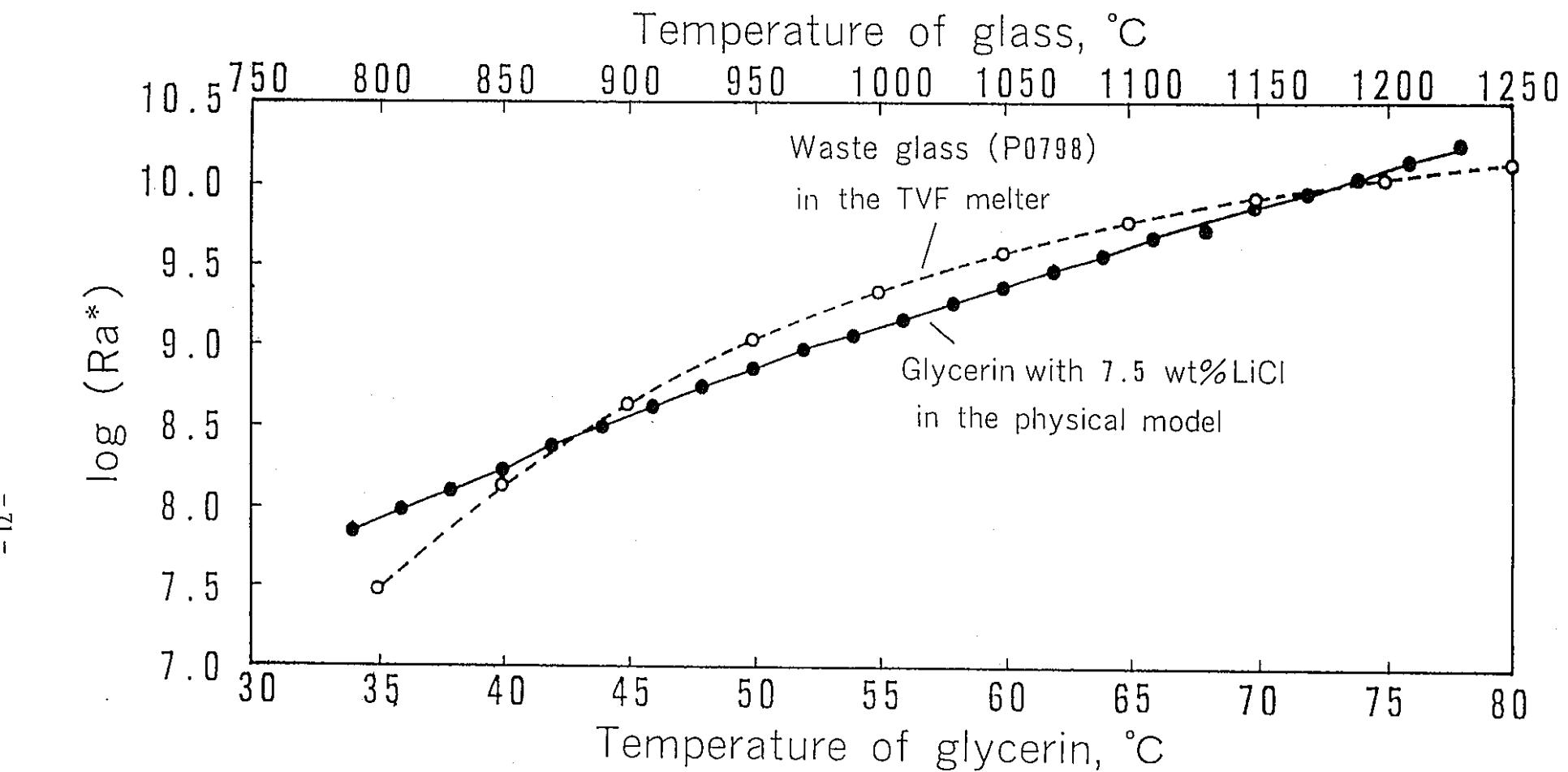
## Modified Rayleigh Number : $Ra^*$

$$Ra^* = g \beta \sigma V_0 L^3 / \alpha \nu \lambda$$

( = Gr Pr )

-70-

- Gr : Grashof Number , Pr : Prandtl Number ,  
g : Gravitational Constant , L : Length ,  
 $V_0$  : Electrode Voltage ,  $\alpha$  : Thermal Diffusivity ,  
 $\beta$  : Thermal Coefficient of Volumetric Expansion ,  
 $\sigma$  : Electrical Conductivity of Fluid,  
 $\lambda$  : Thermal Conductivity ,  $\nu$  : Kinematic Viscosity .



Comparison of modified Rayleigh number,  $Ra^*$  between the physical model and the TVF melter  
(P0798 : PNC's glass code)

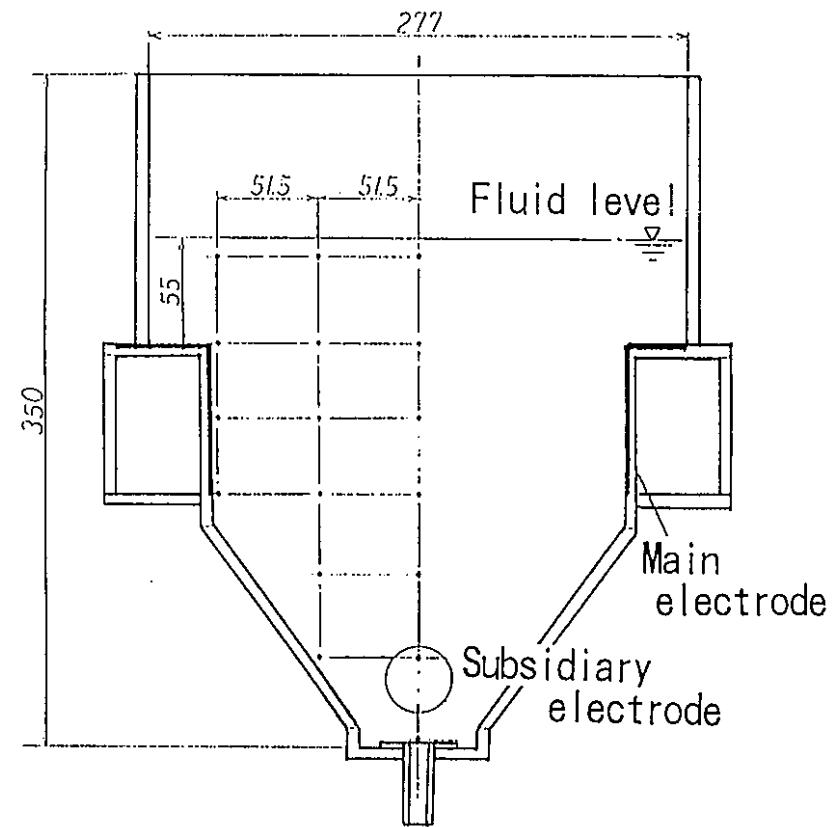
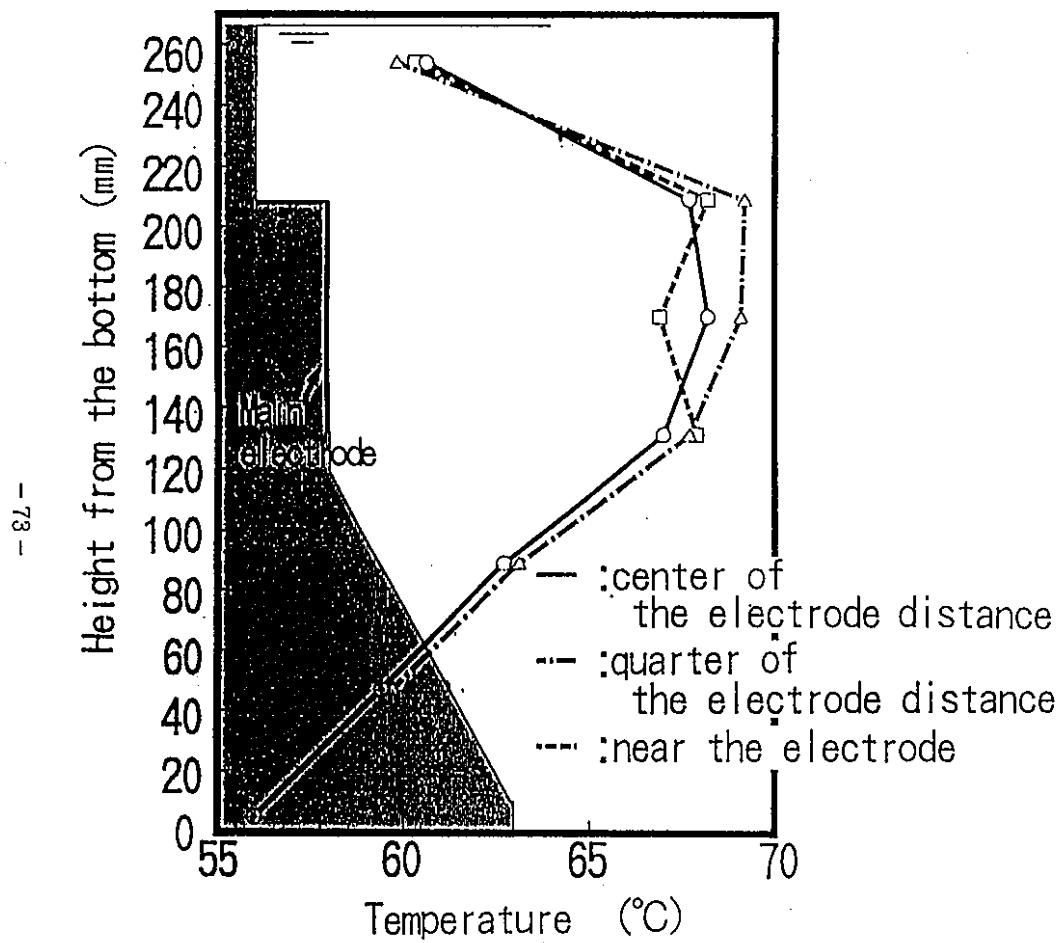
# Experiment Condition of Physical Modeling

## Joule heating

Electrodes	Electric power (W)		
	Case 1	Case 2	Case 3
Main electrodes	74	86	96
Subsidiary electrodes	3	3	3
Drain nozzle and main	3	3	3

## Water bath temperature

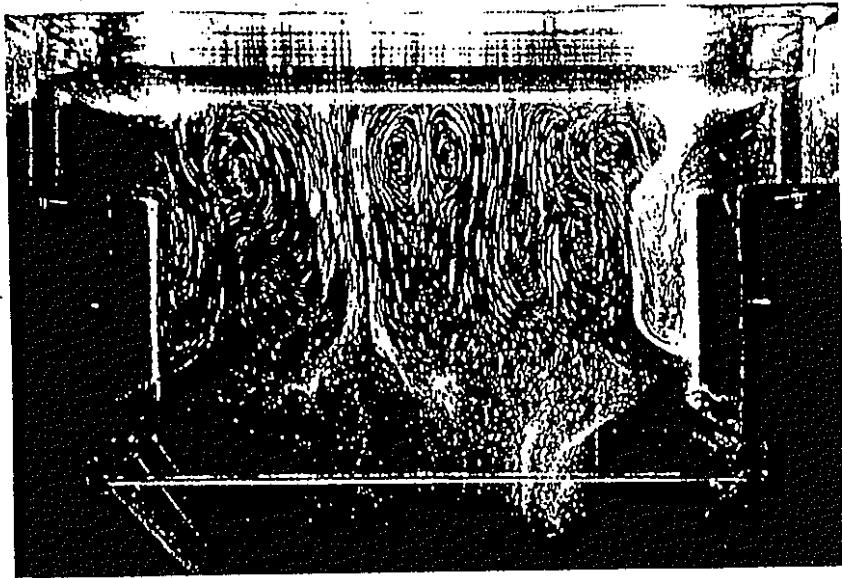
Water bath	Water temperature (°C)
Top water jacket	62
Side wall	61
Bottom	52
Main electrode	61



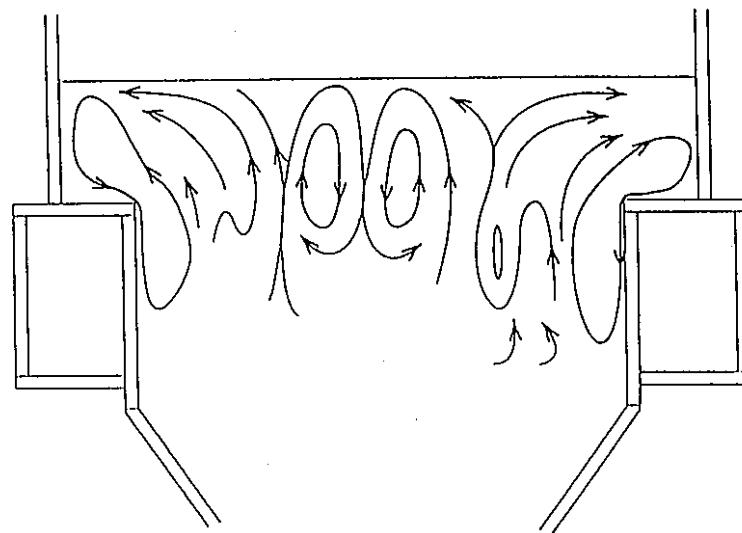
Measuring points of temperature and electric voltage

# Vertical temperature profile (Case 1)

-74-



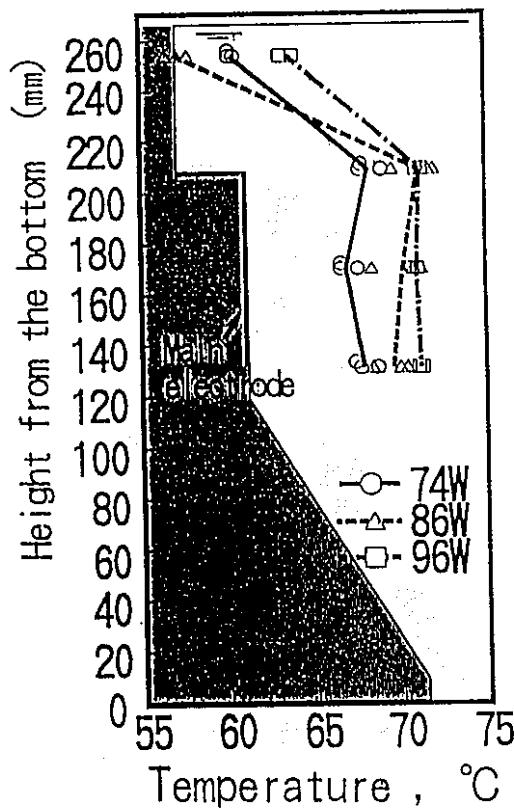
(a) Photograph of traces  
(Shutter speed 20second)



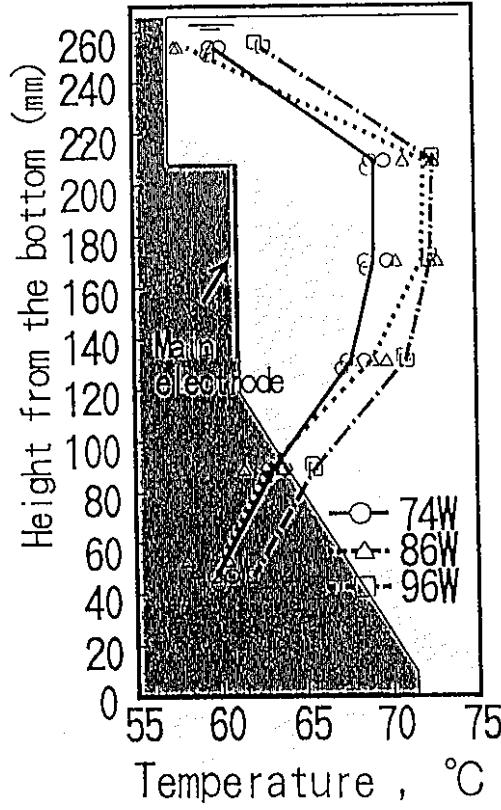
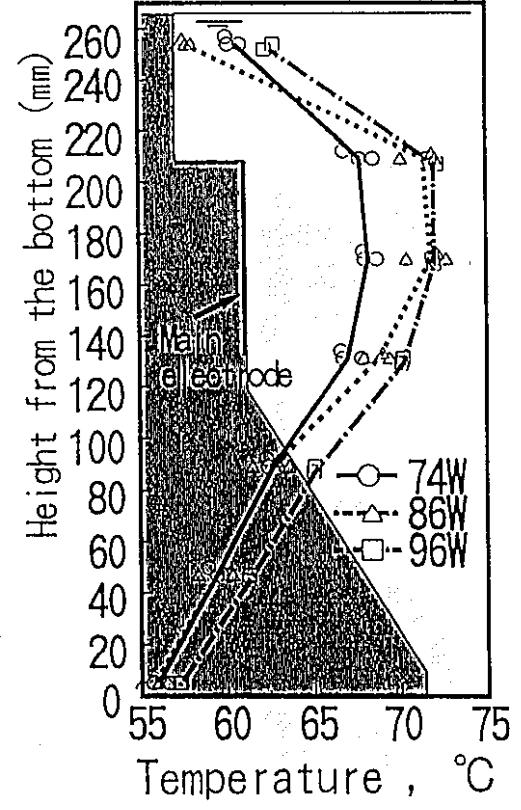
(b) Illustrated patterns

Flow patterns observed in physical model  
for Case 1 (Main electrode power 74W)

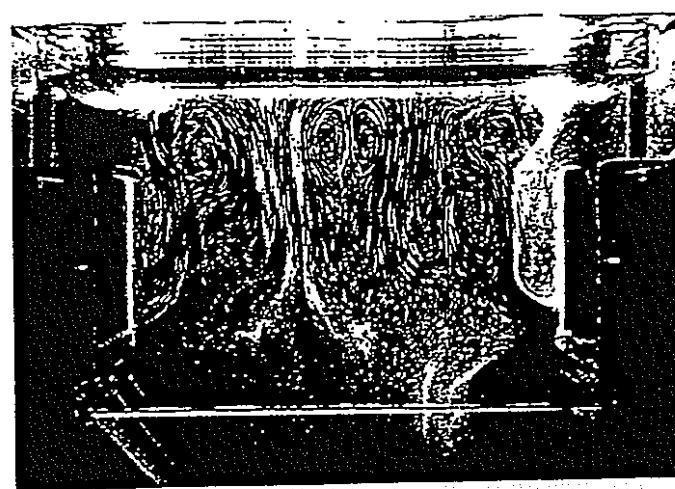
- 92 -



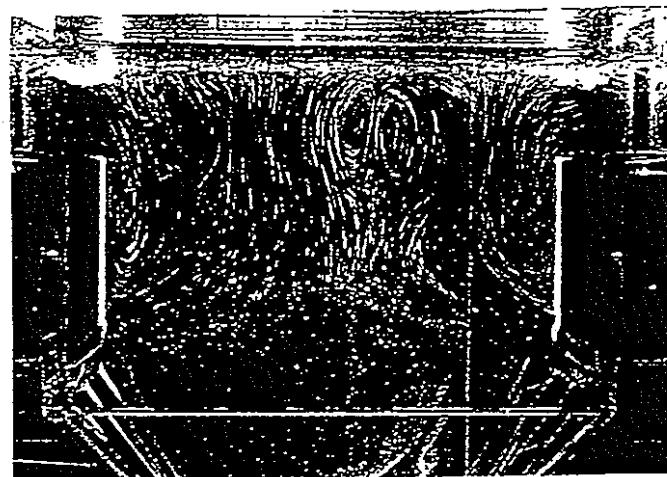
near the electrode

quarter of the electrode  
distancecenter of the electrode  
distance

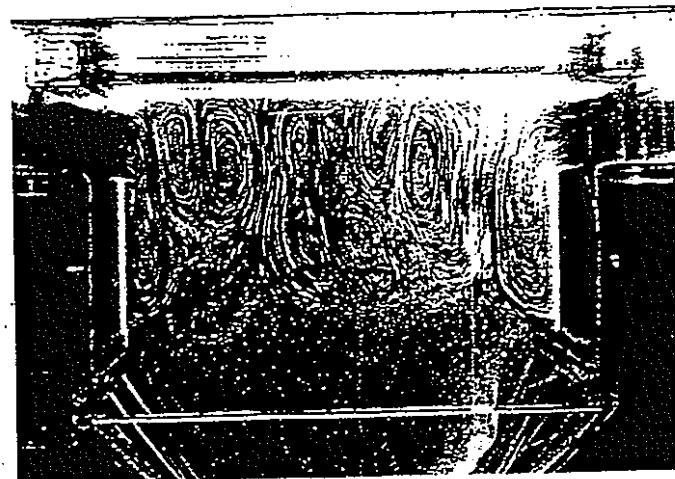
## Experiment results of vertical temperature profiles (Comparison of main electrode power)



(a) Main electrode power 74W

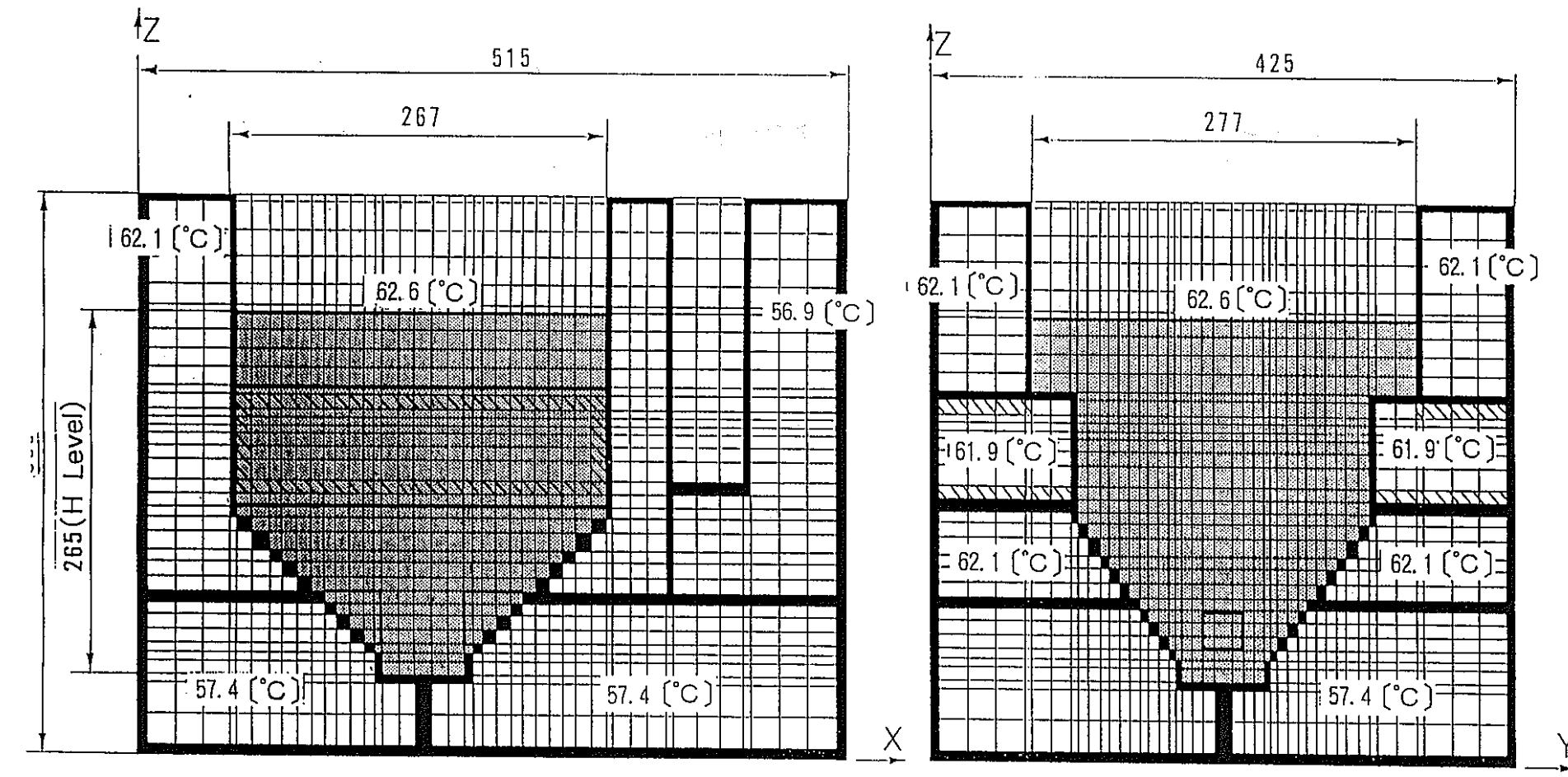


(b) Main electrode power 86W

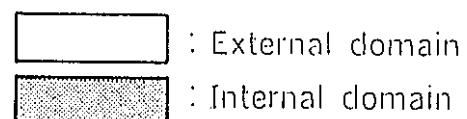


(c) Main electrode power 96W

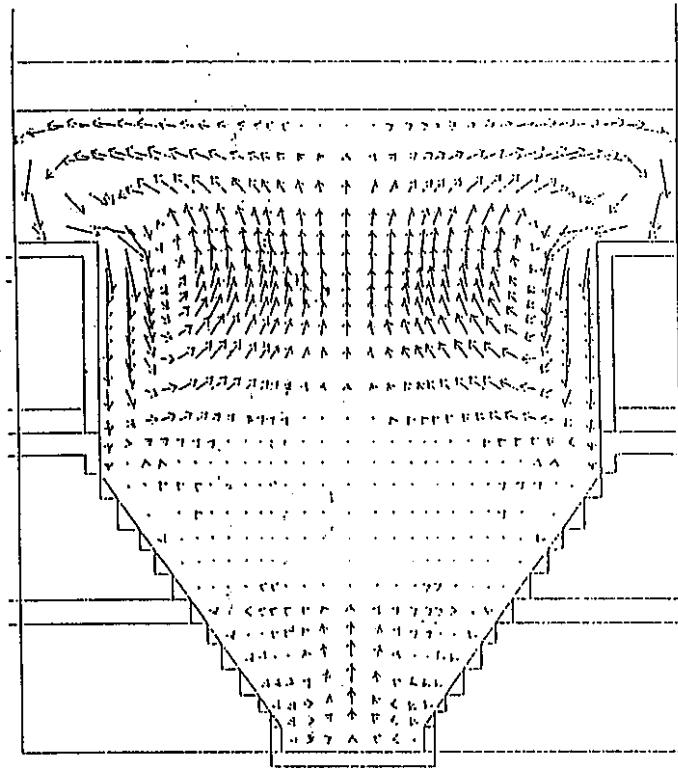
Flow patterns observed in physical model



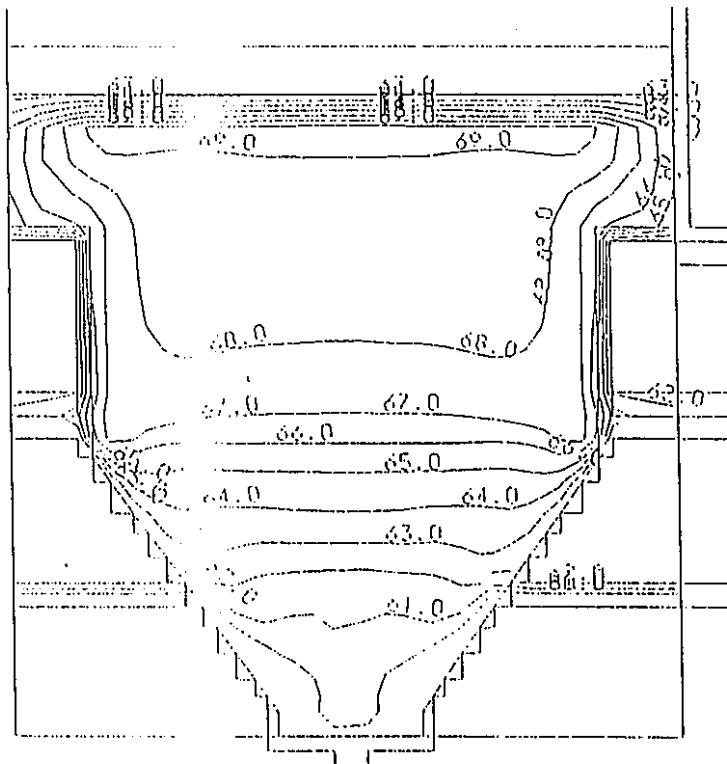
Mesh geometry of physical model for computation  
(Mesh size :  $46 \times 46 \times 38$  with  $28 \times 36 \times 28$  Internal domain)



-78-



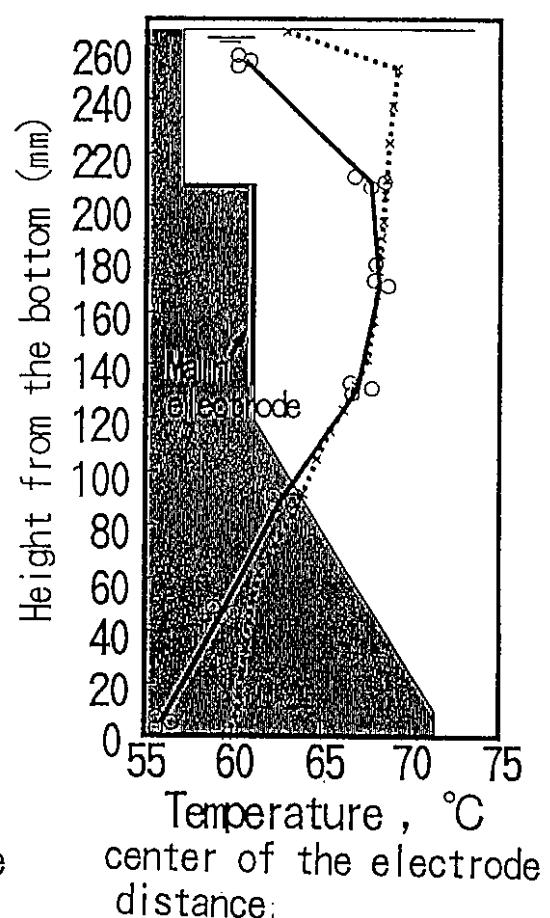
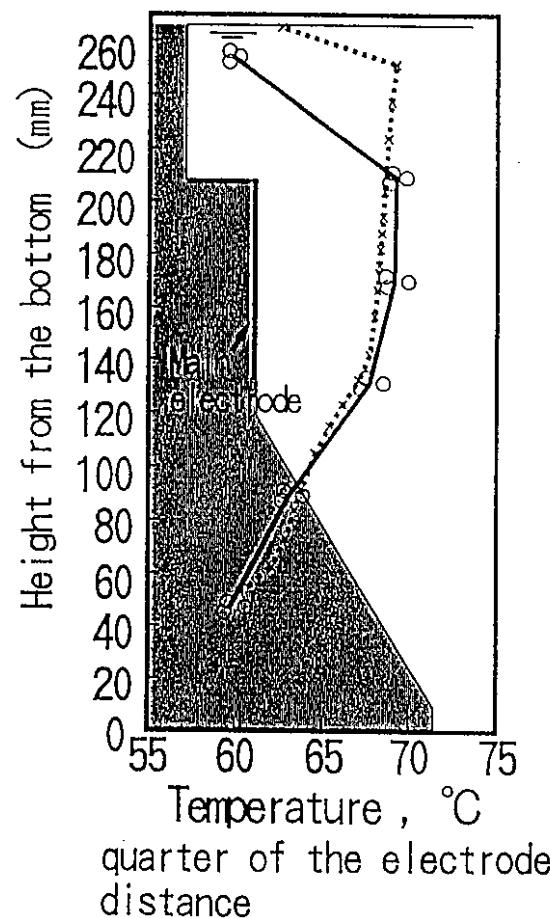
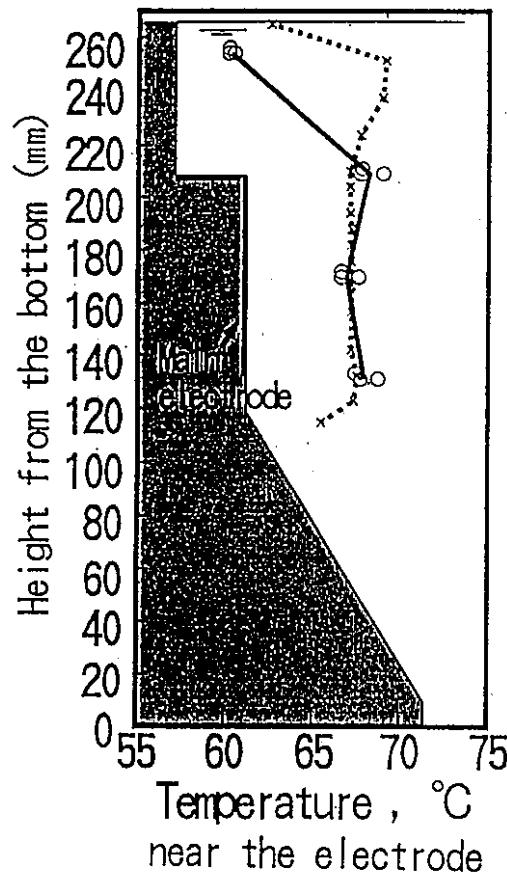
(a) Flow pattern



(b) Temperature distribution, °C

Computational result by mathematical model

- 6L -

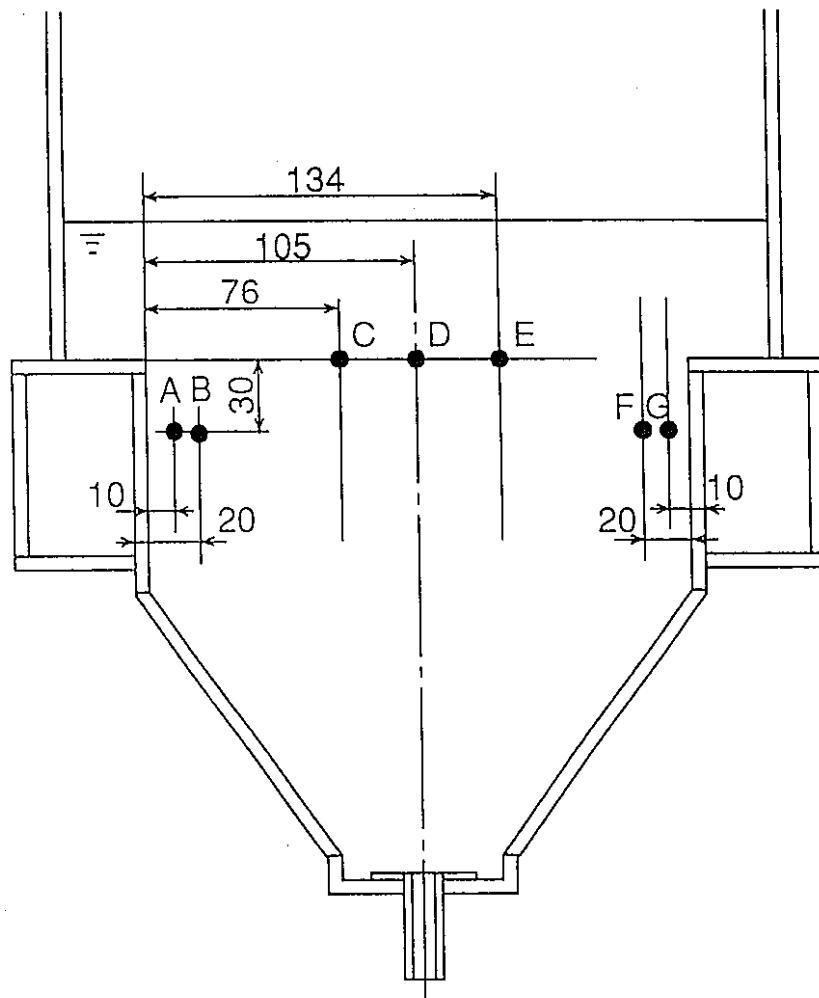


Comparison of vertical temperature profile between computation and experiment

—○— Experiment  
---×--- Computation

Comparison of vertical velocity components  
between computation and experiment , mm/s

Measuring points	experiment		computation
	velocity	standard deviation	
A	-0.32	0.060	-0.56
B	0.40	0.063	-0.03
C	0.67	0.146	0.21
D	-0.50	0.070	0.20
E	0.71	0.120	0.21
F	0.40	0.066	-0.03
G	-0.39	0.088	-0.56



Measuring points of velocity  
in physical model

# Conclusion

- Influence which the electric power exerts the flow patterns and temperatuer profile was clearly by physical modeling.
- The flow patterns and temperature behavior was proved by physical and mathematical modeling.
- The chief flow patterns agreed between the experiment and computational result.
- The temperatuer profile agreed qualitatively and quantitatively between the experoment and computatinal results.

2.4 Inspection technique based on LASER for melter and  
Dismantling of spent melter

# **Inspection technique based on LASER for melter and Dismantling of spent melter**

Prepared for  
The 12th Annual KfK-PNC Meeting  
on  
Cooperation in High-Level Waste Management

by KOBAYASHI, H.

December 7-9, 1993

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Power Reactor and Nuclear Fuel  
Development Corporation



## Development of the In-Melter Inspection System

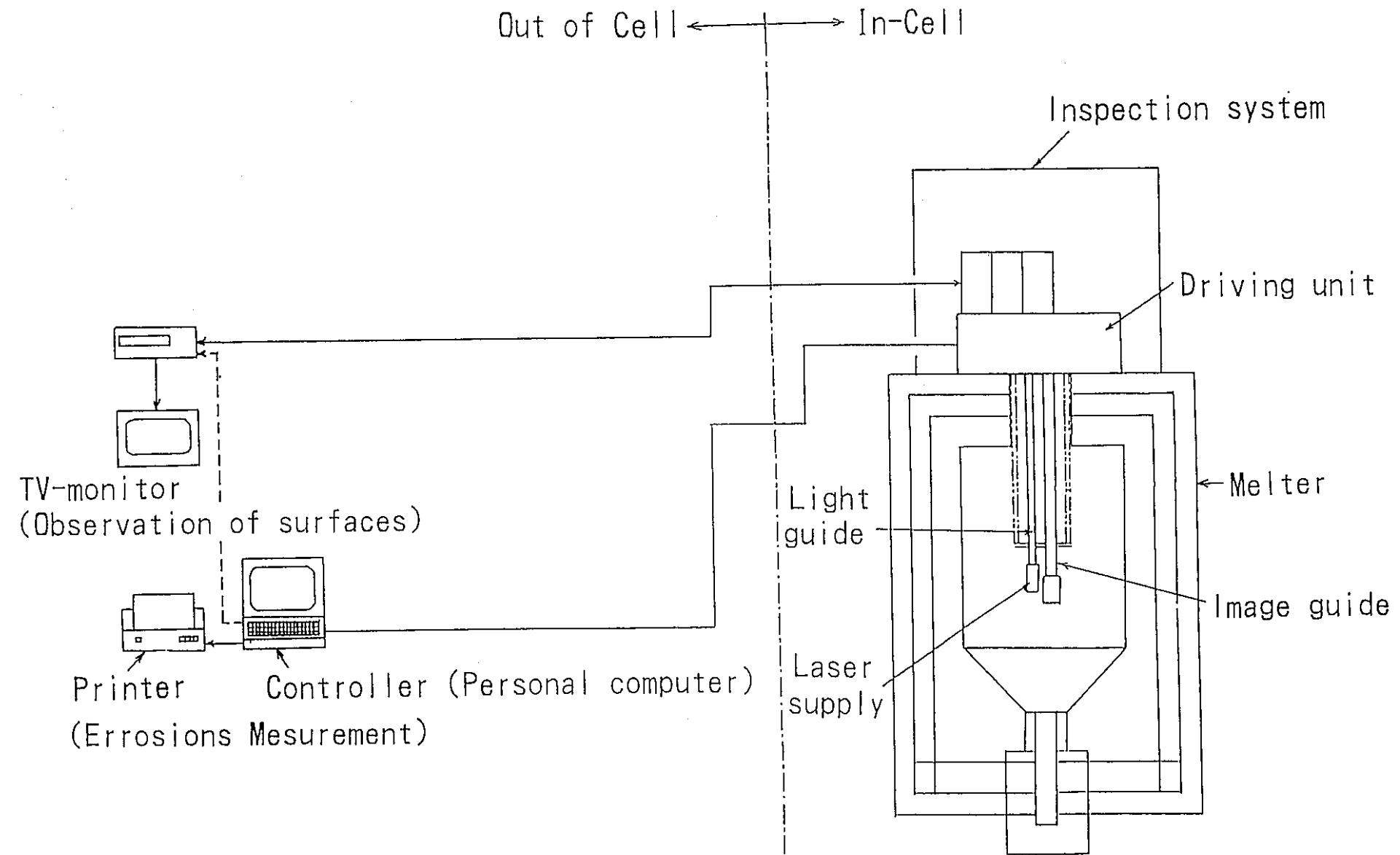
### Purpose

Check durability of the melter

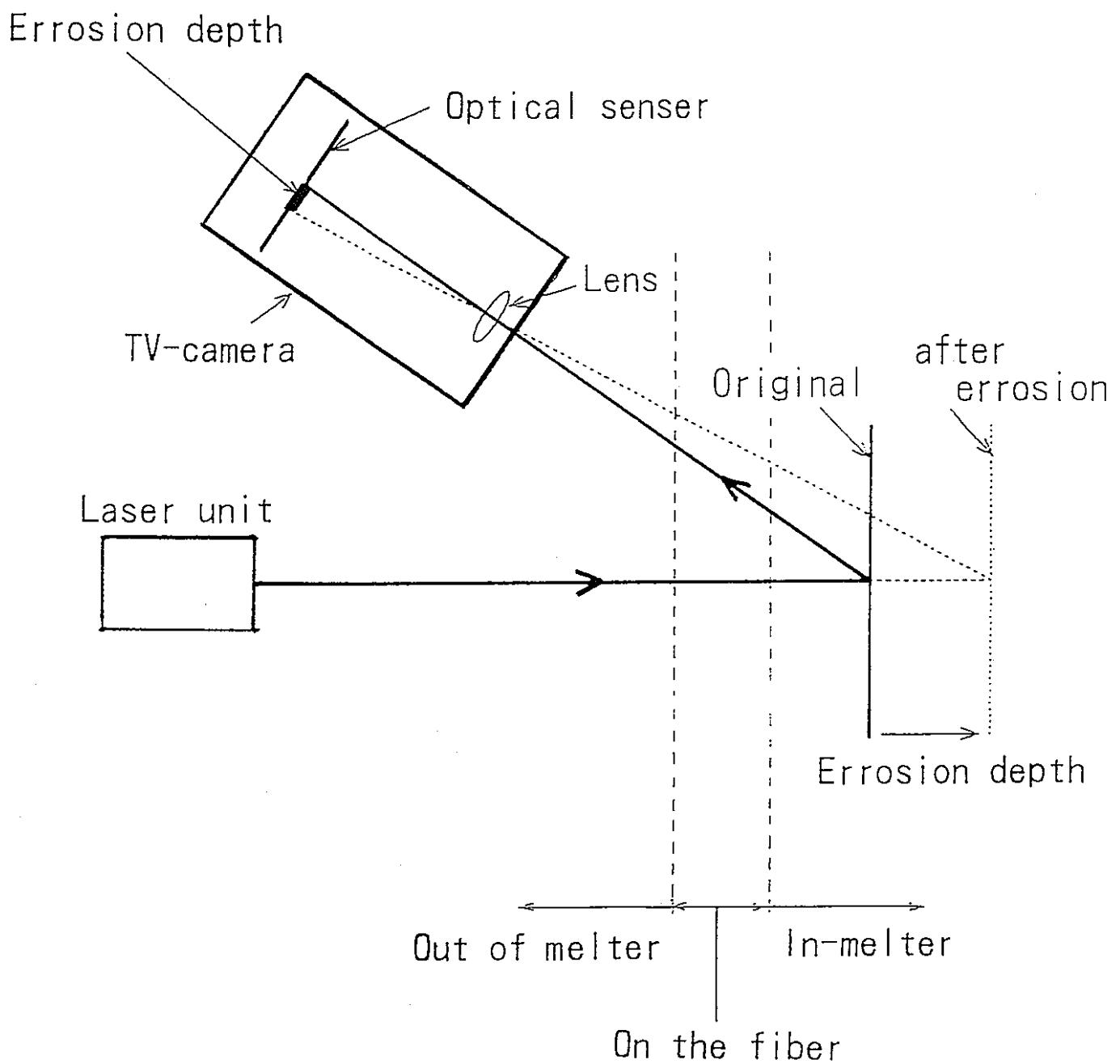
- Observation of Surfaces      (Electrode)
- Erosion Measurement      (Refractory)

### Radiation in melter

- $10^6 \text{ R/h}$  ( $258 \text{ C/kg} \cdot \text{h}$ )

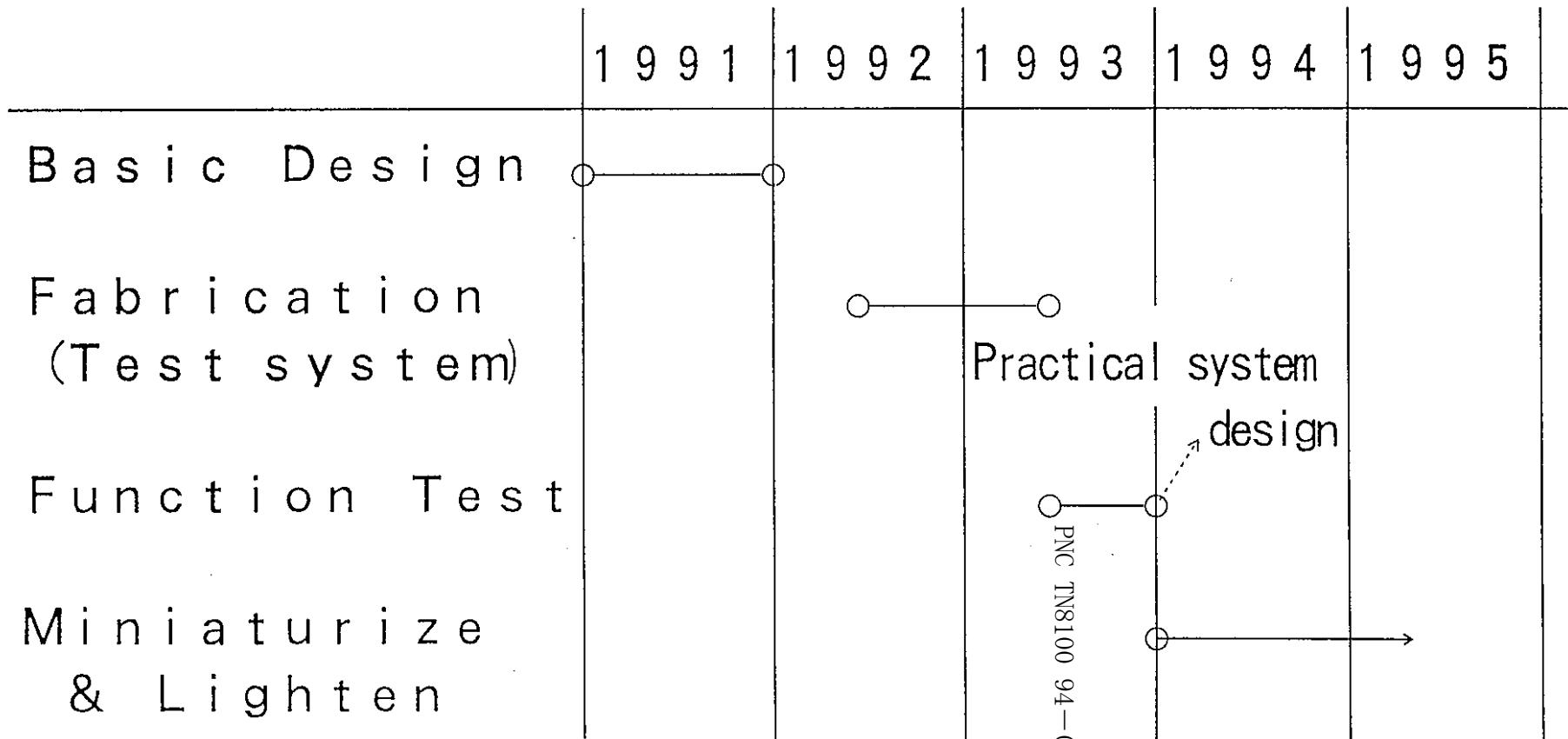


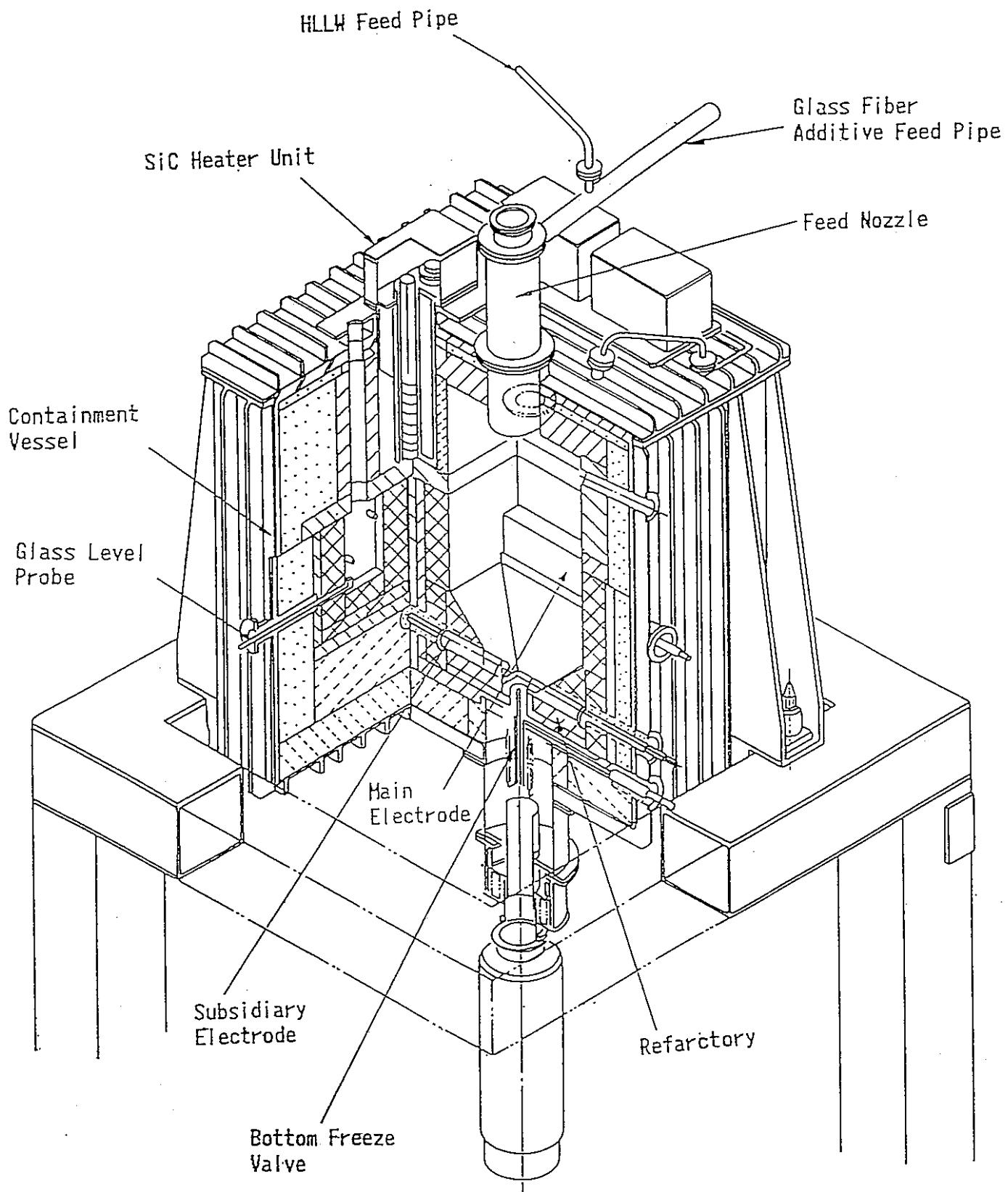
SCHEMATIC OF INSPECTION SYSTEM



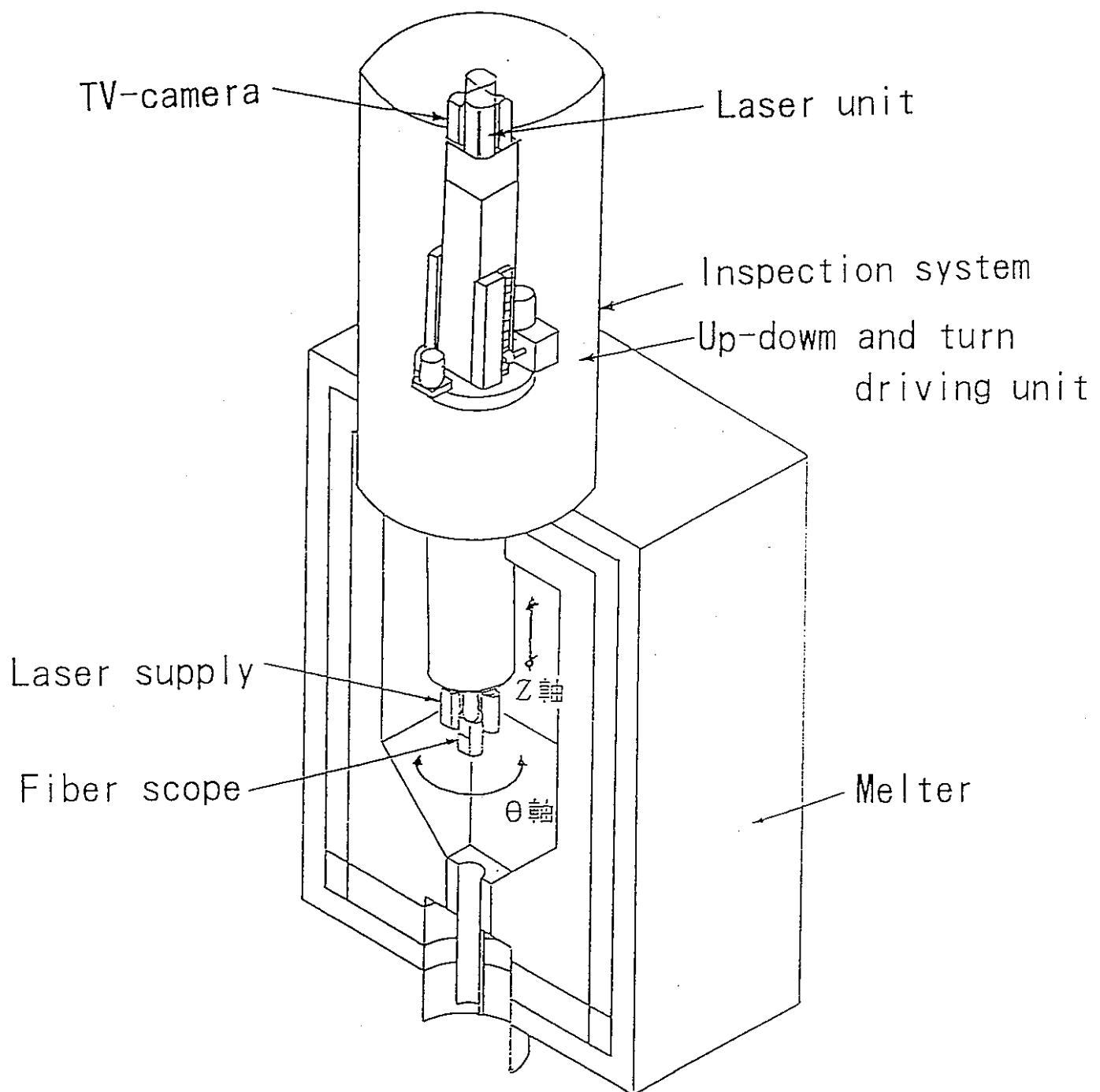
## DISTANCE MEASUREMENT BY TRIANGULATION

## Development plan

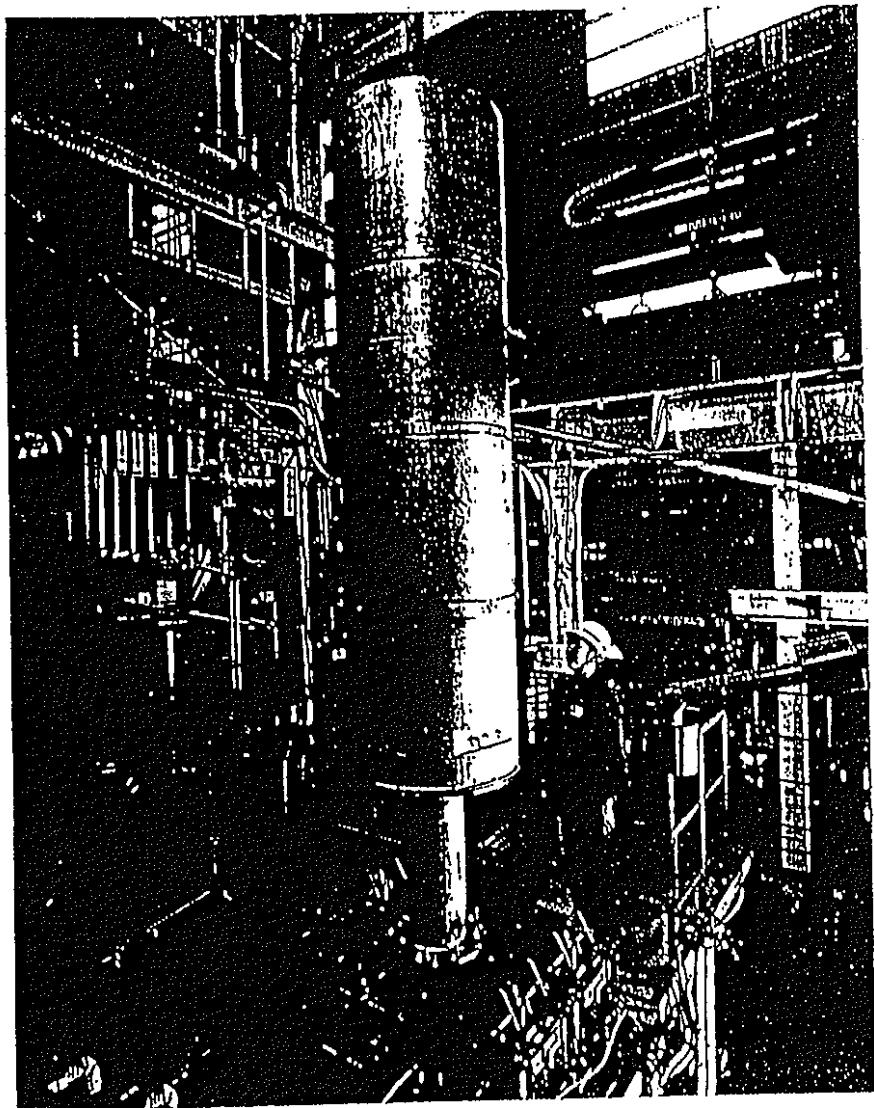




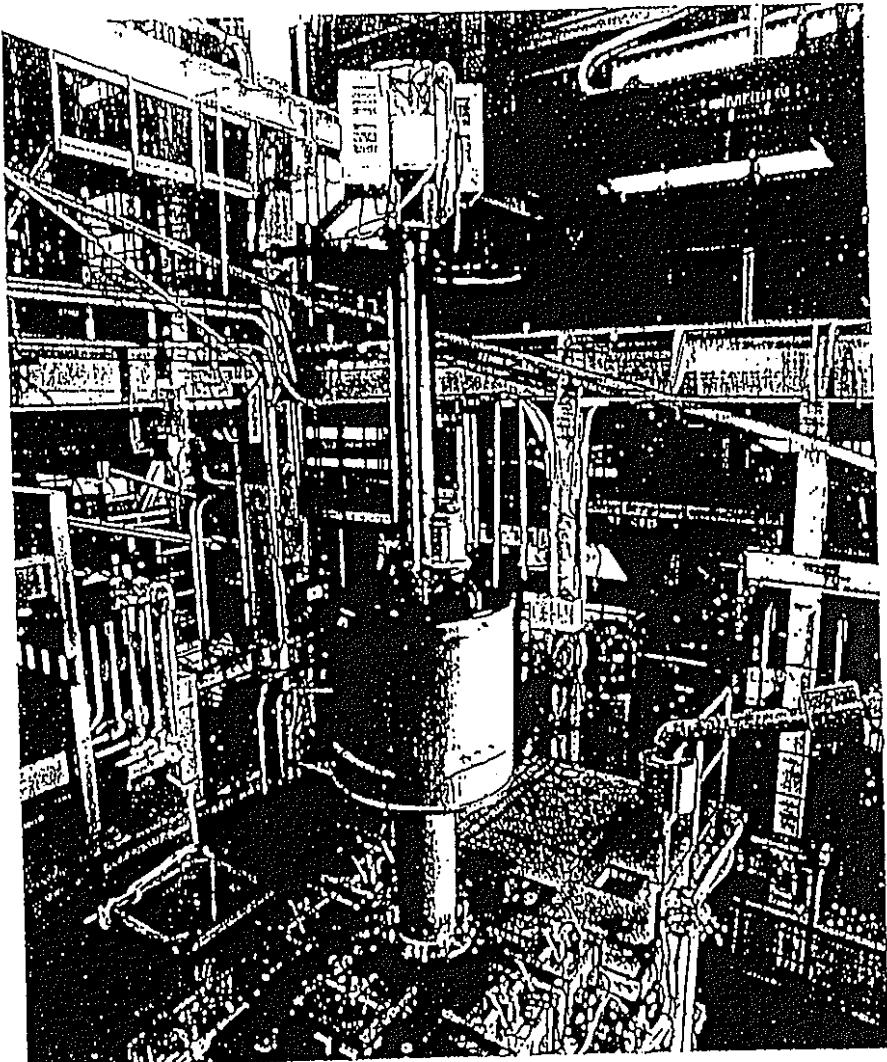
## Structure of Melter



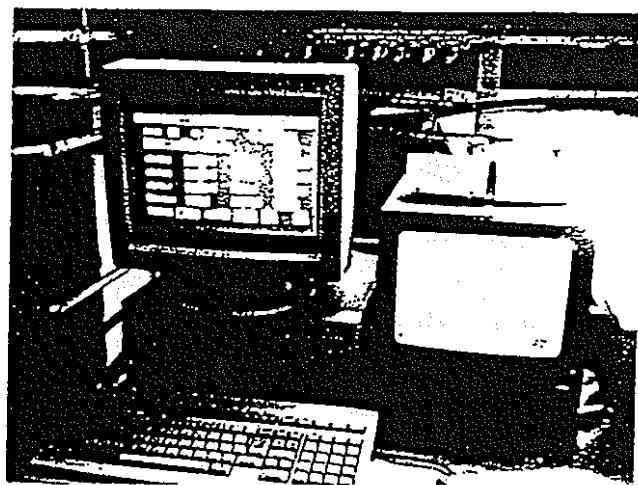
## CONCEPT OF THE IN-MELTER INSPECTION SYSTEM



Inspection Instrument  
on the melter



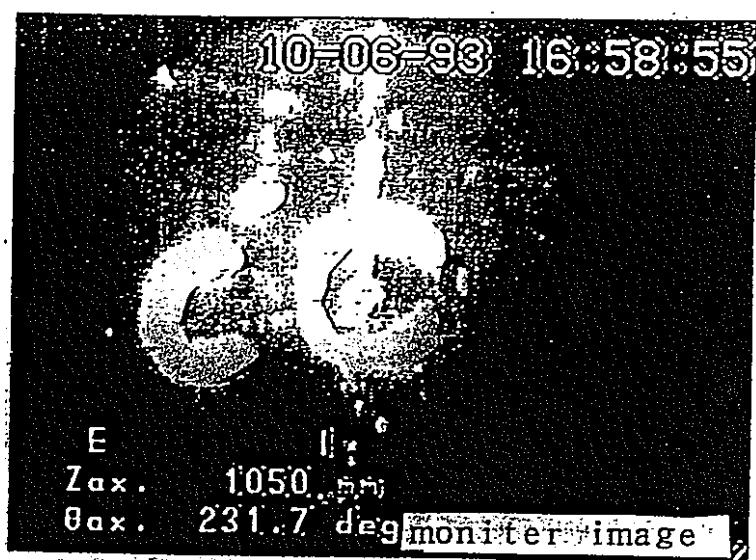
Inside of The Instrument



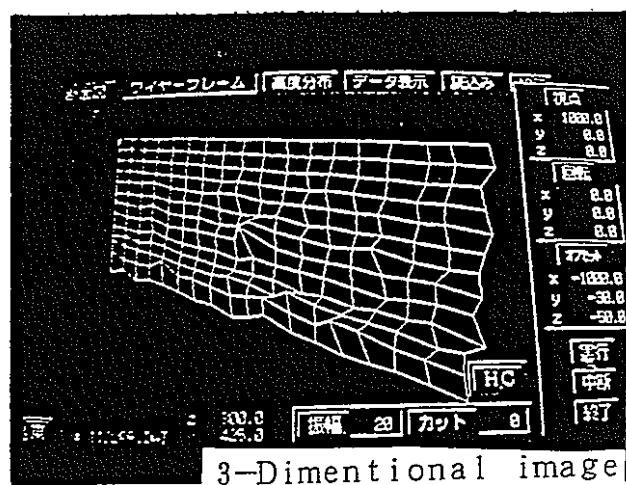
Control system



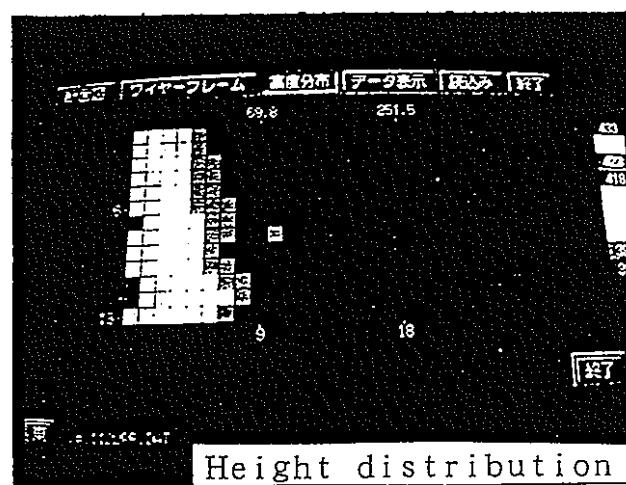
Clear picture



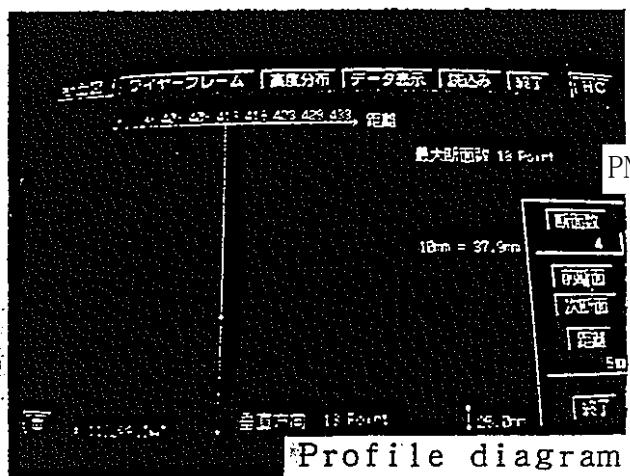
DATA DISPLAY with THE INSPECTION SYSTEM



3-Dimentional image



Height distribution



Profile diagram

DATA DISPLAY with THE INSPECTION SYSTEM

## Melter Dismantling Test

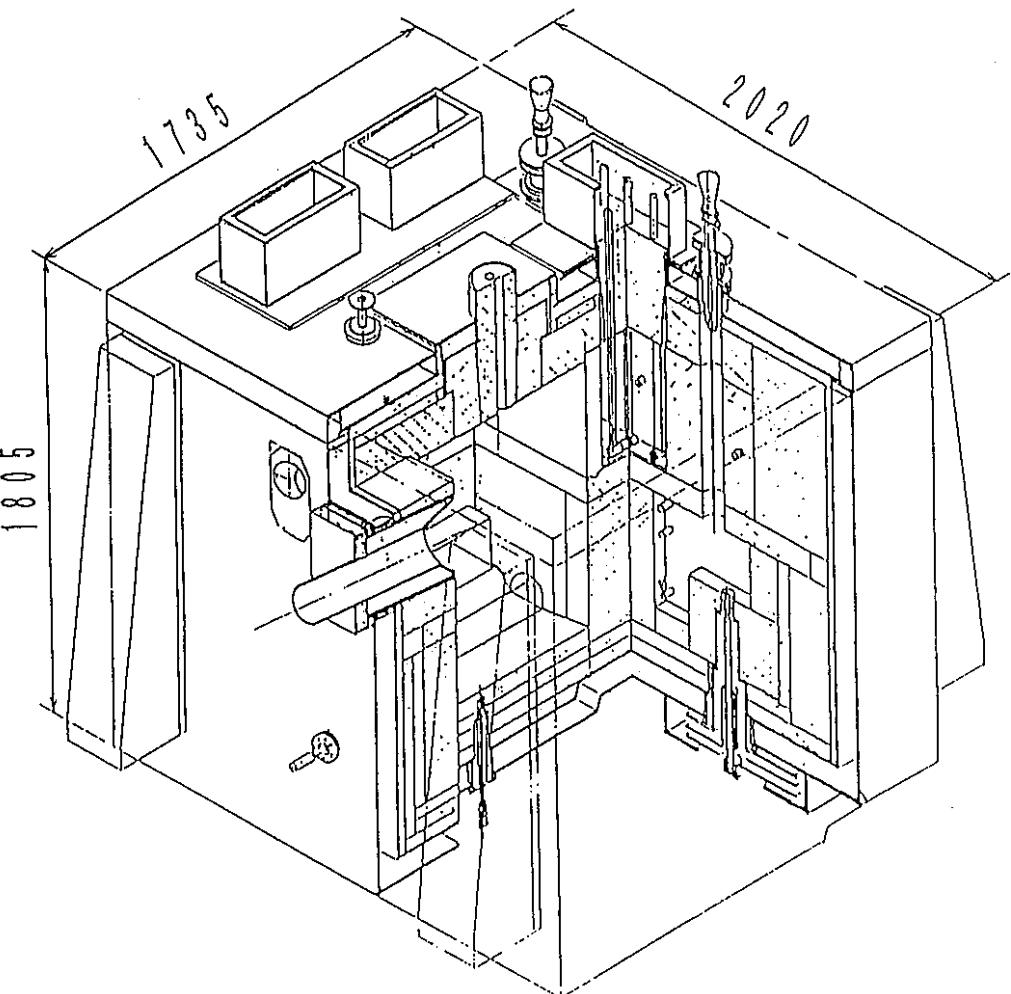
### Purpose

- Estimate of dismantling process
- Ability test of dismantling tools

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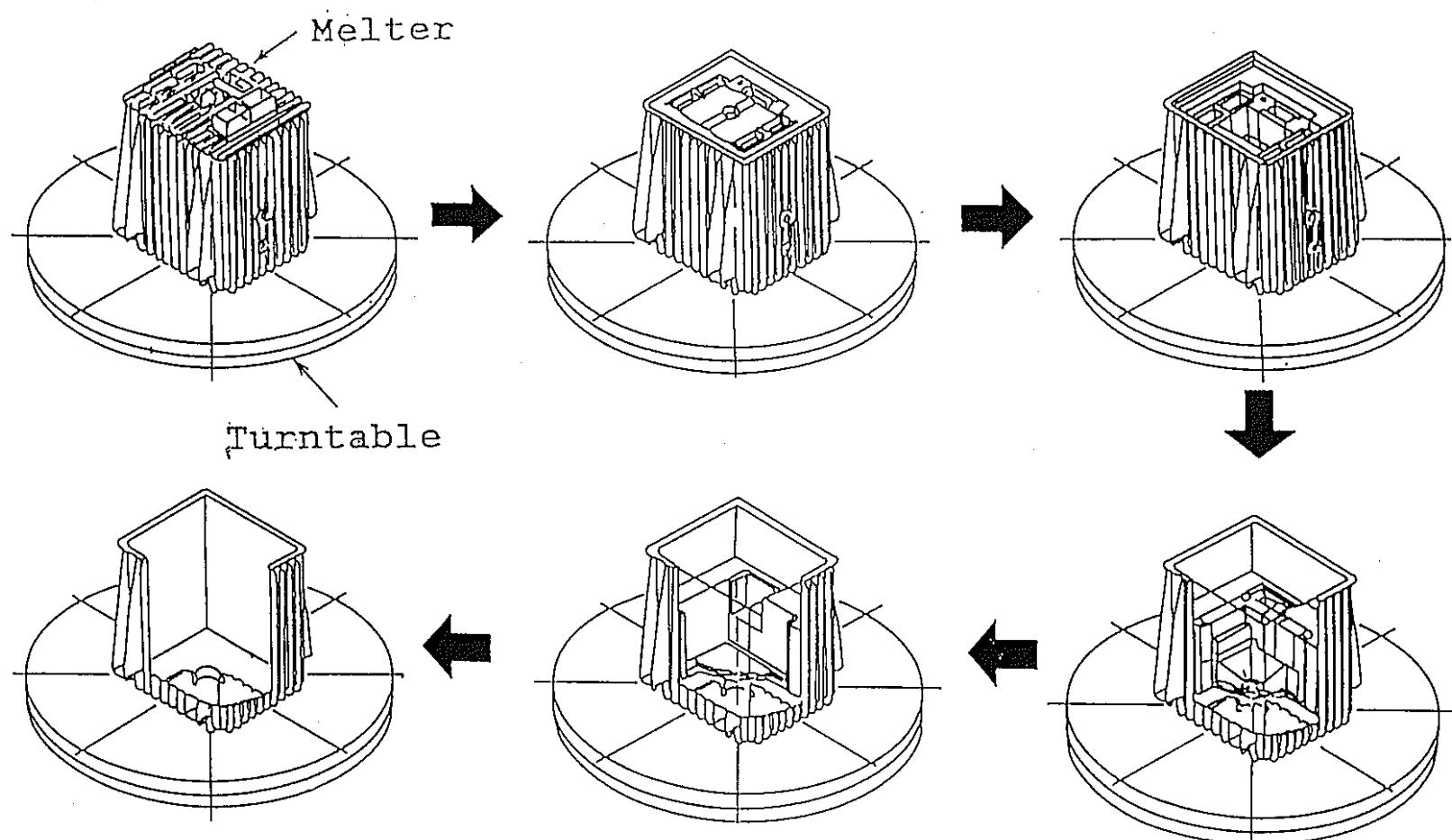
### An object

- Mock-up 2 melter



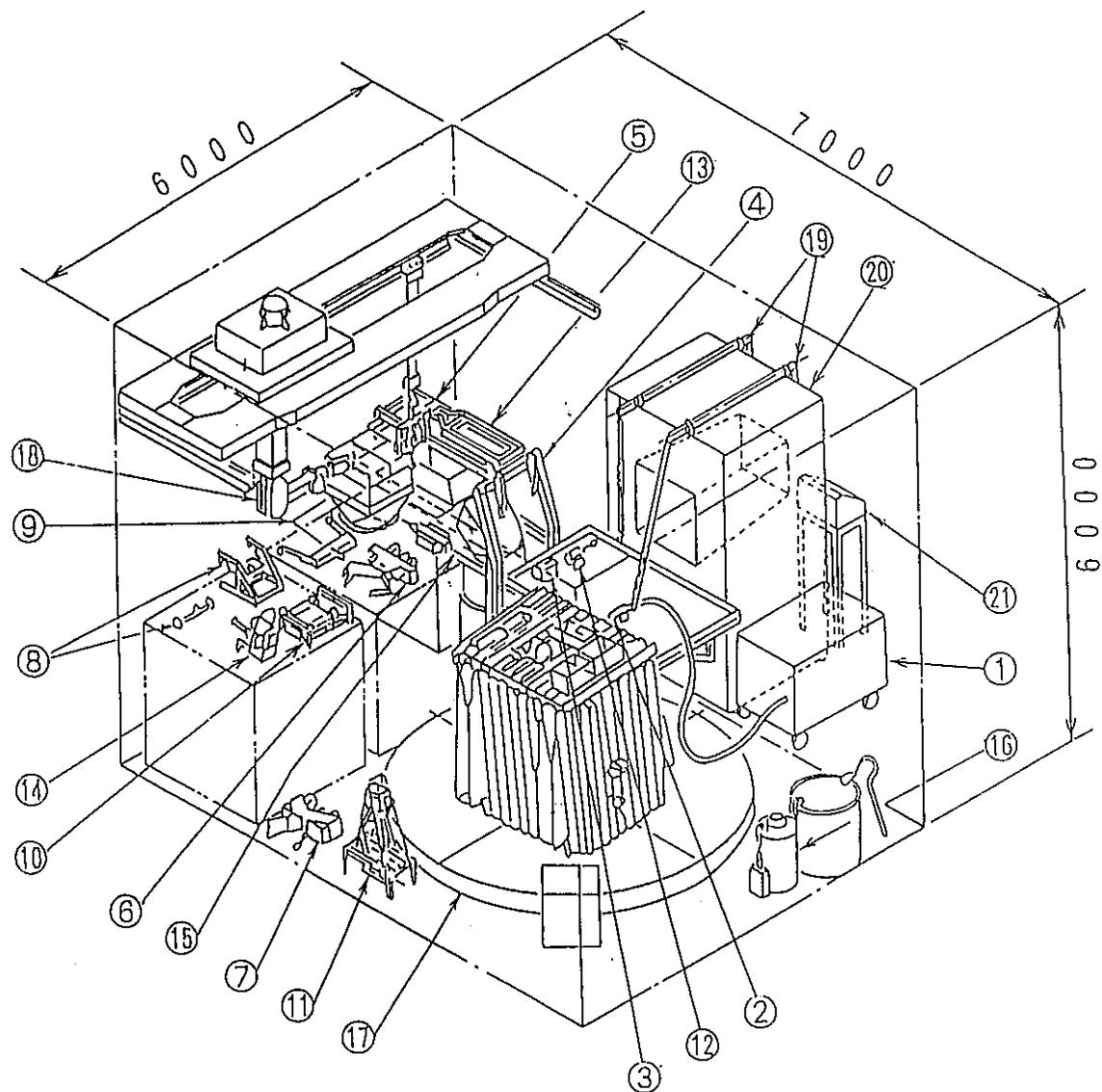
Heating	Joule-Heating
Containment Vessel	SUS 304
Electrode	Inconel 690
External Dimension	W2020XL1735XH1805
Refractory	K-3 2020 kg Other 4970 kg
Total Weight	16015 kg

## SPECIFICATION OF M/U-2 MELTER



## PROCEDURE OF DISMANTLING FOR MELTER(TVF)

- 88 -

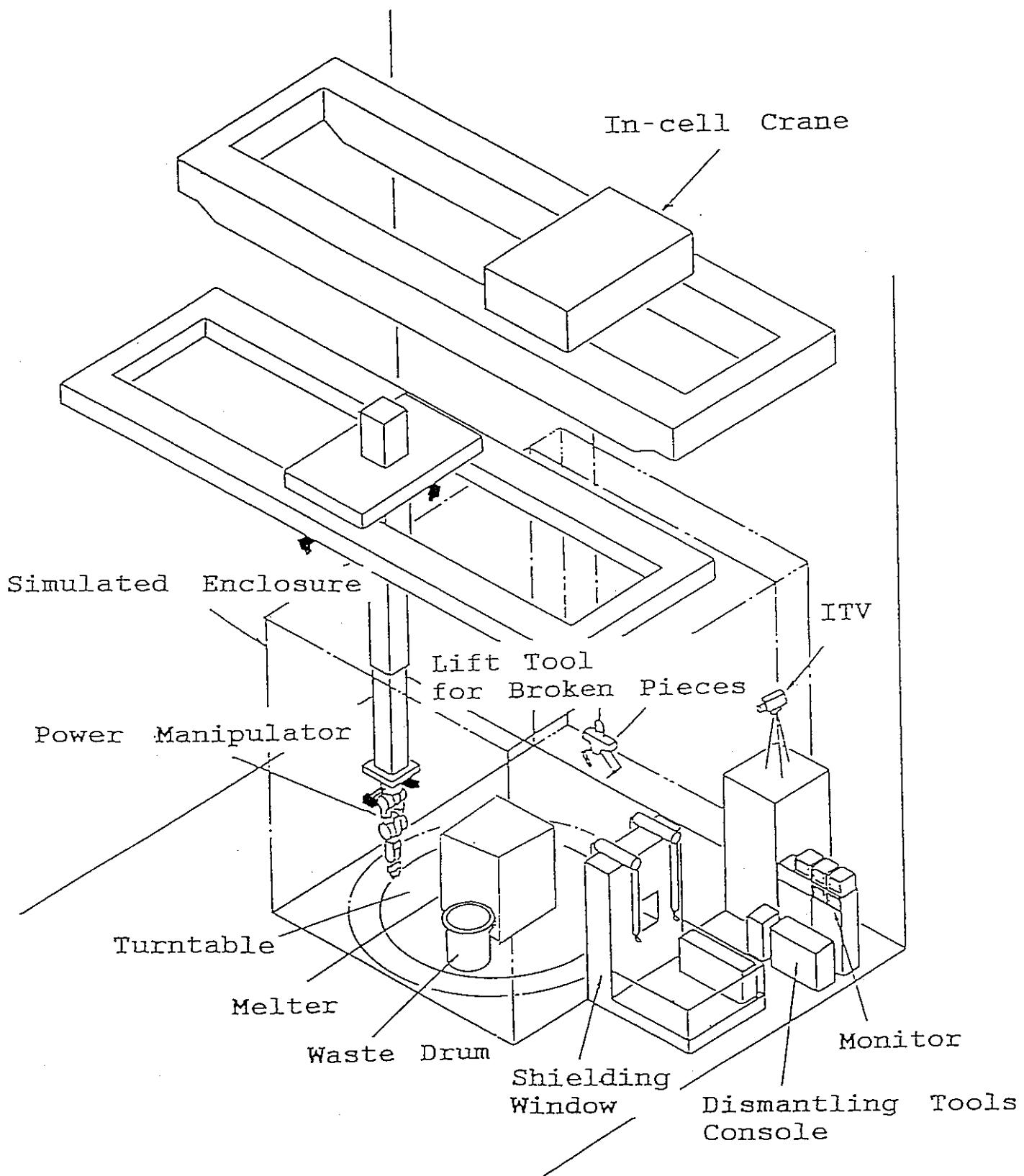


1 ~ 3	Plasma cutter
4	Hacksaw
5	Hoist scale
6 ~ 7	Handling tools of cut piece
8	Handling tools of electrode
9	Receiver
10 ~ 11	Internal dismantling breaker
12	Impact wrench
13 ~ 14	Handling tools of electrode
15	Breaker
16	Vacuum cleaner
17	Turn-table
18	Power manipulator
19	Master/slave manipulator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

CONCEPT OF THE DISMANTLING AREA (TVF)

Equipment and tools used at melter dismantling

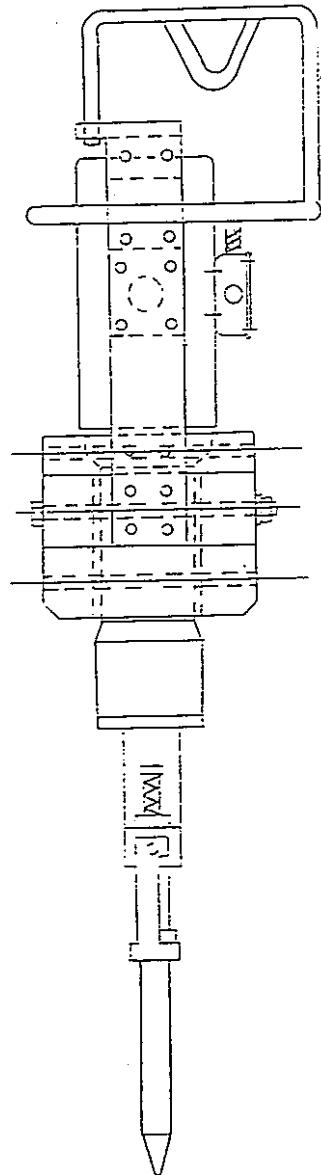
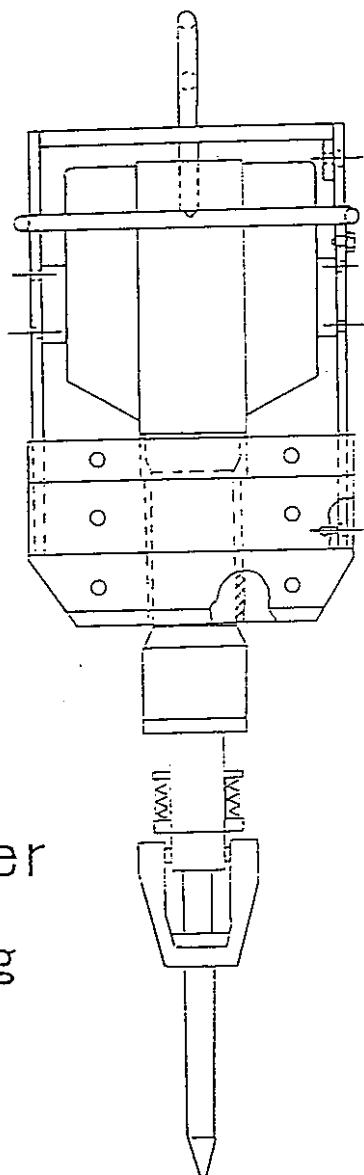
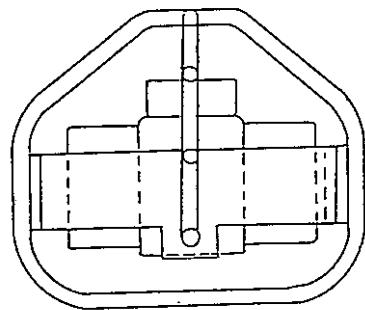
Plasma torch,	power	350A
Breaker,	electric power system	
Gripper,	electric power system	
Power Manipulator,	handeling weight	~ 100kg
Turntable,	~ 4m	diameter



### Outline of Dismantling Test Equipments

## Dismantling Test Schdule

	1993		1994	
	DEC.	JAN.	FEB.	MAR.
Melter Lid portion (Casing & Refractories)	○	○		
Melter Middle portion (Refractories & Electrode )		○	○	
Melter bottom portion (Refractories & Casing)			○	○

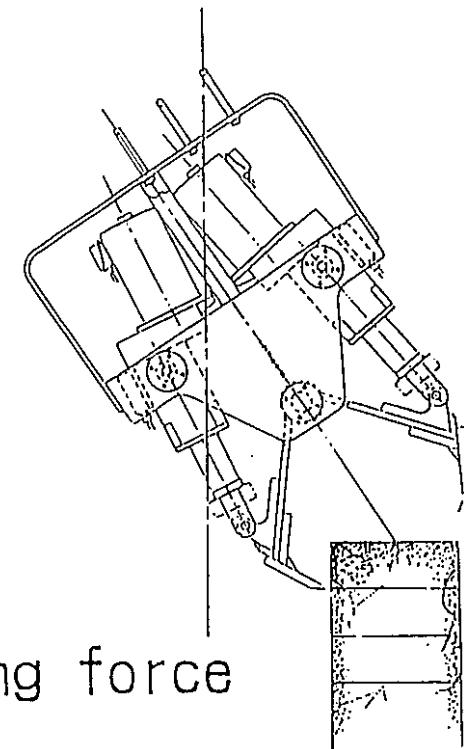
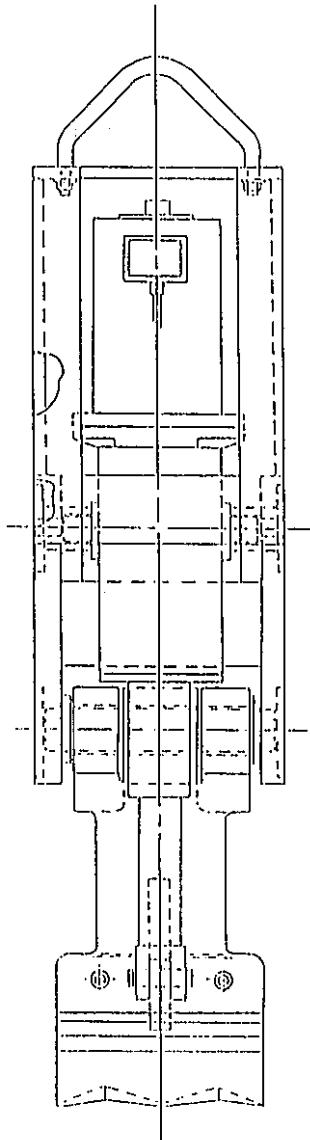
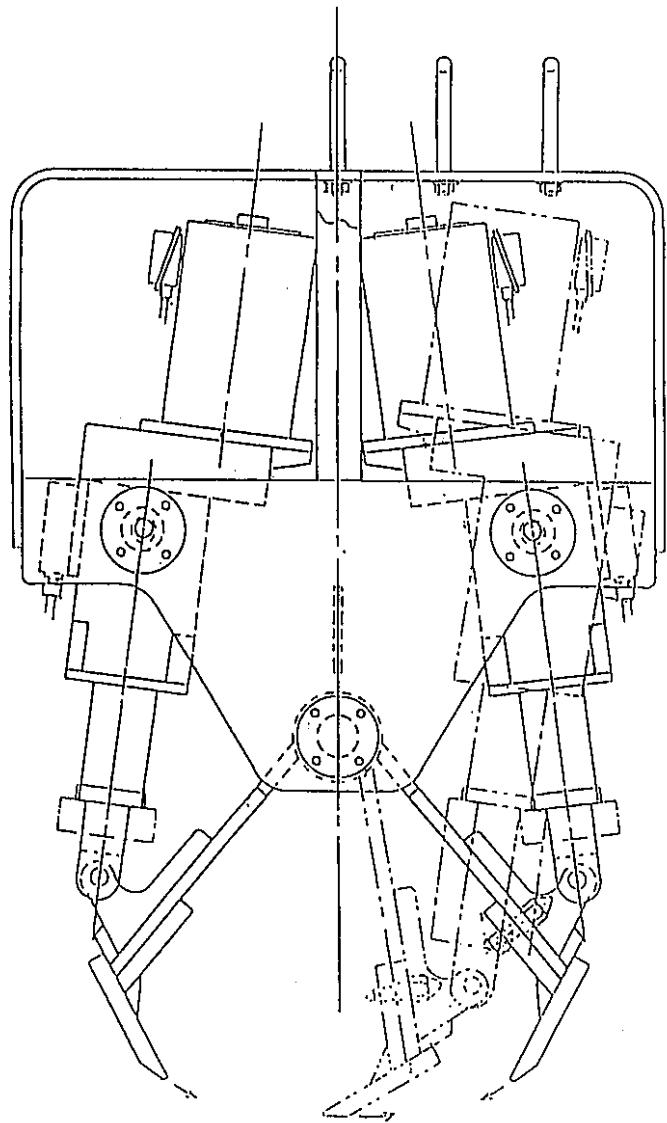


Driving force

: electric power

weight : 140kg

# Breaker



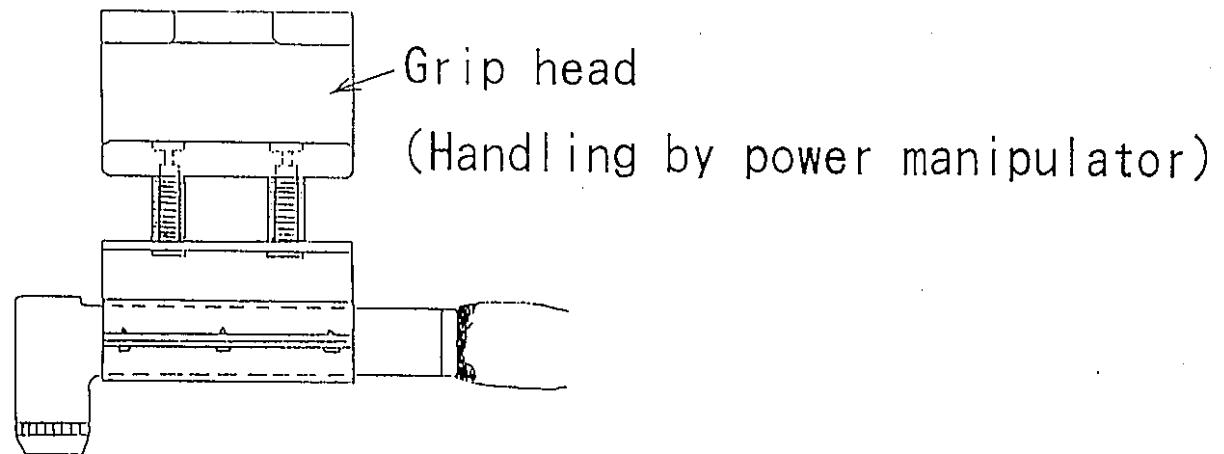
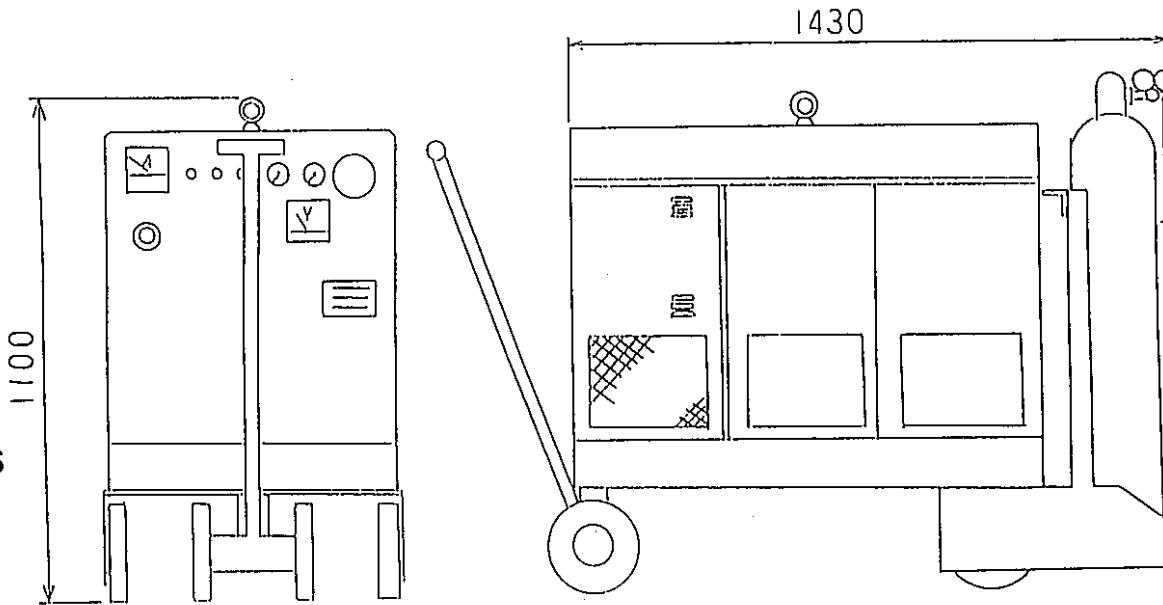
Driving force  
: electric power  
Handling capacity  
: 300kg  
weight : 240kg

Gripper tool

Plasma gas:  $\text{CO}_2 \times \text{N}_2$  gas

Cutting speed  
: 10mm/s(25mm-SUS304)

Weight : 500kg



Cutting machine and Plasma torch

2.5 New glass melter

# New glass melter

Prepared for  
The 12th Annual KfK-PNC Meeting  
on  
Cooperation in High-Level Waste Management

by IGARASHI, H.

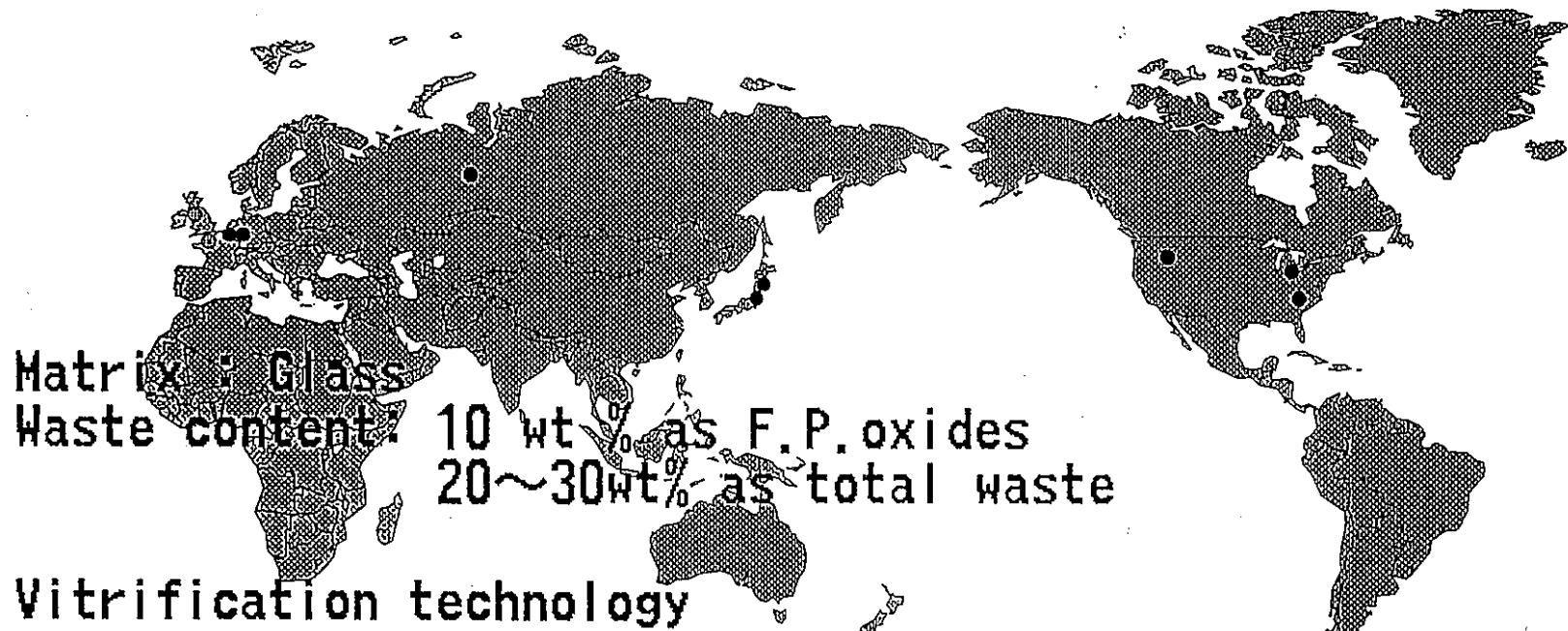
December 7-9, 1993

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Power Reactor and Nuclear Fuel  
Development Corporation



# Commercialized Technology for Vitrification of HLLW



## Vitrification technology

- Joule-heated melter: Japan (TVF, Rokkasho)  
Germany (PAMELA, WAK)  
USA (SRP, WVDP, HWVP)  
Russia (MAYAK)
- Calcining/Induction heating  
France (AVM, AVH), UK(HVP)

# Trend of Advanced Vitrification Technology

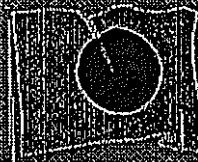
USA: Stirred melter, High-temperature melter  
(longer life, noble metals, higher-processing rate,  
higher waste loading)

Application to treatment of TRU, LLW, Mixed waste

France, Russia: Cold crucible induction melting  
(Higher melting temperature/ diversified glass composition  
application to medium and low level waste)

Germany: Joule-heated melter with steeply sloped floor  
UK: ?

# Development of Advanced Melting Technology in Japan



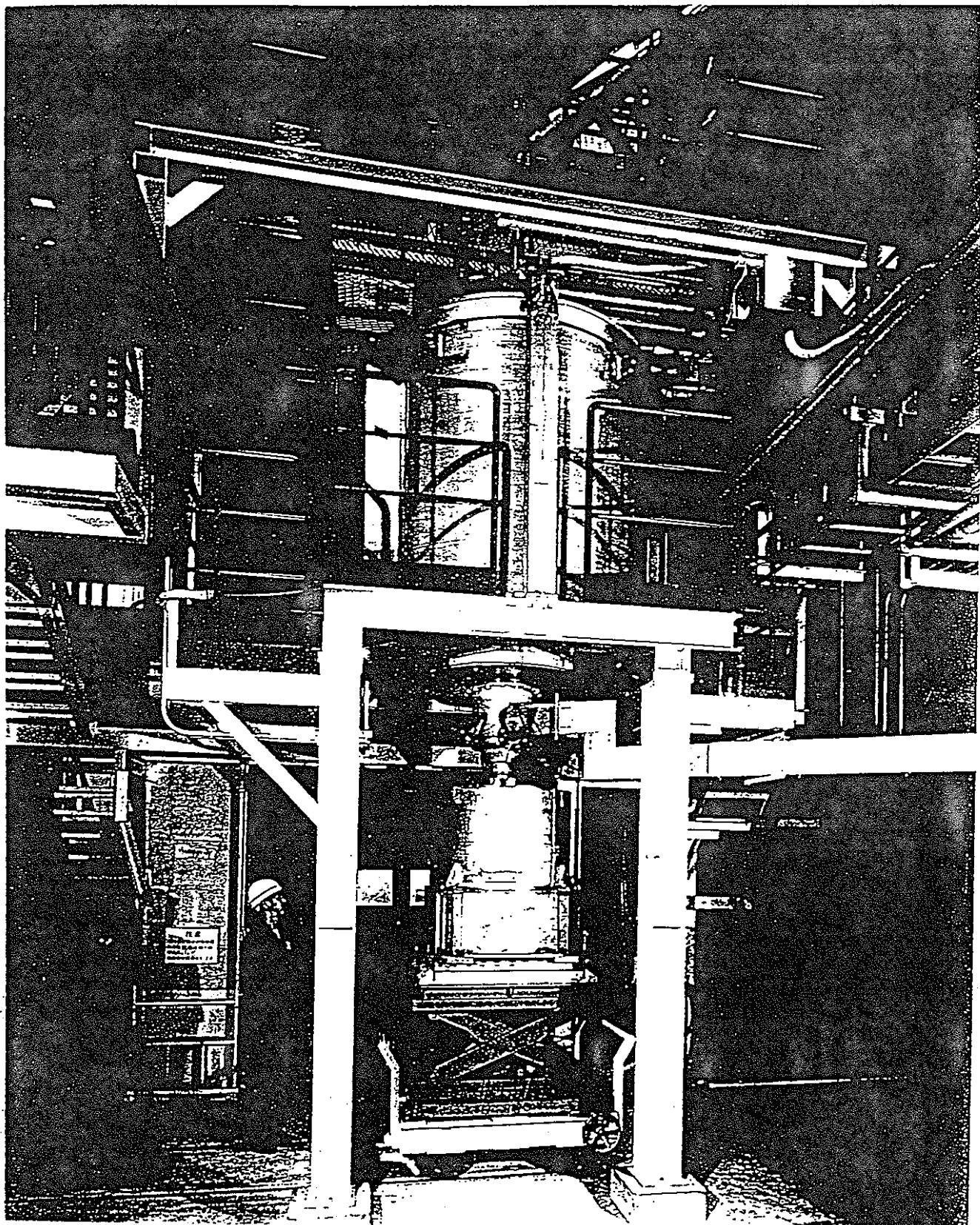
## 1. JUCIM (Double-heated cylindrical electrode melter)

- Reduction of slagging waste
- Size reduction, longer life
- Simplification of components to be exchanged
- Operability by solid state materials in higher concentration
- Possibility to realize higher temperature FBR reprocessing in higher waste loading

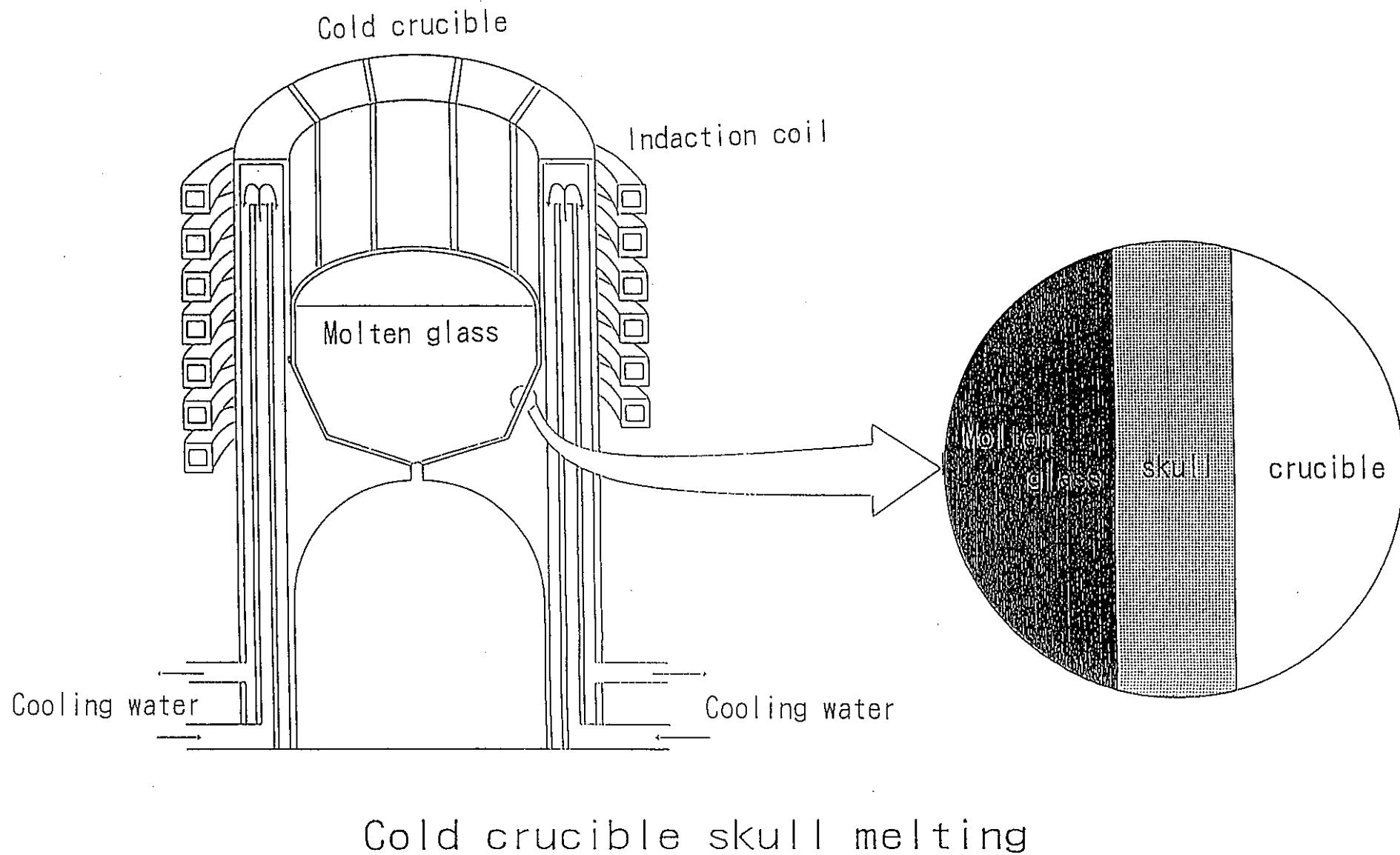
## Development of structure (electrode arrangement)

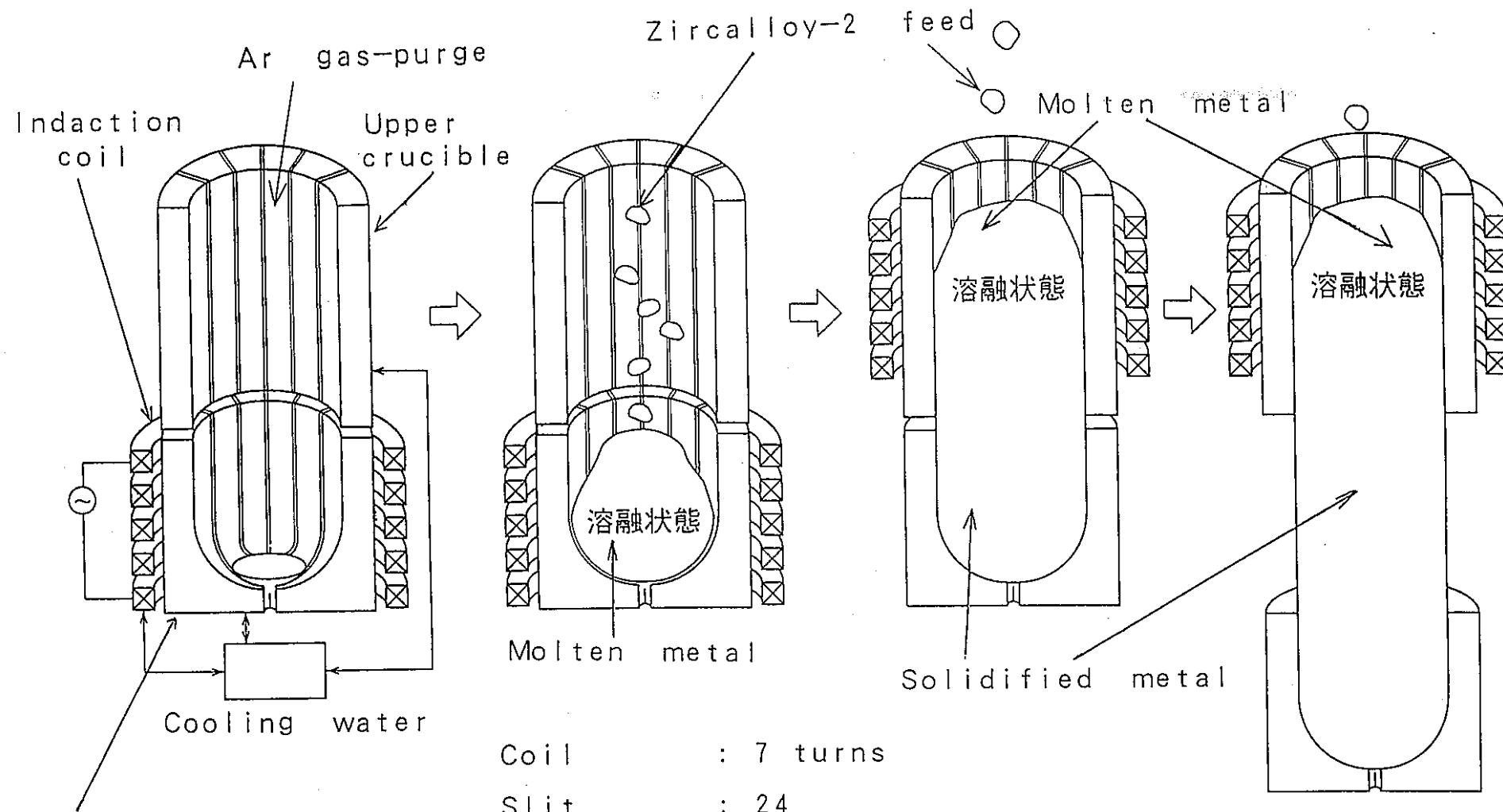
## 2. Crucible Induction Melting

- Performance at low temperatures/flexibility of various composition com.
- Simplification of crucible
- High energy input, melting rate



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Coil : 7 turns

Slit : 24

Power : Max. 110 kW

Frequency : Average 67 kHz

Production of solidified Metallic wastes by crucible induction melting

## OPERATIONAL RESULTS OF SMALL SCALE TEST MELTER OF UICBM

Demonstrated technical feasibility of the UICBM concept  
by achieving the basic operational characteristics  
of higher melting rate than conventional UICM  
stable operation for the waste pure metal melt  
(gross thermal efficiency 95-102%)

# JCET-M-E 1 Campaign

## 10 Dec. Optique

## Evaluation of basic performance of engineering scale reactor in cooling operation

2010-09-01 00:00:00 2010-09-01 00:00:00

- Melting temperature—900, 1000, 1100°C
  - Glass level—L, M, H-level
  - Electrode cooling air—0 ~80 Nm<sup>3</sup>/h
  - Pelletum pressure—0 mmHg

## 30 Results

- Appropriate flow rate of cooling air of electrodes  
Inner: 40Nm<sup>3</sup>/h Outer: 30Nm<sup>3</sup>/h

# JCEM-E1 Campaign (continued)

## 3) Results (continued)

- Heat balance

(e.g., 1100°C, - level, Inert: 40m<sup>3</sup>/h, Outer : 20m<sup>3</sup>/h)

Power	31 kH
Heat loss from surface	66.6 (%)
Cooling air(Inner)	7.5
Cooling air(Outer)	8.4
Off-gas	17.2
Cooling of feed nozzle	1.3

- Cooling off outer electrode promotes natural convection of molten glass.
- Effect of glass level and melting temperature
- Two tests of glass draining, evaluation of operational procedure

# JCEM-E2 Campaign

## 1) Objectives:

Preliminary evaluation of feeding system and operational characteristics under waste feeding

## 2) Operational condition:

- Feeding rate ~14.2 /h (glass production ~8.7kg/h)
- Heating temperature  $1150 \pm 50^{\circ}\text{C}$
- Simulated waste SW-18 (no noble metals)
- Glass additives - Beads additives (PFT98)
- Glass composition - P07PBT(TVF glass)

## 3) Results:

- Production 1.3 m<sup>3</sup> of waste, 816 kg of glass
- Two glass forming test
- Maximum feeding rate was 14.2 /h

# JCEM-E 3 Campaign

Objective: Evaluation of maximum melting rate

Time: Oct. 18~Nov. 19, 1993

          interrupted by malfunction in control system

Processed amount: 6 batches, 3m<sup>3</sup>, 1.9 ton

Maximum melting rate: less than 14kg/h

Melting temperature: 1150±50° C

Simulated waste: SH-18.23 (no mobile metals)

Glass additives: Beads additives(PFT98)

Glass product composition: P0798(TVF glass)

## Future work

- Evaluation of maximum melting rate
- Temperature distribution of melting cavity
- Thermal balance
- Evaluation of heat resistance of electrode materials (corrosion, creep, thermal cycle)
- Confirmation of compatibility with noble metal elements
- Entrapments into off-gas
- Feasibility study of application to TUF and other waste treatment field (TRU, LLW)

## 2.6 High waste loading glass

# High waste loading glass

Prepared for  
The 12th Annual KfK-PNC Meeting  
on  
Cooperation in High-Level Waste Management

by SASAGE, K.

December 7-9, 1993

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Power Reactor and Nuclear Fuel  
Development Corporation



# **Selection of glass composition**

## **Effect of high Na<sub>2</sub>O concentration**

# Requirements of high waste loading glass

Heat : Separation of Cs & Sr

Yellow phase :

Separation of Mo

Operability :

Separation of noble metals

# Objective

**Waste loading**

Waste 45wt.% and No yellow phase

**Leaching rate**

Equivalent to 25wt.% waste glass

**Producibility**

Possible to melt and drain by LFCM

# Plan

Increase  $\text{MoO}_3$  solubility

Increase  $\text{Cr}_2\text{O}_3$  solubility

Increase chemical durability

# Sample compositions

Glass code		P1029	P1030	P1031	P1032	P1033	P1034	P1035
Glass frit	$\text{SiO}_2$	32.27	31.27	30.27	30.27	30.27	30.27	30.27
	$\text{B}_2\text{O}_3$	11.45	12.45	12.45	12.45	12.95	12.45	12.45
	$\text{Li}_2\text{O}$	2.2	2.2	2.2	1.2	0.2	1.2	0.2
	$\text{CaO}$	2.2	2.2	2.2	2.2	2.2	3.2	4.2
	$\text{ZnO}$	2.2	2.2	2.2	3.2	4.2	2.2	2.2
	$\text{Al}_2\text{O}_3$	4.69	4.69	5.69	5.69	5.19	5.69	5.19
Waste	$\text{MoO}_3$	1.62	1.62	1.62	1.62	1.62	1.62	1.62
	$\text{Na}_2\text{O}$	11.75	11.75	11.75	11.75	11.75	11.75	11.75
Total Waste		45.01	45.01	45.01	45.01	45.01	45.01	45.01

# Result

Melting temperature : 1100°C

MoO<sub>3</sub> concentration : 1.62wt.%

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Glass code	P1029	P1030	P1031	P1032	P1033	P1034	P1035
Yellow phase	Nothing (Good)						
Leaching rate ( $\times 10^{-4}$ kg/m <sup>2</sup> d)	3.5	4.2	3.1	2.8	2.5	2.9	2.5

# Result

Melting temperature : 1100 °C

MoO<sub>3</sub> concentration : 2.50wt.%

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# Result

Melting temperature : 1050 °C

MoO<sub>3</sub> concentration : 2.50wt.%

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Glass code	P1029	P1030	P1031	P1032	P1033	P1034	P1035
Yellow phase	Nothing (Good)	Nothing (Good)	Separated	Separated	Nothing (Good)	Separated	Separated

# Objective

Characterization of  
high waste loading glass

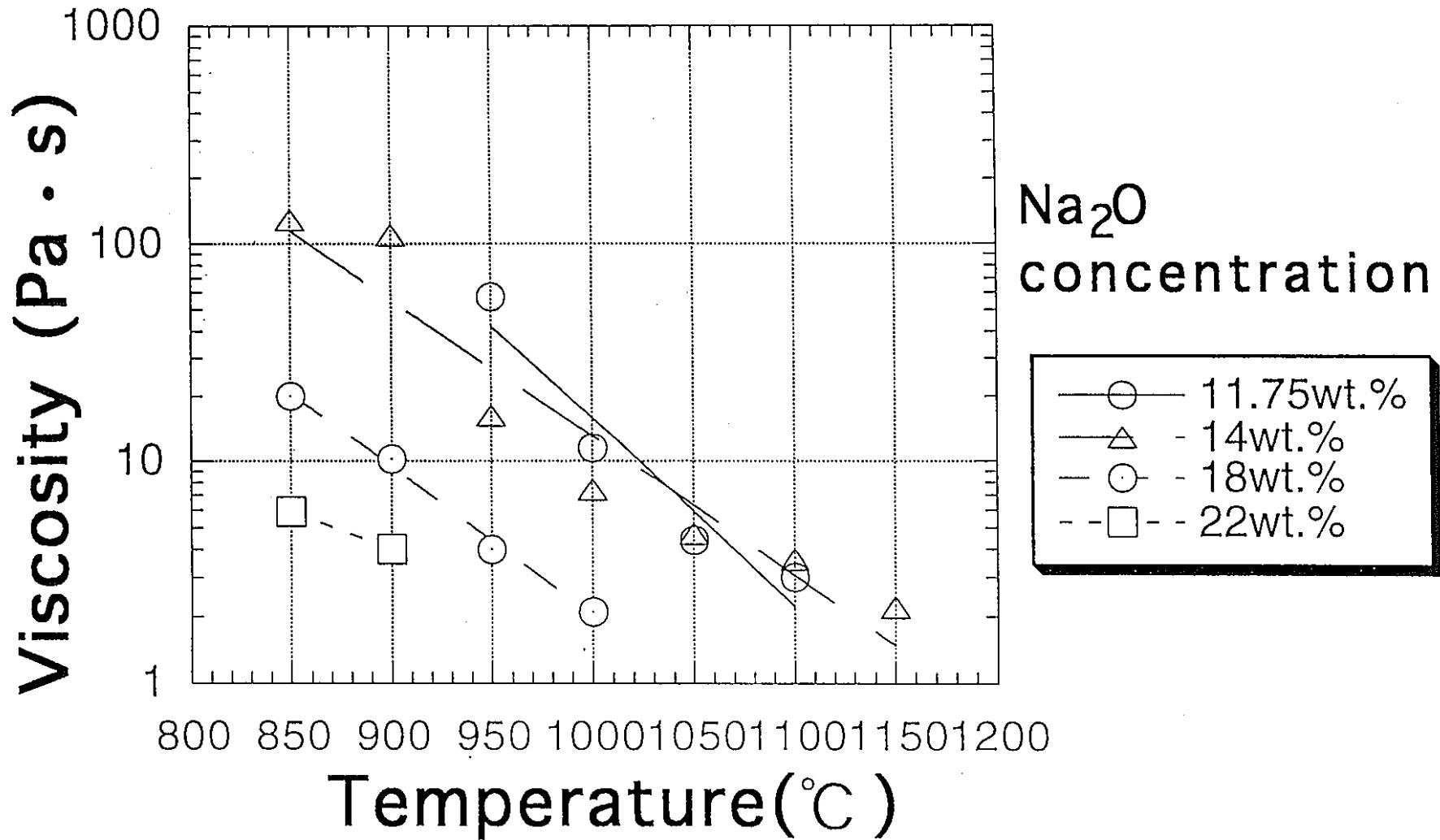
## Evaluation items

Producibility  
(Viscosity and Resistivity)

Chemical durability

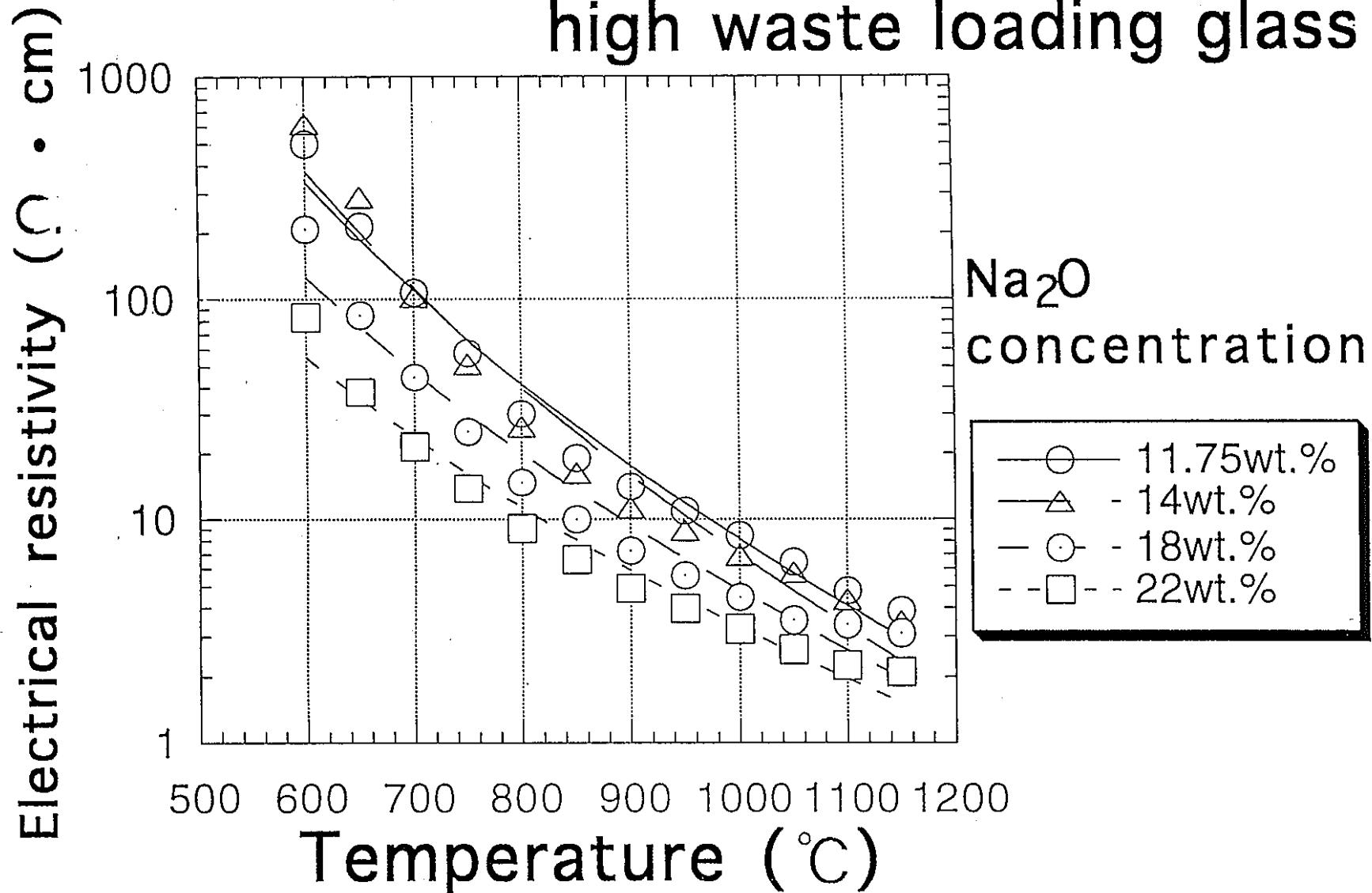
Yellow phase

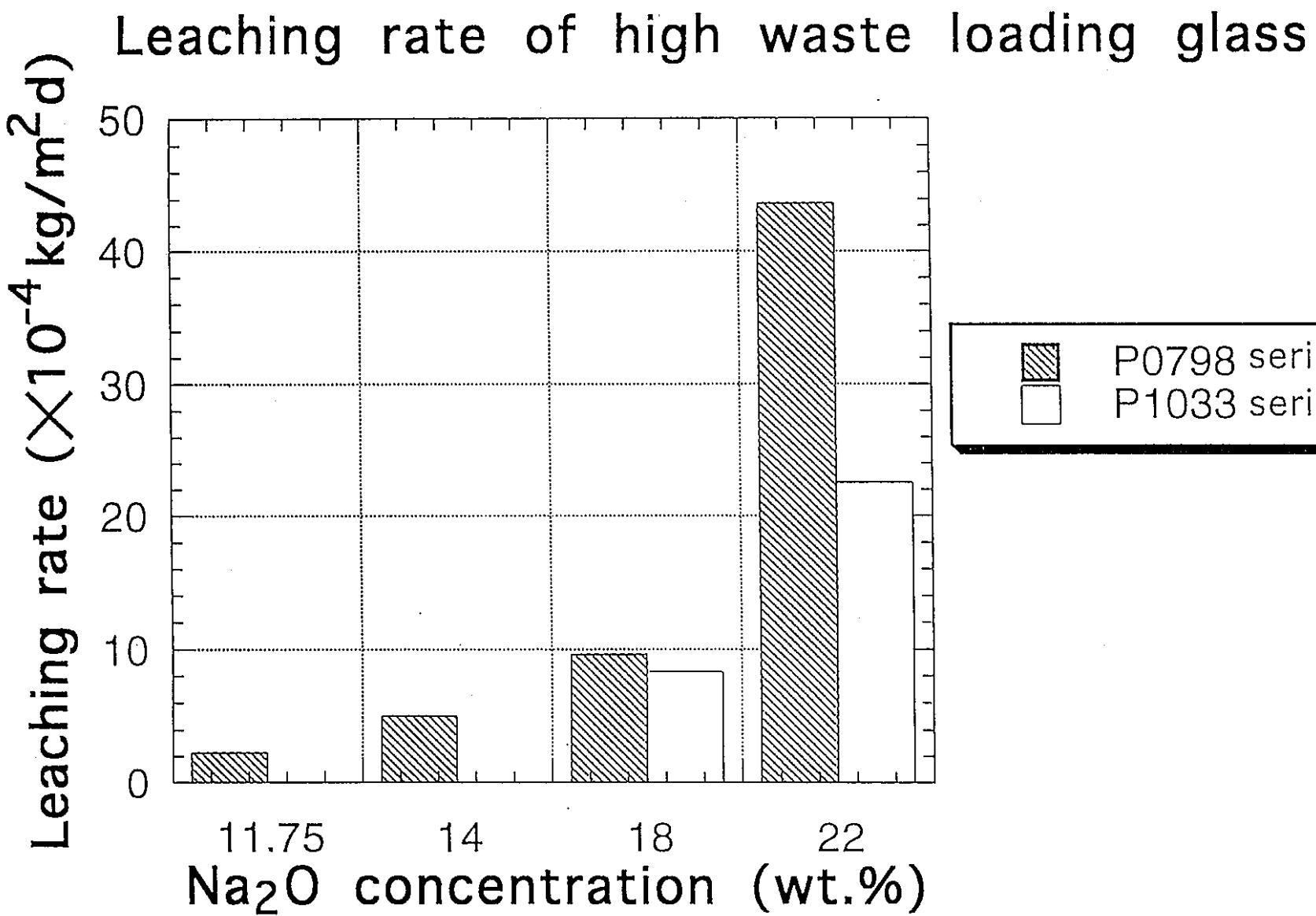
# Viscosity of high waste loading glass



# Electrical resistivity of high waste loading glass

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# Solubility of MoO<sub>3</sub>

Glass code	P0798 series	P0798 series	P1033 series	
Na <sub>2</sub> O concentration (wt.%)	18	22	22	
MoO <sub>3</sub> concentration (wt.%)	0.6	Nothing	Nothing	Nothing
	0.8	Nothing	Y	Nothing
	1.0	Y	Y	Nothing
	1.2	Y	Y	Y

# Summary

We made new glass composition with  
high MoO<sub>3</sub> solubility & high chemical durability.

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If waste glass contain much Na<sub>2</sub>O,  
its MoO<sub>3</sub> solubility & chemical durability decrease.

New glass composition showed high MoO<sub>3</sub>  
solubility & high chemical durability.

2.7 Separation of heat-generating elements from high-level liquid waste by denitration

# **Separation of Heat-Generating Elements from High-Level Liquid Waste by Denitration**

Prepared for  
The 12th Annual KfK-PNC Meeting  
on  
Cooperation in High-Level Waste Management

by YONEZAWA, S.

December 7-9, 1993

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Power Reactor and Nuclear Fuel  
Development Corporation



## Heat-Generating Elements : Cs , Sr

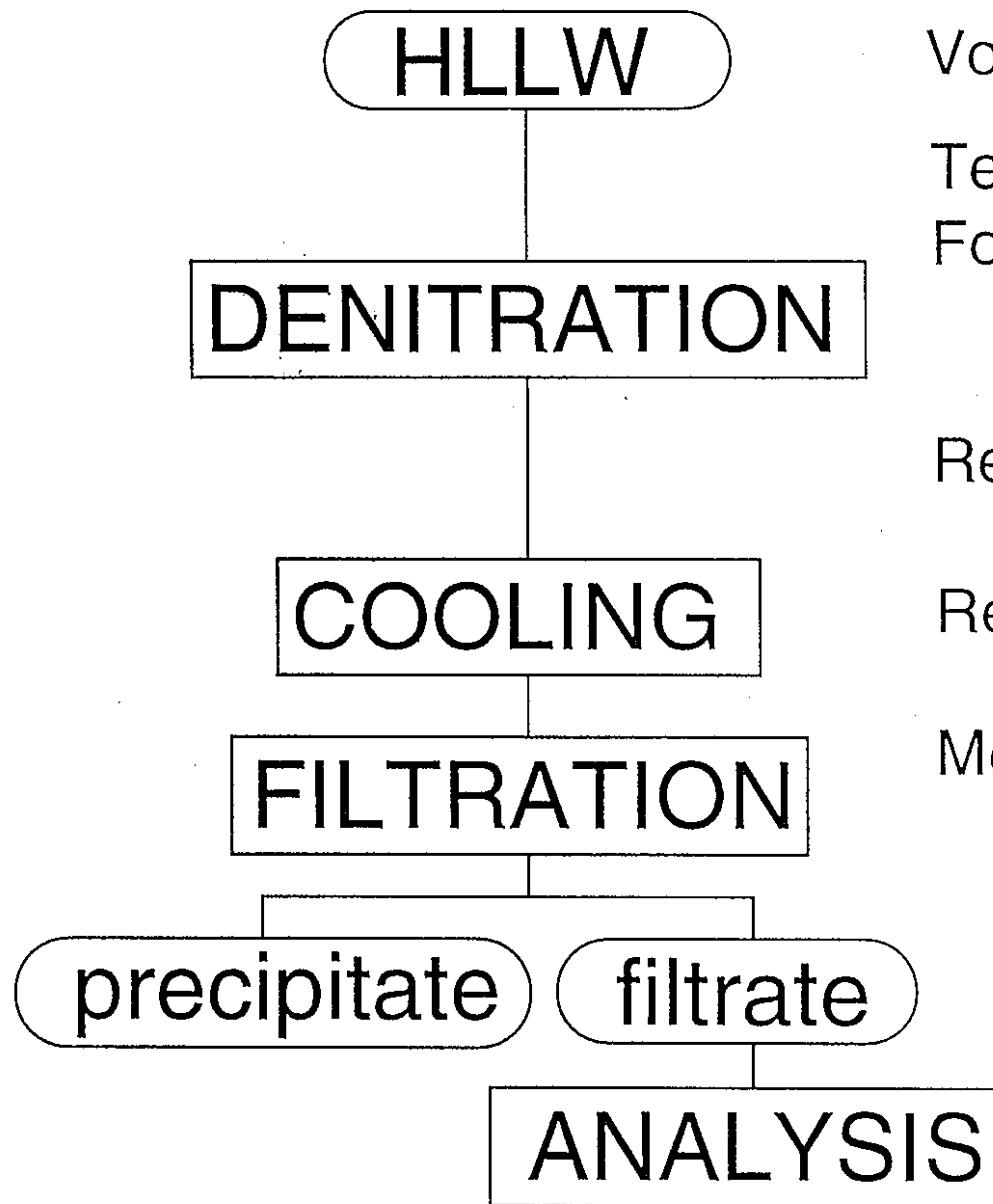
### Reason of choice

- 1 : No Secondary Products
- 2 : Based on Experience

### Purpose

Behavior of Elements in HLLW

Best Condition for Separation



Volume : 100 mL

Temp. : 95°C

Formic Acid

Feed Rate : 0.4 mL/min

$[HCOOH] / [HNO_3] = 1.9 \sim 2.1$

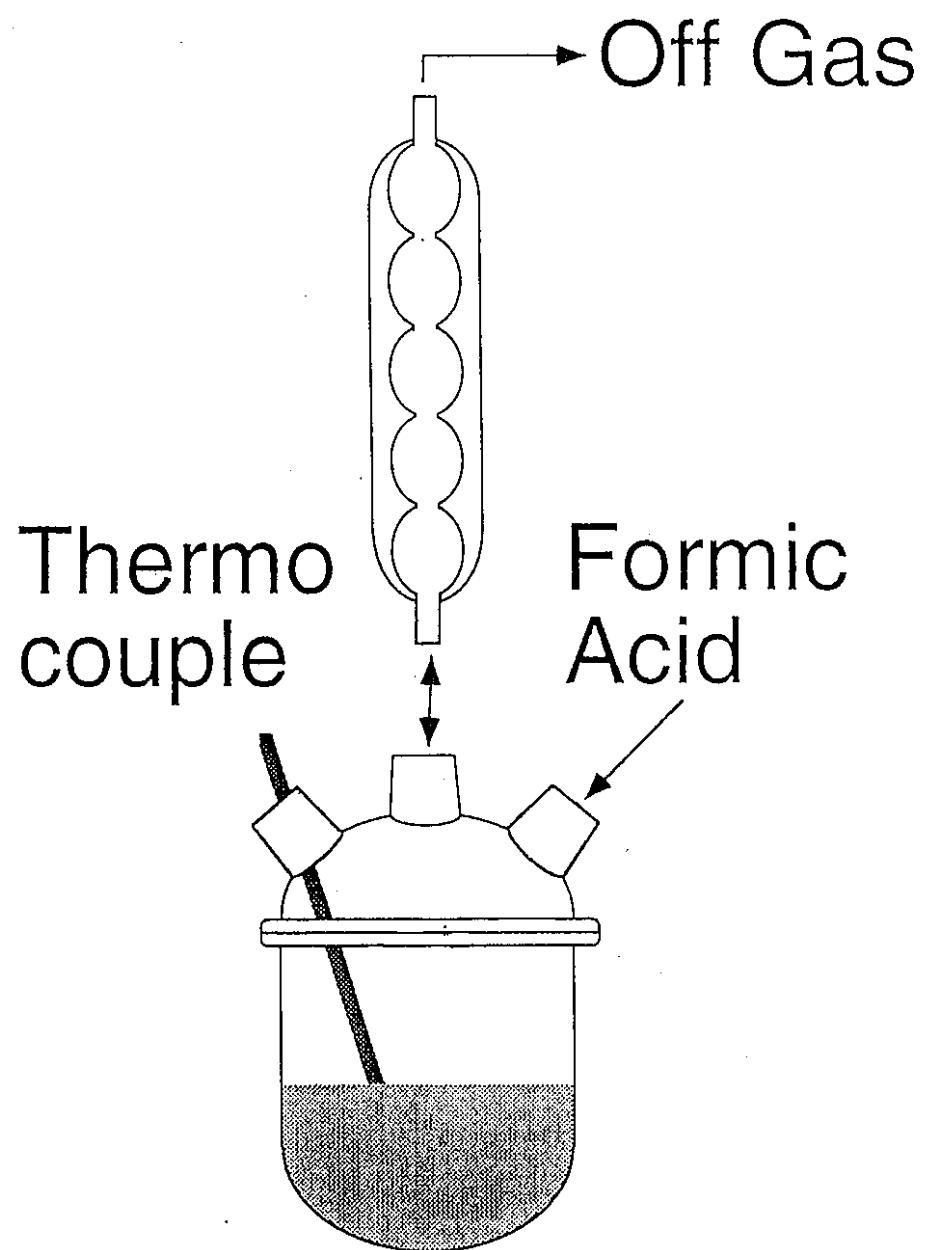
Retention Time : 6 hr

Retention Time : 16 hr

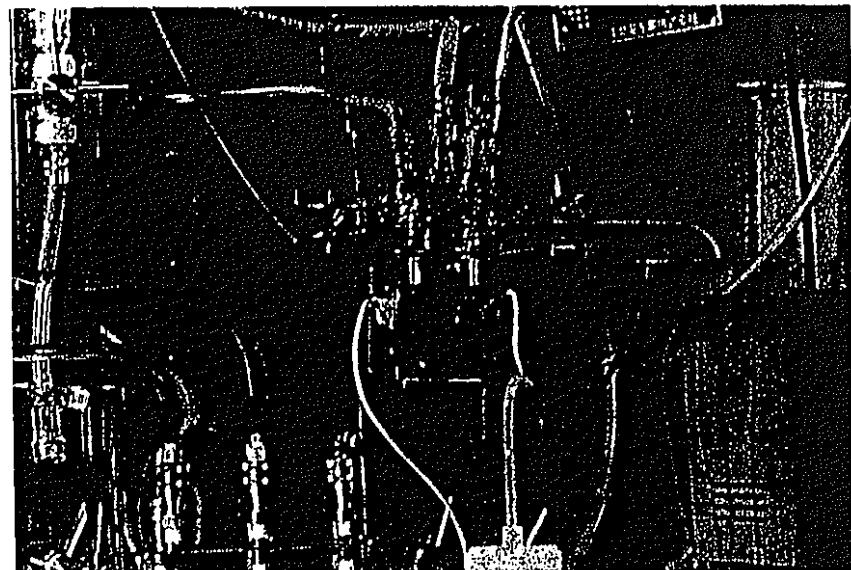
Membrane Filter

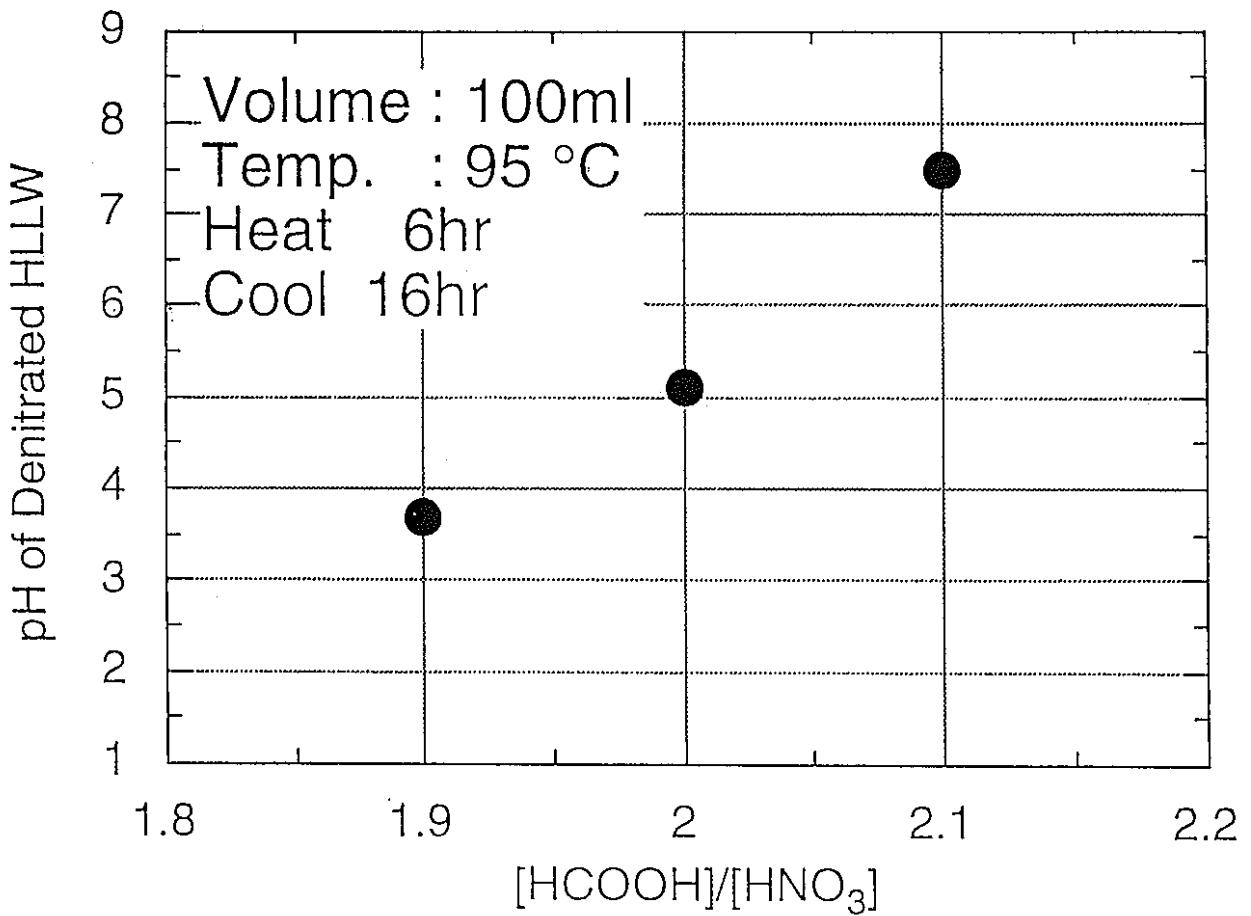
Pore Size : 0.45 μm

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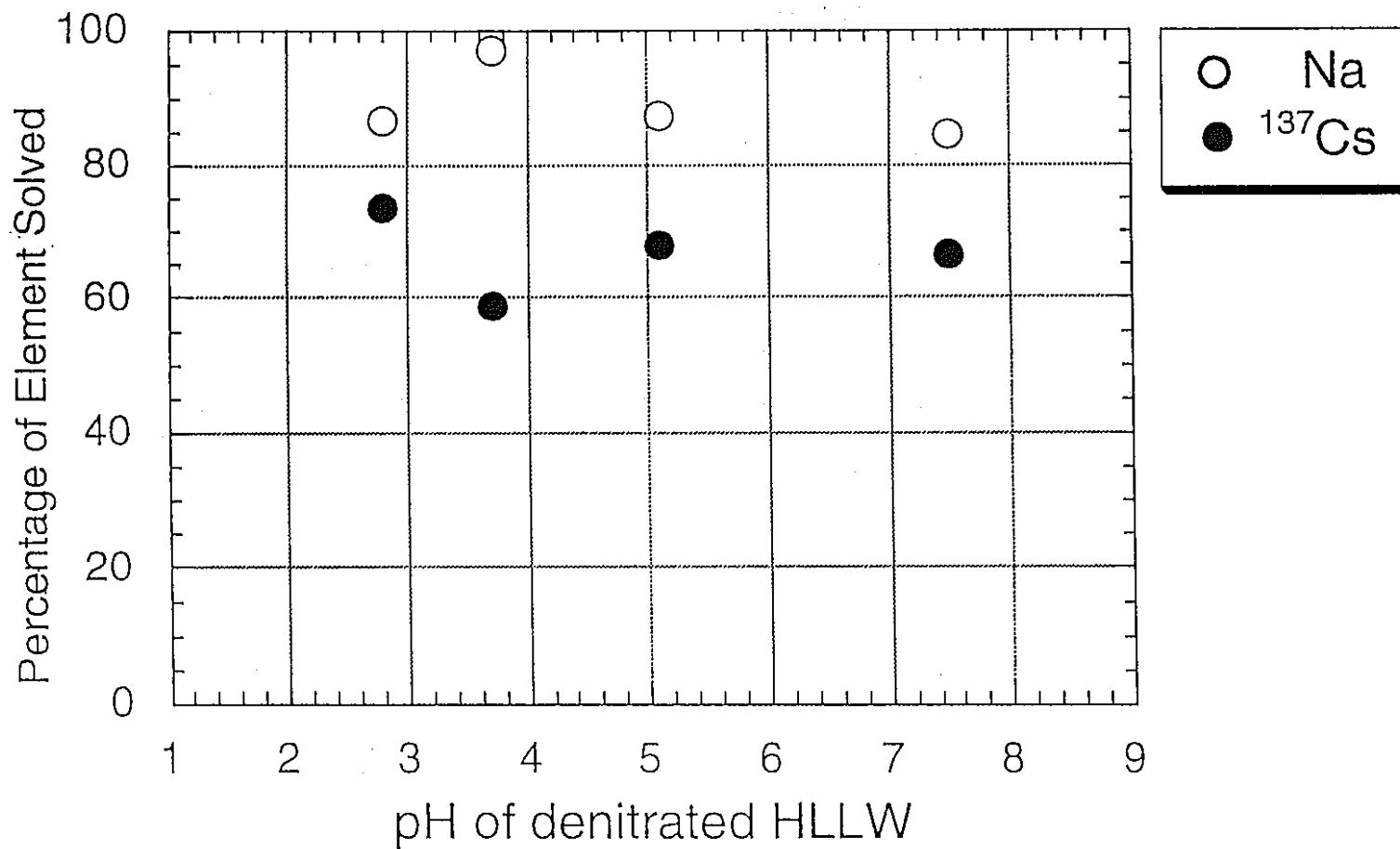
Photo



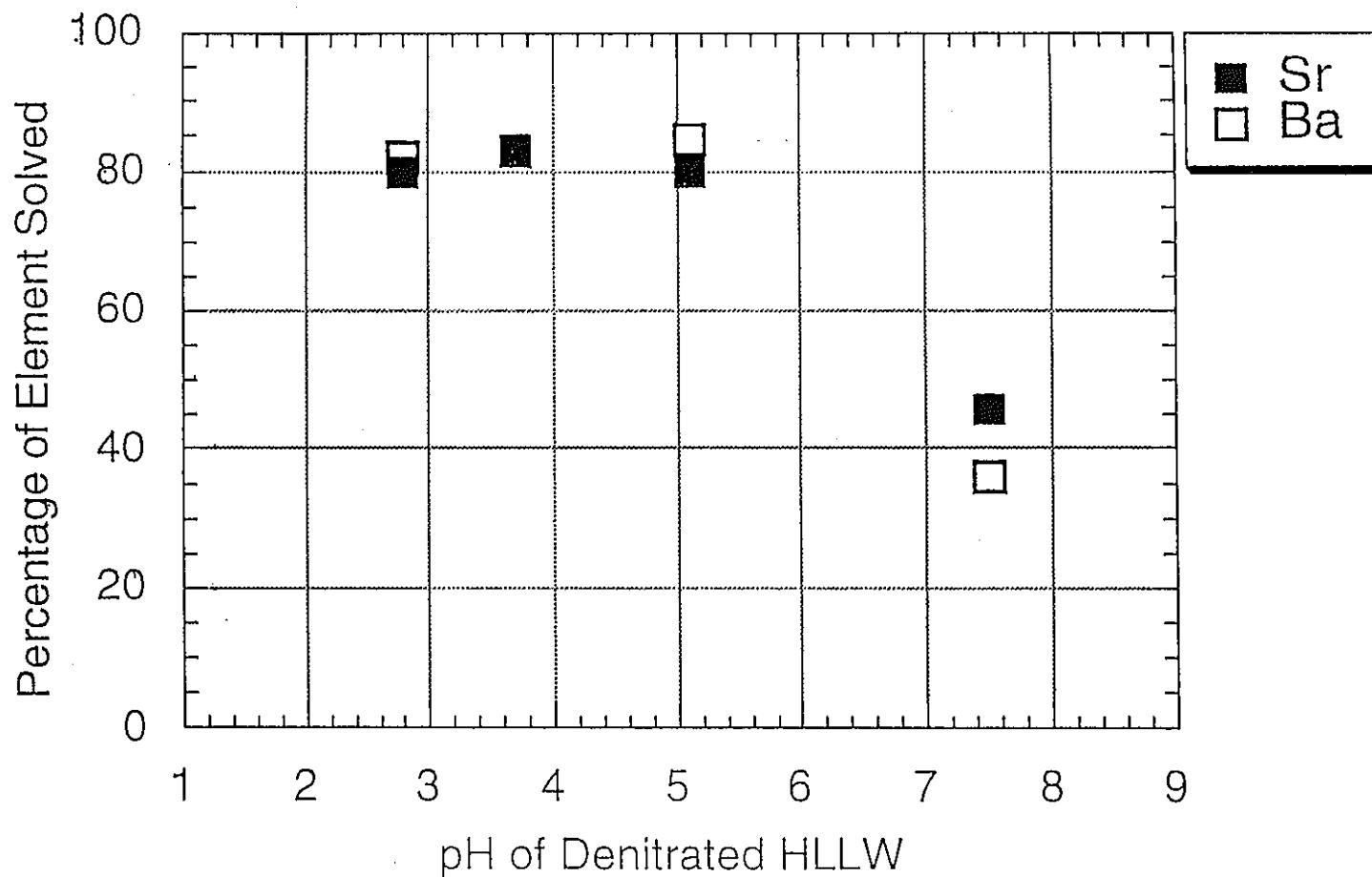


Relationship between [HCOOH]/[HNO<sub>3</sub>]  
and pH of Denitrated HLLW

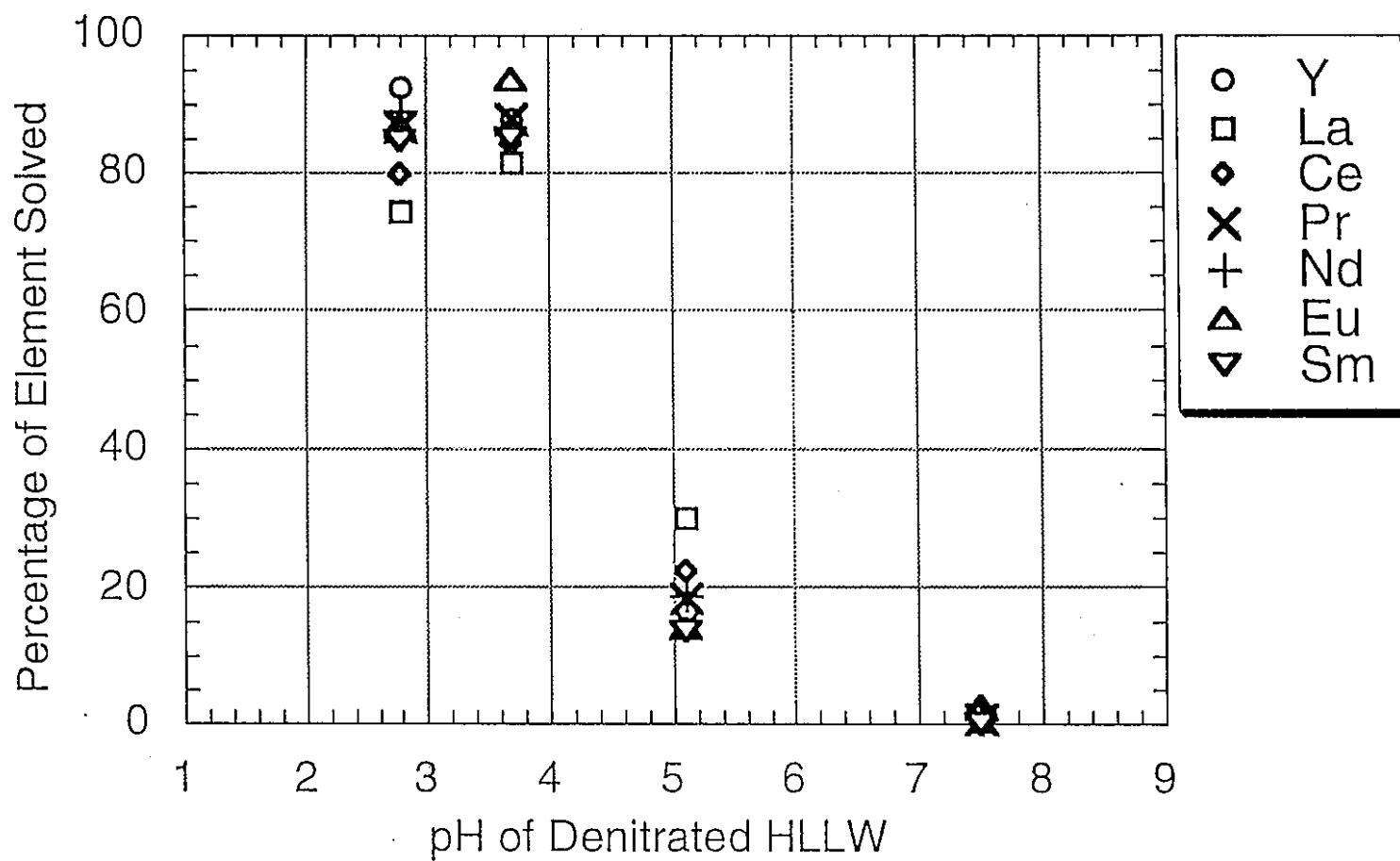
$$\text{Percentage of Element Solved} = \frac{\text{Concentration in Denitrated-HLLW}}{\text{Concentration in HLLW}} \times 100$$



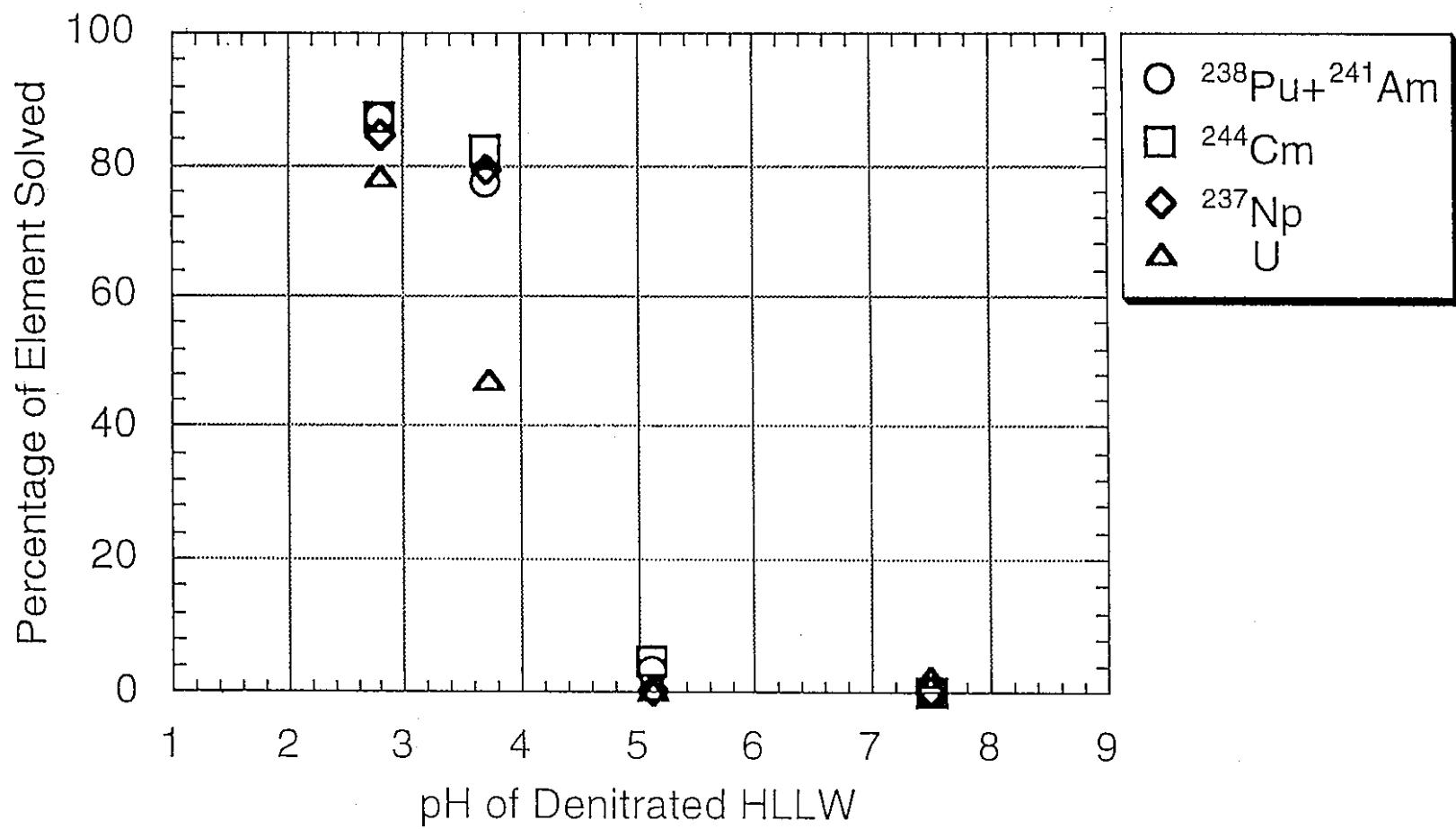
Relationship between pH of Denitrated HLLW and  
Percentage of Element Solved  
(Alkaline Metal Elements)



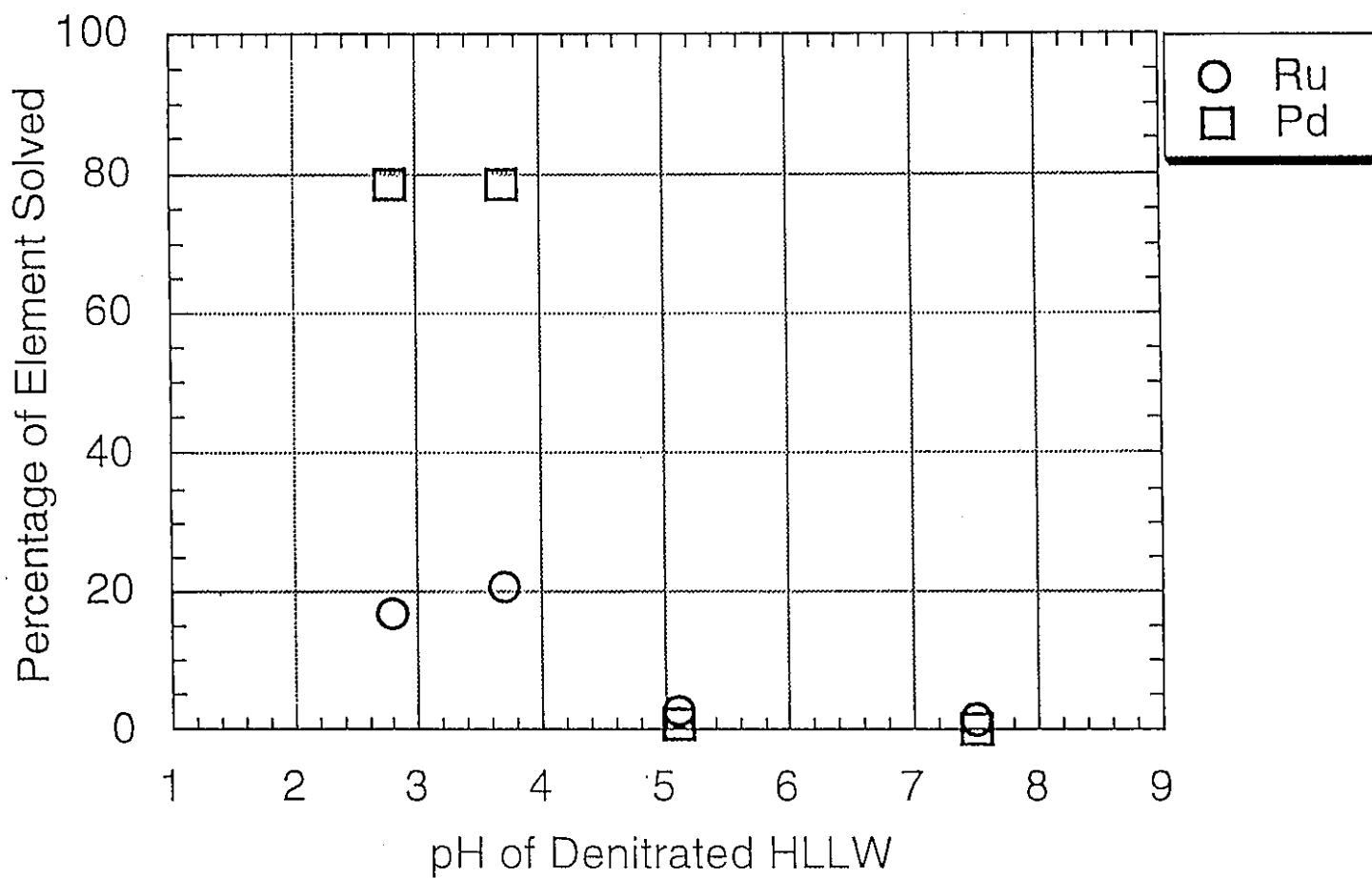
Relationship between pH of Denitrated HLLW and  
Percentage of Element Solved  
(Alkaline Earth Metal Elements)



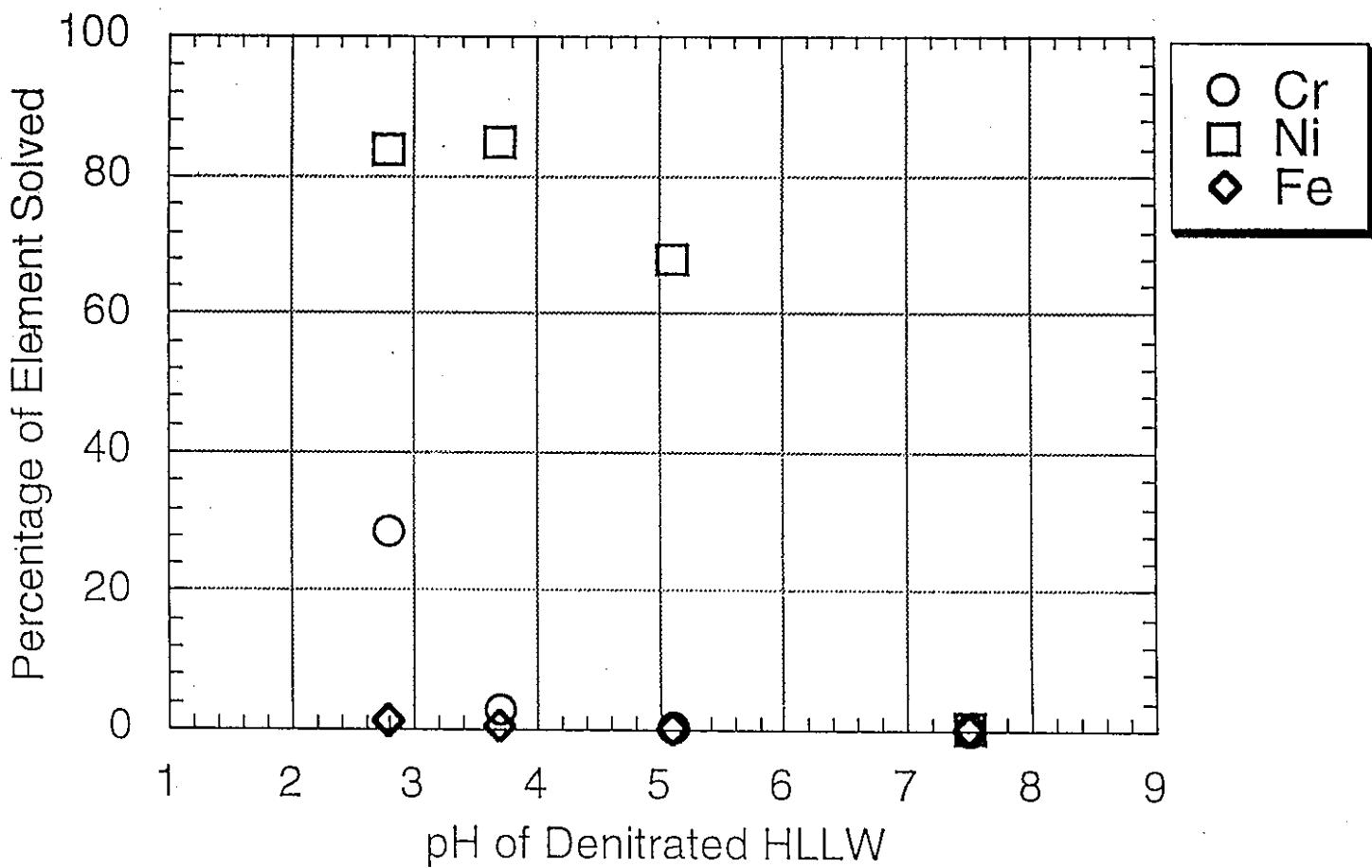
Relationship between pH of Denitrated HLLW and  
Percentage of Element Solved  
(Rare Earth Elements)



Relationship between pH of Denitrated HLLW and  
Percentage of Element Solved  
(TRU Elements)

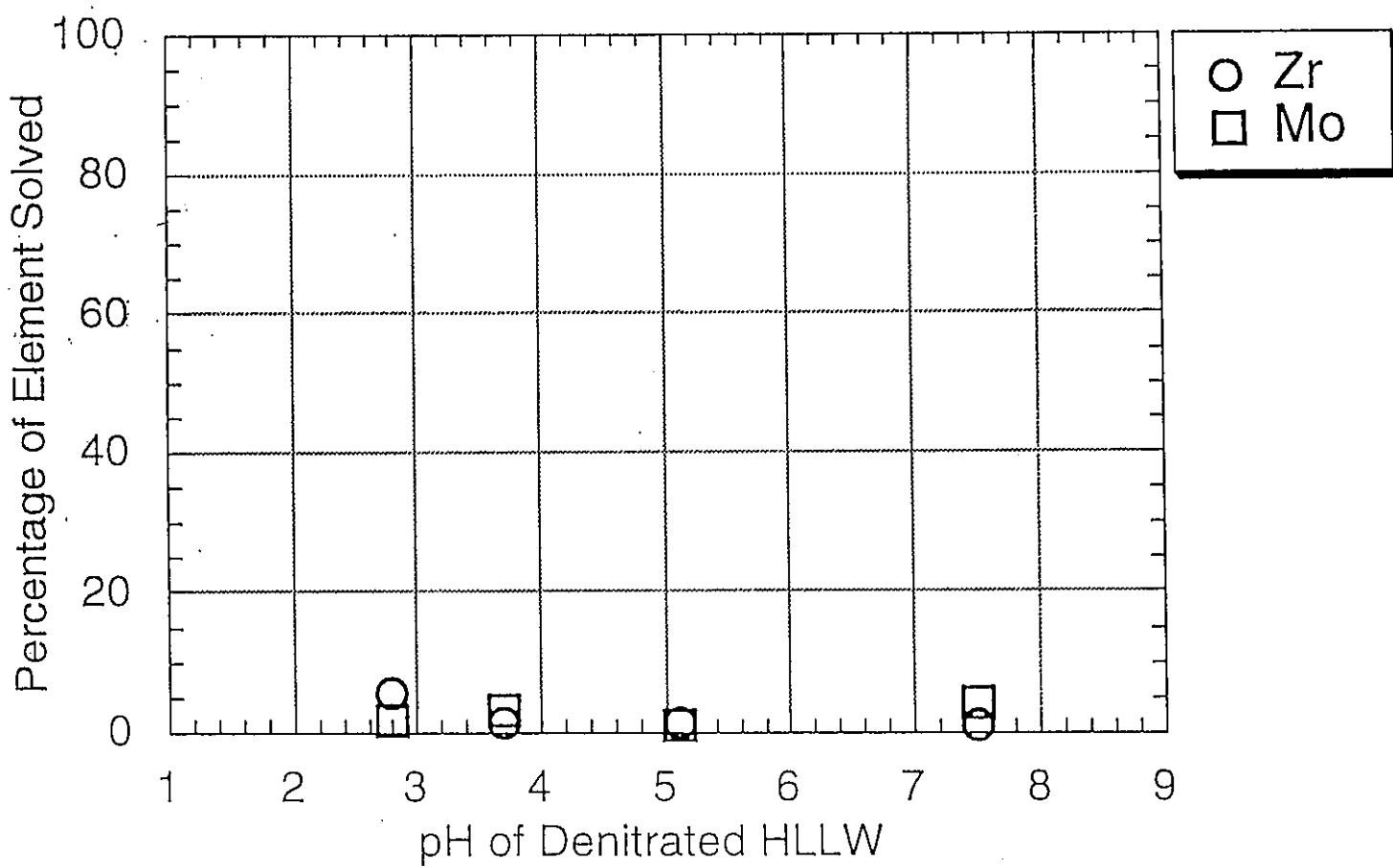


Relationship between pH of Denitrated HLLW and  
Percentage of Element Solved  
(Noble Metal Elements)



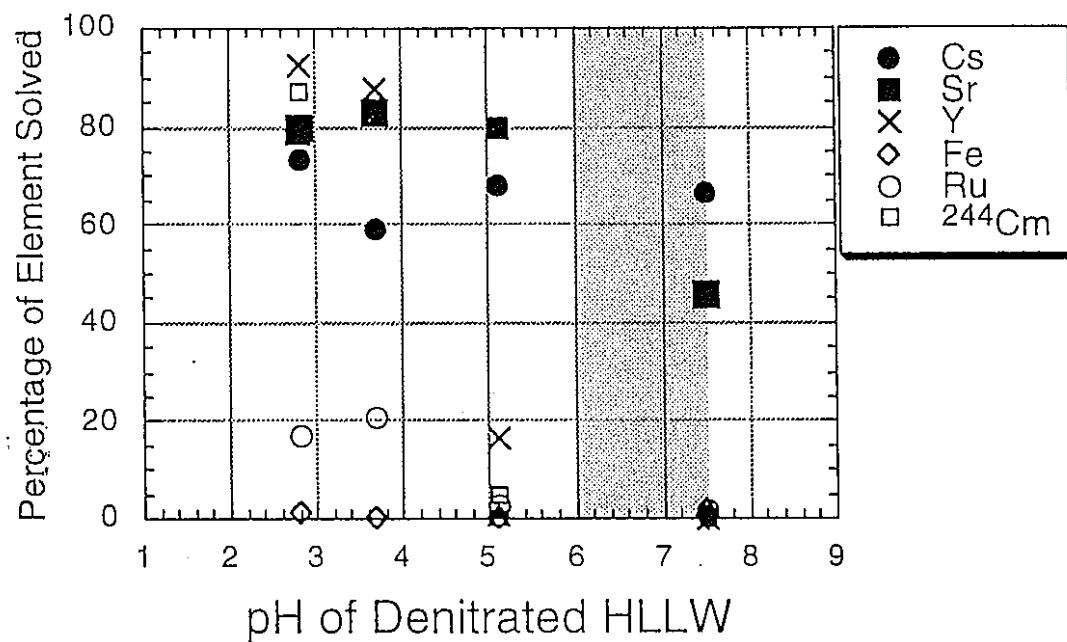
Relationship between pH of Denitrated HLLW and  
Percentage of Element Solved  
(Corrosion Products)

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Relationship between pH of Denitrated HLLW and  
Percentage of Element Solved  
(Zr,Mo)

# Conclusion



Heat-Generating elements are separated by denitration of HLLW

The best pH range = 6 ~ 7.5

$[HCOOH] / [HNO_3] = 2.0$

## 2.8 Characterization of TVF glass

# Characterization of TVF glass

Prepared for  
The 12th Annual KfK-PNC Meeting  
on  
Cooperation in High-Level Waste Management

by KAWAMURA, K.

December 7-9, 1993

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Power Reactor and Nuclear Fuel  
Development Corporation

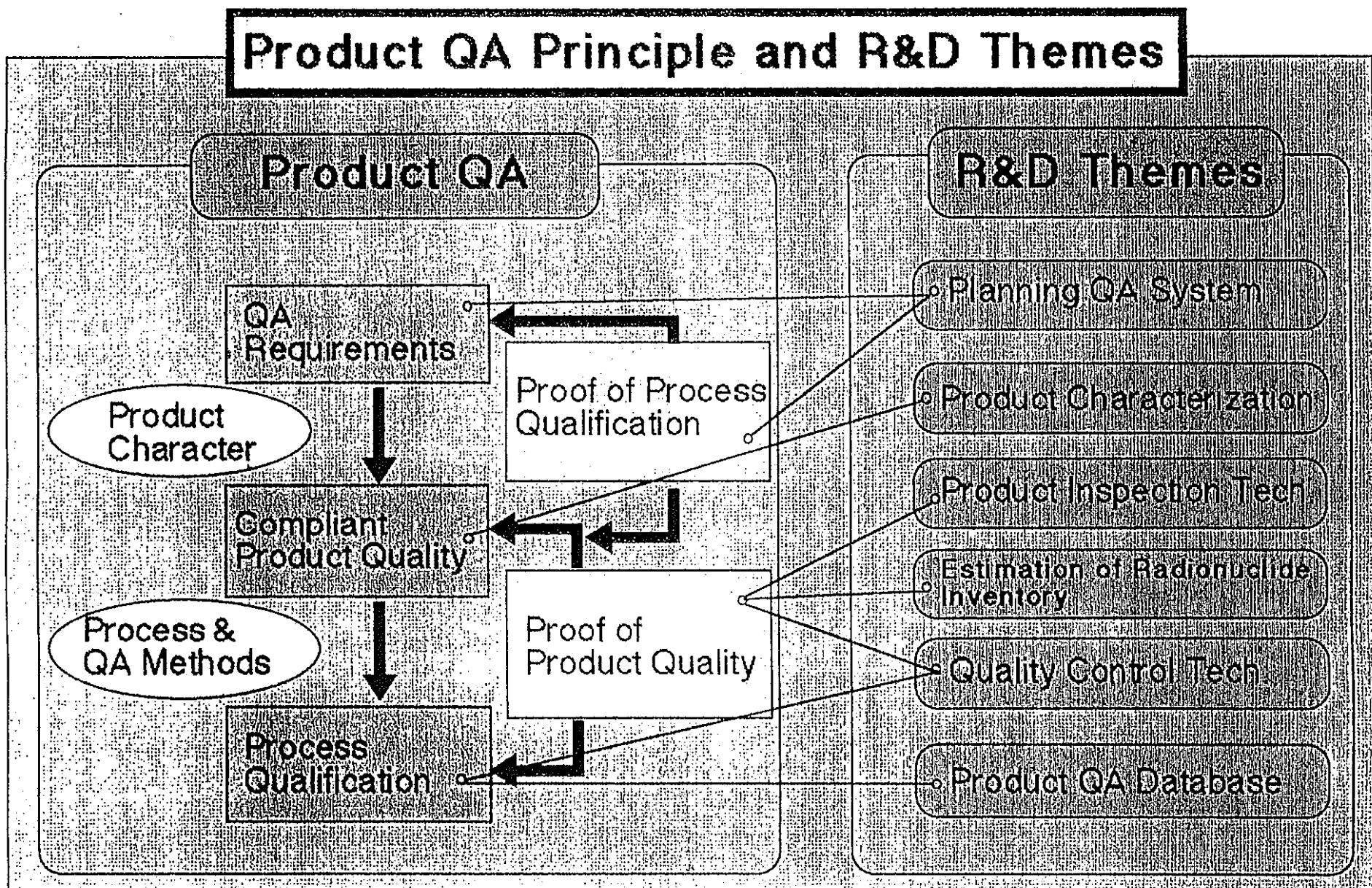


# CONTENTS

1. Product quality assurance
2. Characterization of TVF glass

# PRODUCT QUALITY ASSURANCE

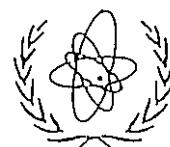
- QA Principle and R&D themes
- QA Requirements & methods
- Estimation of radionuclide inventory



IAEA-TECDOC-680

*Quality assurance  
requirements and methods for  
high level waste package acceptability*

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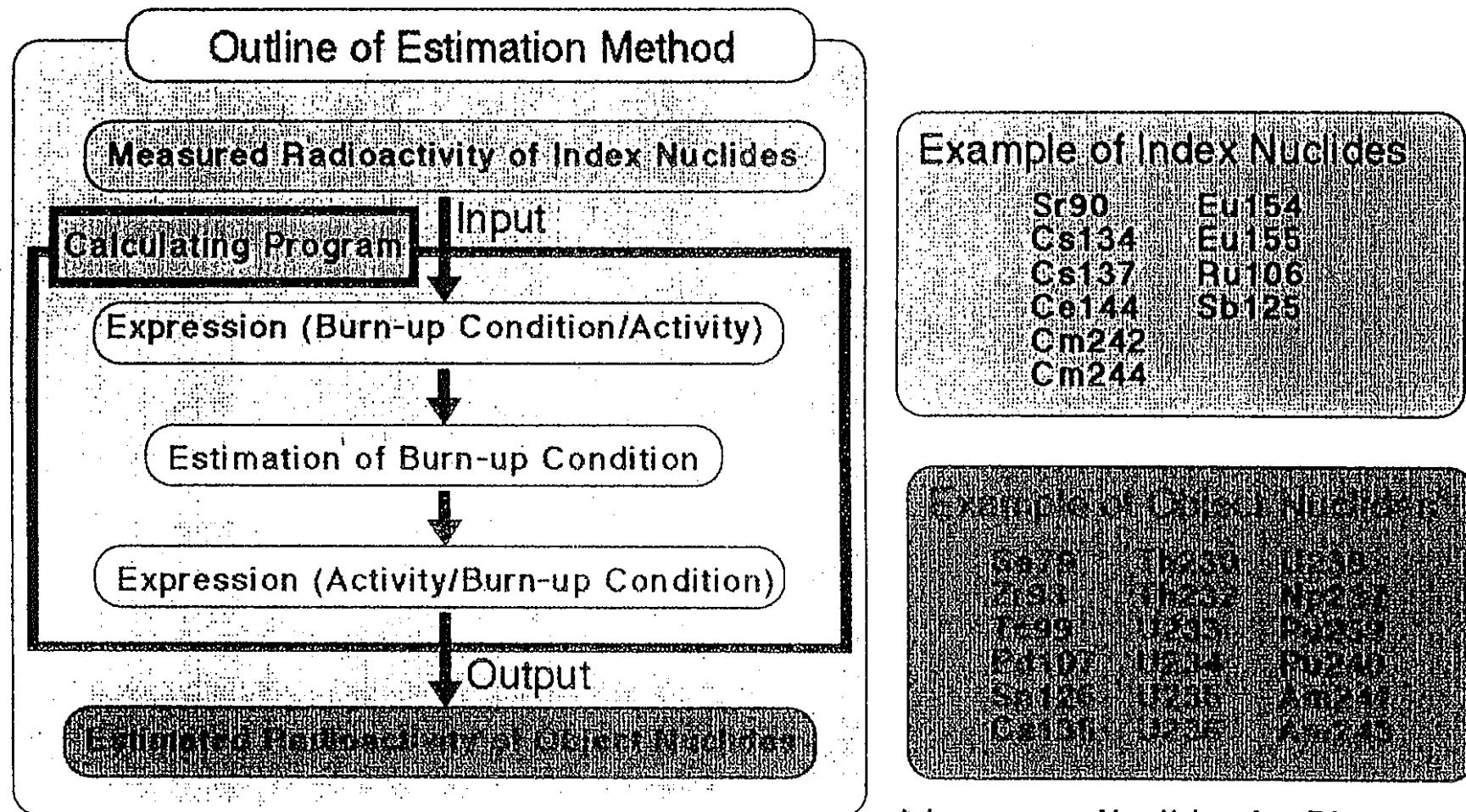


INTERNATIONAL ATOMIC ENERGY AGENCY

IAEA

December 1992

# Estimation of Radionuclide Inventory



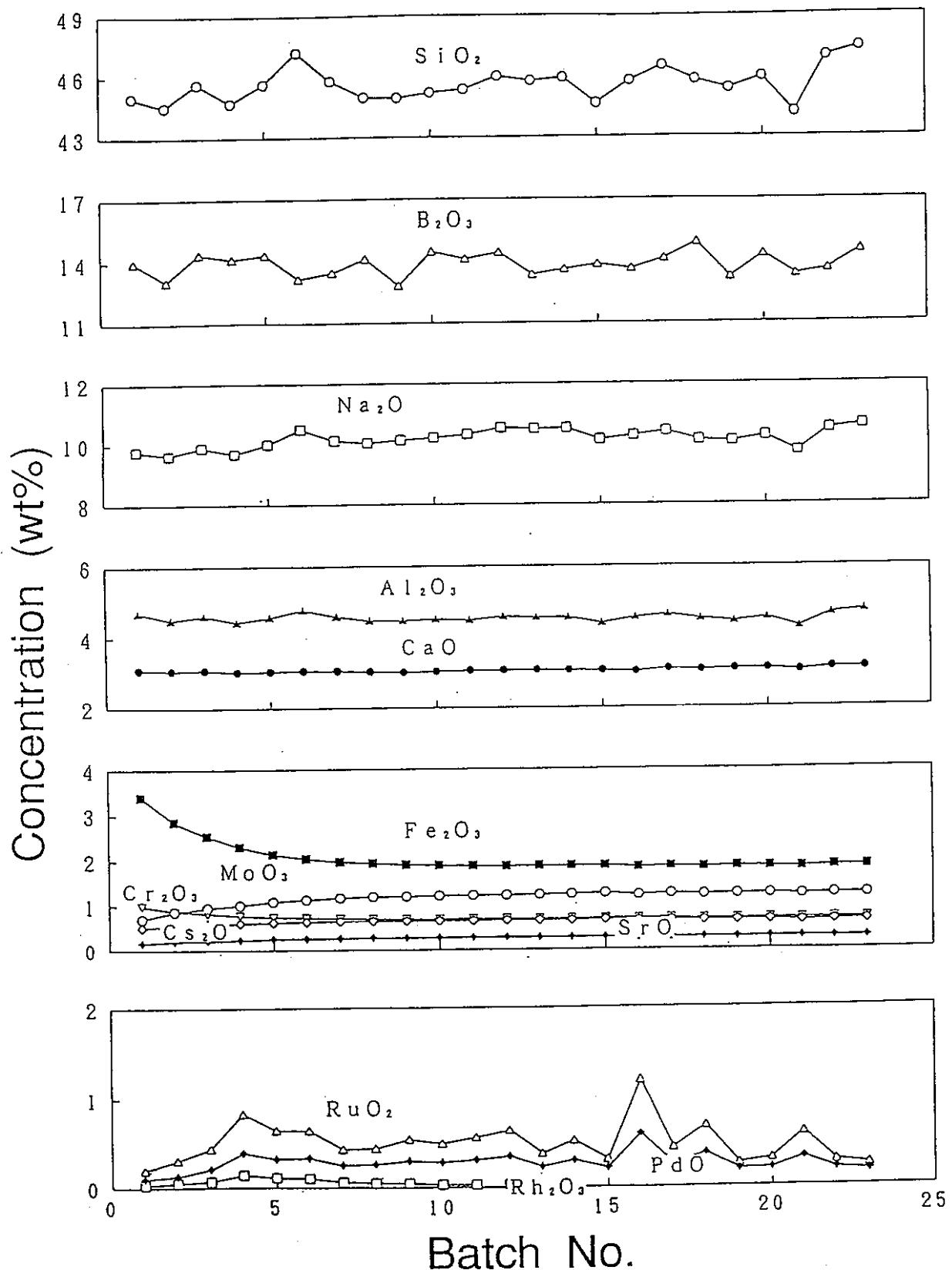
\* Important Nuclides for Disposal  
(PNC '92 Report)

# CHARACTERIZATION OF TVF GLASS

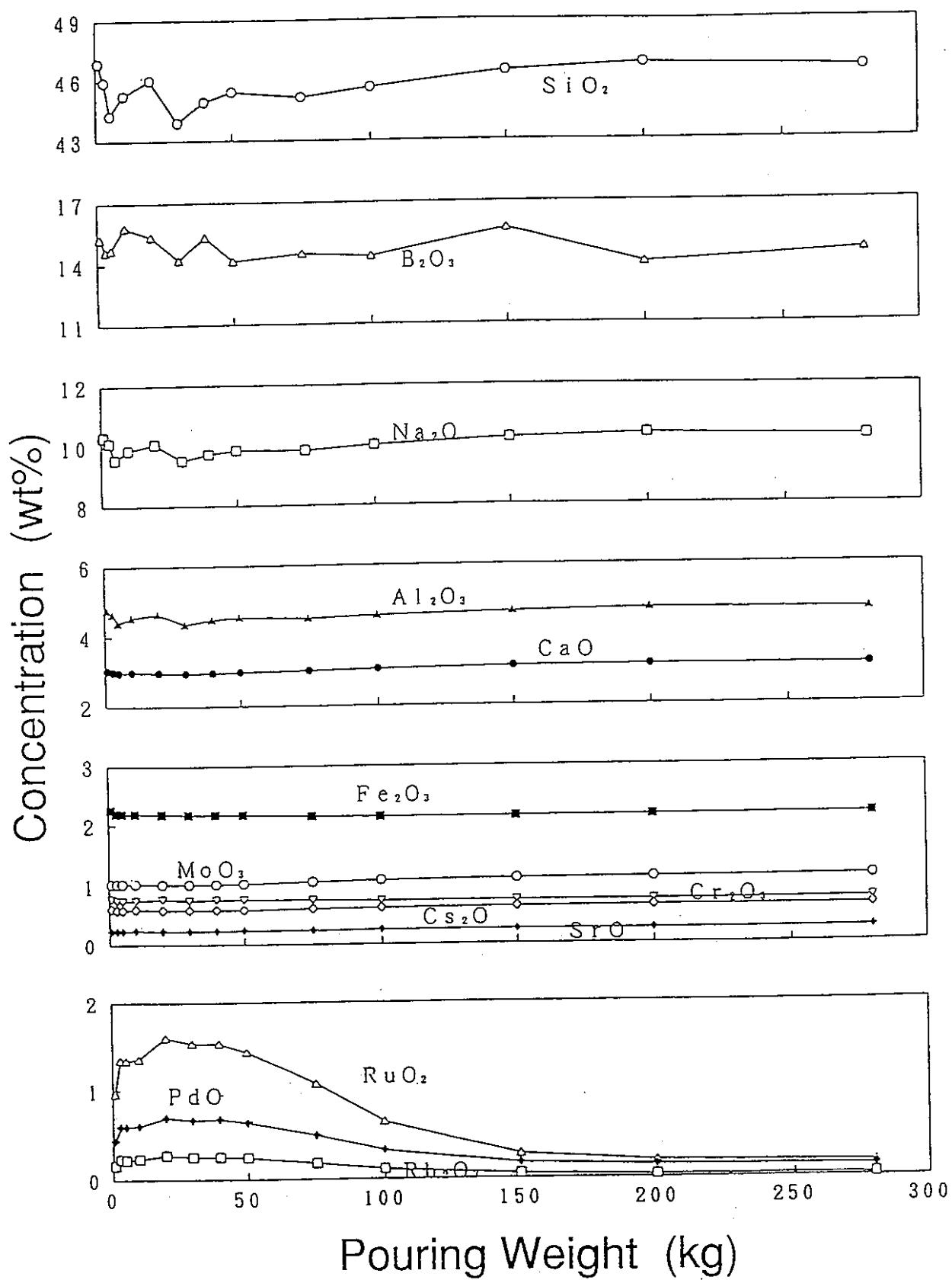
- Homogeneity
  - Composition etc. (XRF, XRD, EPMA)
- Character
  - Viscosity, E. Resistivity, Density, T. Expansion, T. Conductivity, Tg, Ts, Leaching Rate

## Target Composition of TVF Glass (2nd Campaign)

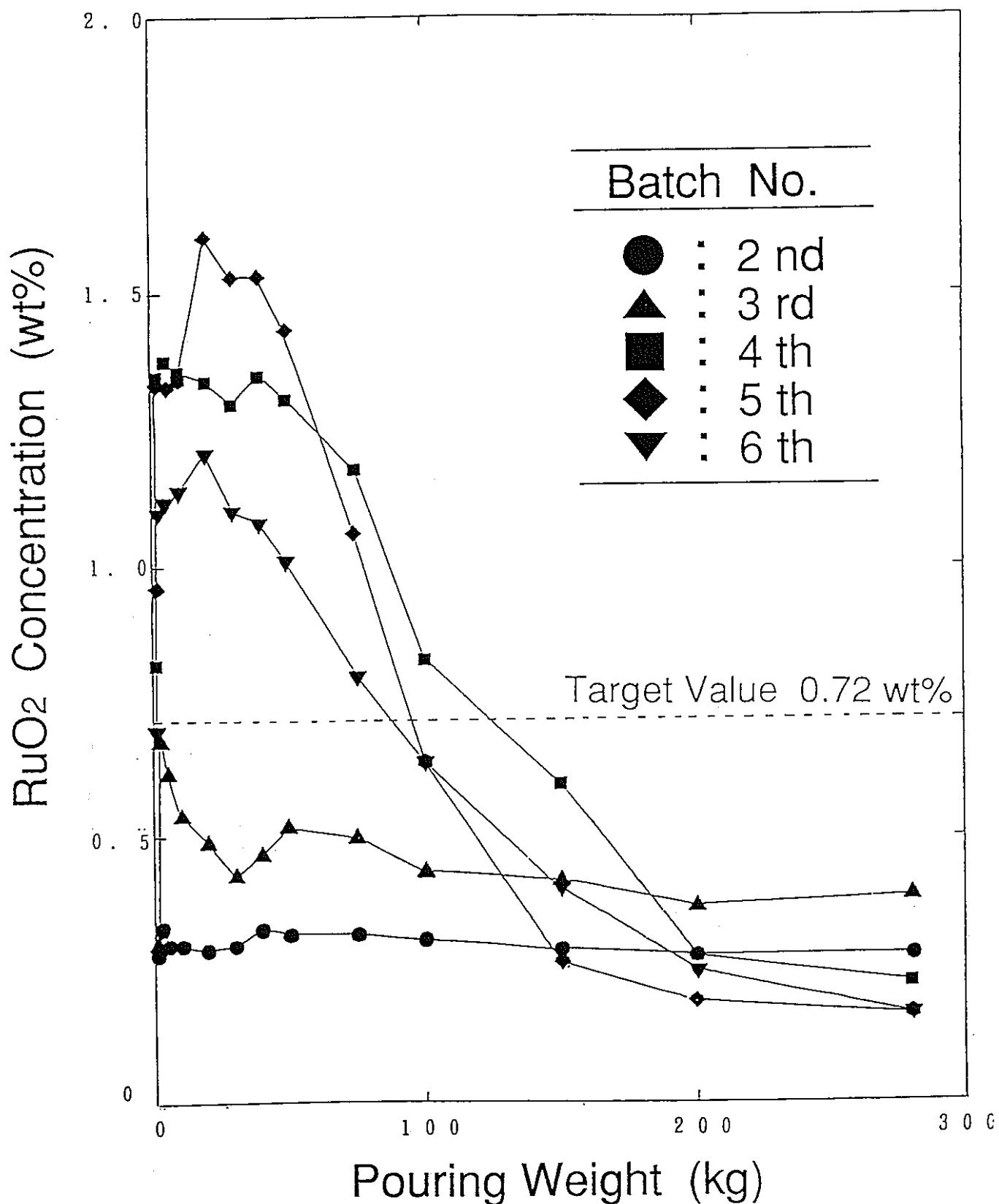
Component	wt%	Component	wt%
SiO <sub>2</sub>	46.73	ZrO <sub>2</sub>	1.41
B <sub>2</sub> O <sub>3</sub>	14.25	MoO <sub>3</sub>	1.41
Li <sub>2</sub> O	3.00	MnO <sub>2</sub>	0.37
CaO	3.00	RuO <sub>2</sub>	0.72
ZnO	3.00	Rh <sub>2</sub> O <sub>3</sub>	0.16
Al <sub>2</sub> O <sub>3</sub>	5.03	PdO	0.34
		Ag <sub>2</sub> O	0.01
Na <sub>2</sub> O	9.69	CdO	0.02
P <sub>2</sub> O <sub>5</sub>	0.29	SnO <sub>2</sub>	0.02
Fe <sub>2</sub> O <sub>3</sub>	1.92	SeO <sub>2</sub>	0.01
Cr <sub>2</sub> O <sub>3</sub>	0.51	TeO <sub>2</sub>	0.15
NiO	0.45	R.E.	5.84
Rb <sub>2</sub> O	0.11	Total	100.01
Cs <sub>2</sub> O	0.75		
SrO	0.30		
BaO	0.52		



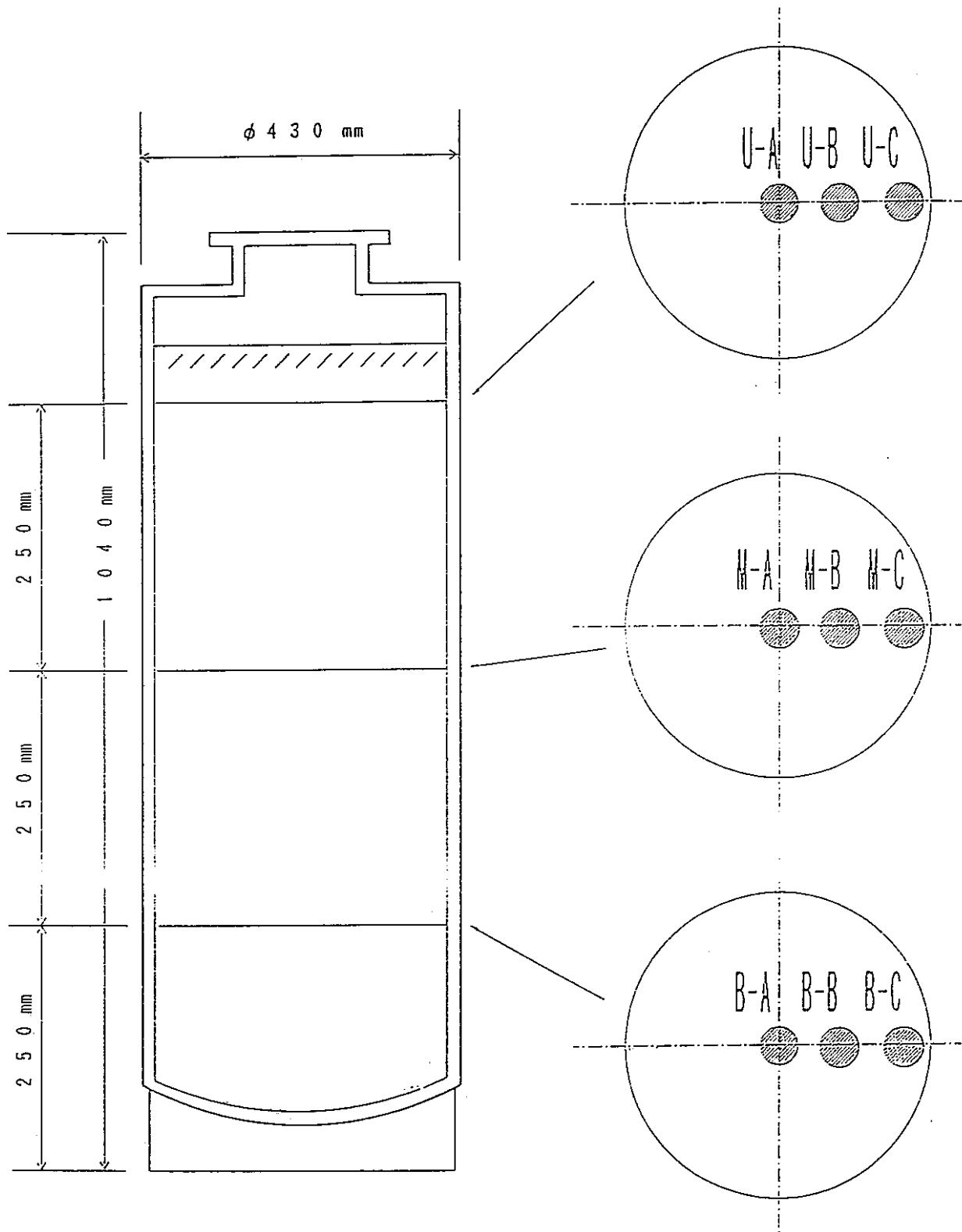
Composition of 100kg-Pouring  
Glass (TVF 2nd Campaign, XRF).



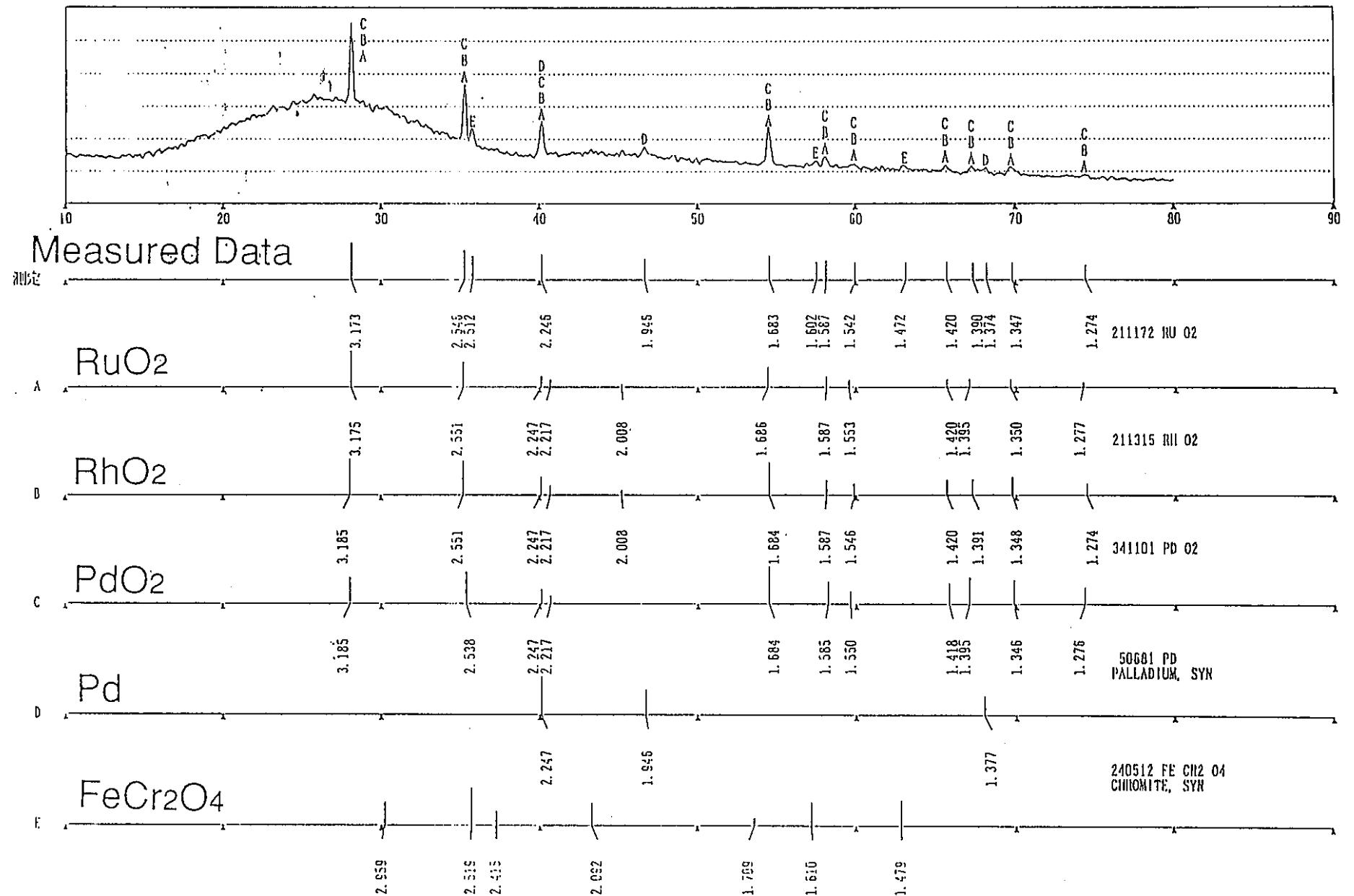
Composition of Pouring Glass  
(TVF 2nd Campaign, 5th Batch,  
XRF).



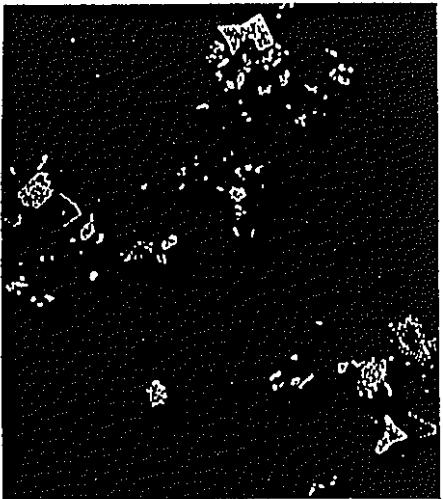
RuO<sub>2</sub> Concentration of Pouring Glass  
(TVF 2nd Campaign, 2nd-6th Batch,  
XRF).



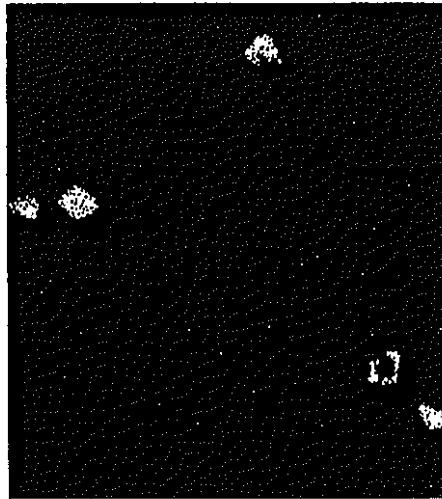
Sampling Point



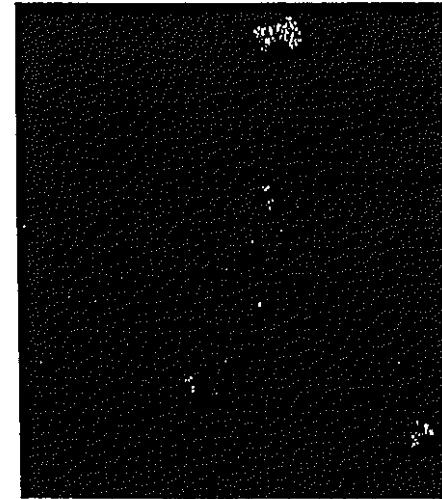
XRD Data of Glass Sampled at M-A  
(TVF 2nd Campaign, 6th Batch).



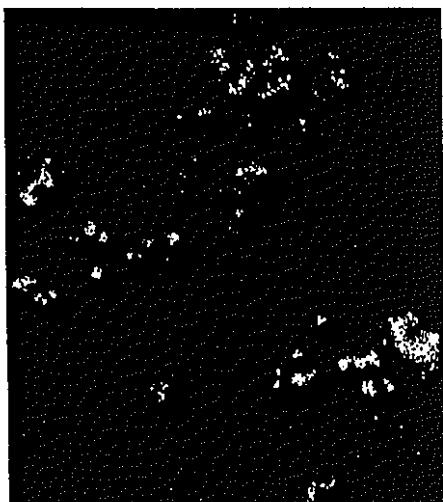
SEI  $10 \mu\text{m}$



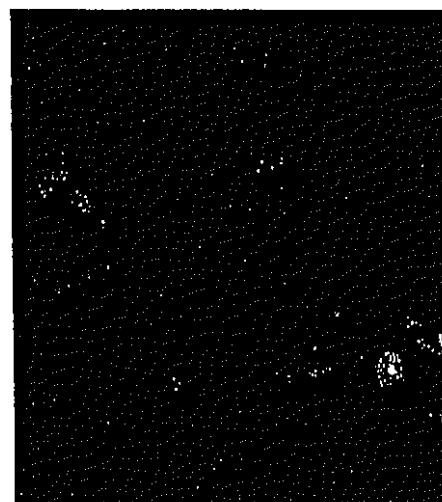
Cr



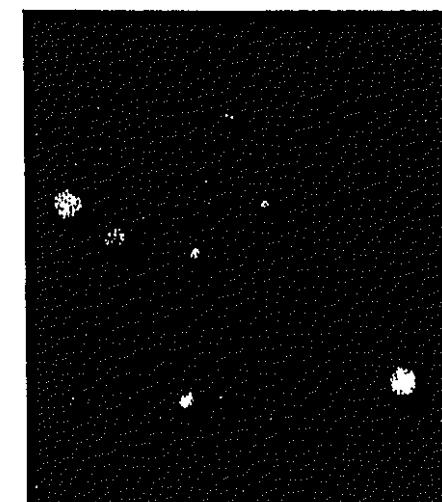
Ce



Ru

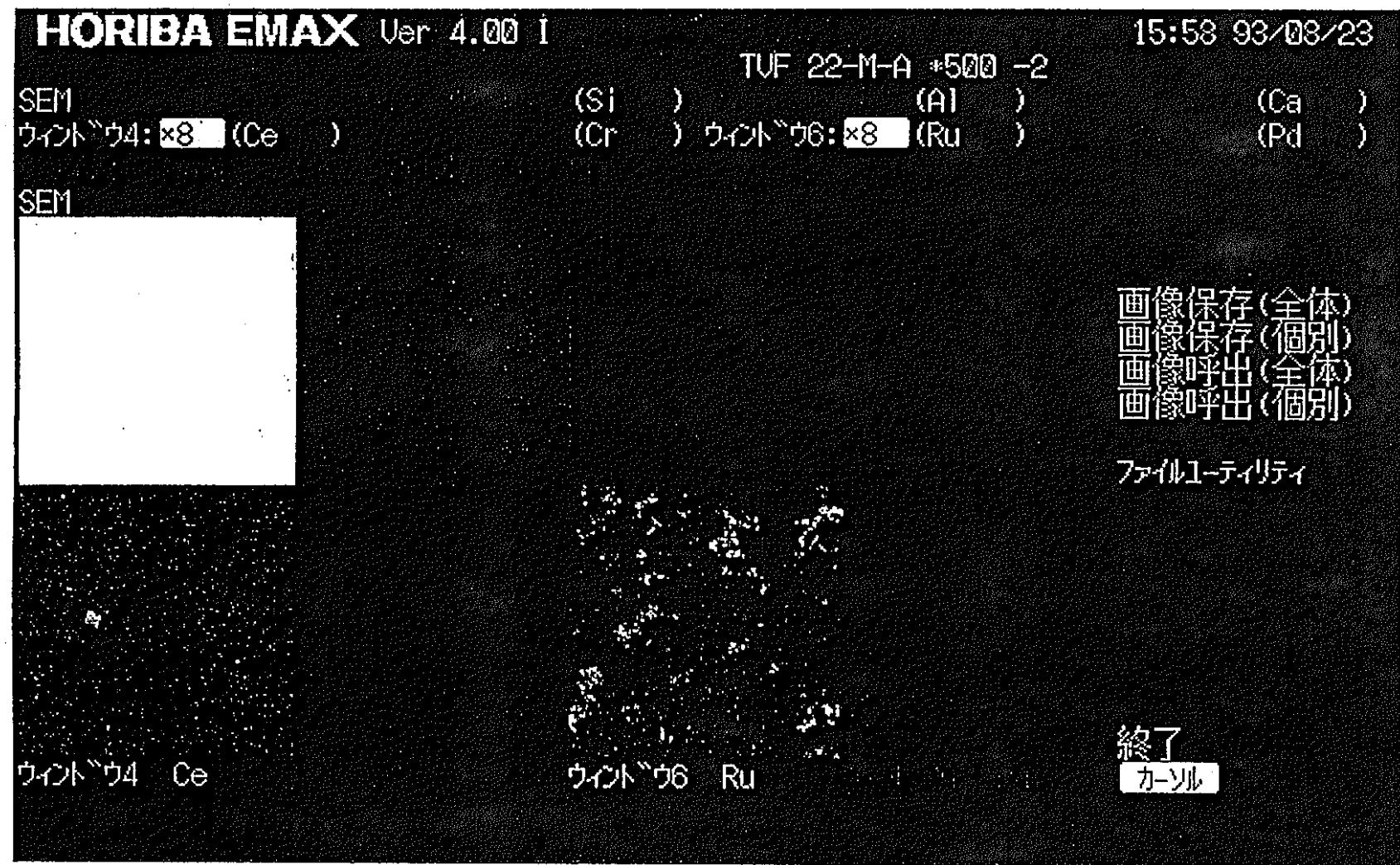


Rh



Pd

Secondary Electron Image and Characteristic  
X-ray Image of Glass Sampled at M-A  
(TVF 2nd Campaign, 6th Batch).



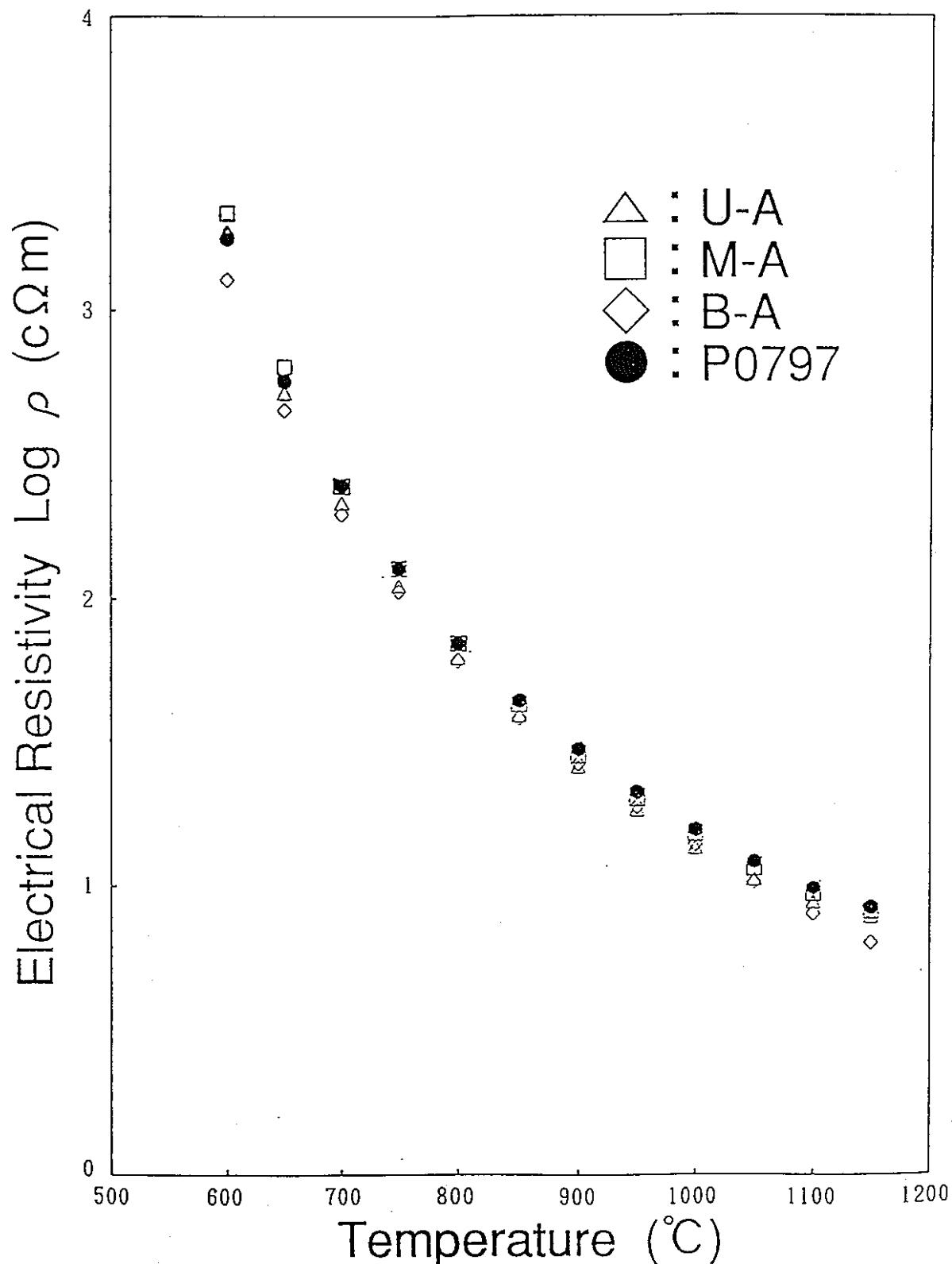
Secondary Electron Image and  
Characteristic X-ray Image of Glass  
Sampled at M-A (TUF 2nd Campaign, 6th batch)

Platinum Metal Content and Density  
of Glasses Sampled at U, M, B  
(TVF 2nd Campaign, 6th Batch)

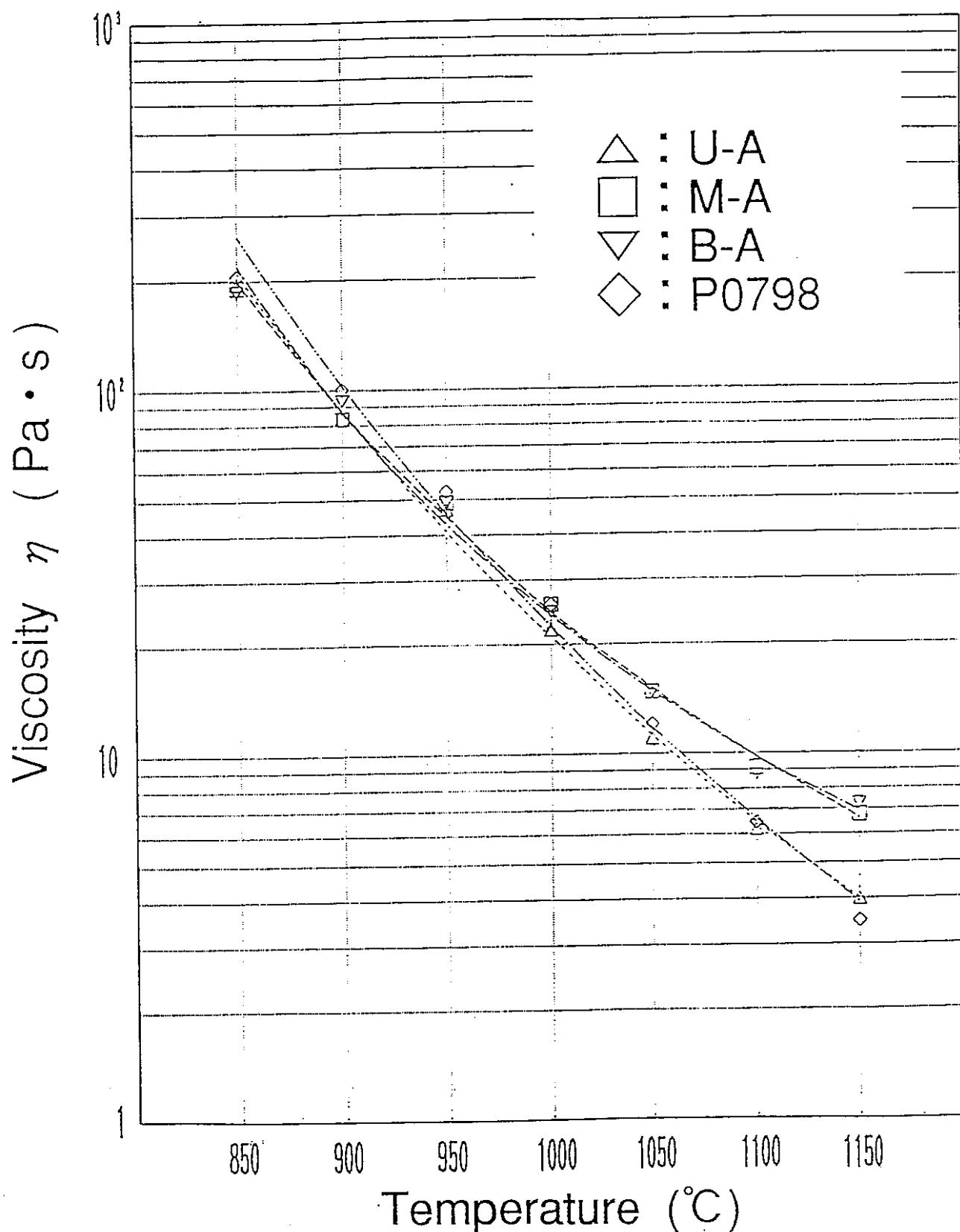
	U	M	B
Platinum Metal Content (wt %)			
RuO <sub>2</sub>	0. 44	0. 26	0. 95
Rh <sub>2</sub> O <sub>3</sub>	0. 05	0. 04	0. 15
PdO	0. 25	0. 17	0. 45
Total	0. 74	0. 47	1. 55
Density ( $\times 10^3$ kg/m <sup>3</sup> )	2. 754	2. 756	2. 763

Characterization of Glasses  
Sampled at U-A, M-A, B-A  
(TVF 2nd Campaign, 6th Batch)

	U - A	M - A	B - A
Density ( $\times 10^3 \text{ kg/m}^3$ )	2.75	2.77	2.77
Thermal expansion ( $\times 10^{-7}/\text{°C}$ )			
Temp. range (30 ~ 300 °C)	84	87	86
Tg (Transformation point, °C)	499	505	503
Ts (Softening point, °C)	613	612	615
Thermal conductivity (W/mK, at R. T.)	0.95	1.00	1.05
Leaching rate (Weight loss) ( $\times 10^{-4} \text{ kg/m}^2 \text{ d}$ )			
J I S   T y p e	2.9	2.6	2.9
M C C - 1   M e t h o d	2.2	2.3	2.1



Electrical Resistivity of Glasses  
Sampled at U-A, M-A, B-A and  
Standard Glass(TVF 2nd Campaign,  
6th Batch).



Viscosity of Glasses Sampled at  
U-A, M-A, B-A and Standard Glass  
(TVF 2nd Campaign, 6th Batch,  
Shear Rate 0.14s<sup>-1</sup>).

# SUMMARY

- Process qualification assures glass product quality.
- Homogeneity :  
Most elements distribute uniformly in vitrified glass.  
Platinum metal crystals [(Ru,Rh)O<sub>2</sub>, Pd-Rh-Te] distribute in the lower part of vitrified glass.
- Character :  
Vitrified glass has uniform properties those are not remarkably affected by distribution of platinum metal crystals.

## 2.9 Characterization of HLLW

# Characterization of HLLW

- 6.1 -

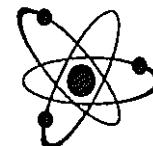
Prepared for  
The 12th Annual KfK-PNC Meeting  
on  
Cooperation in High-Level Waste Management

by YAMASHITA, T.

December 7-9, 1993

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Power Reactor and Nuclear Fuel  
Development Corporation



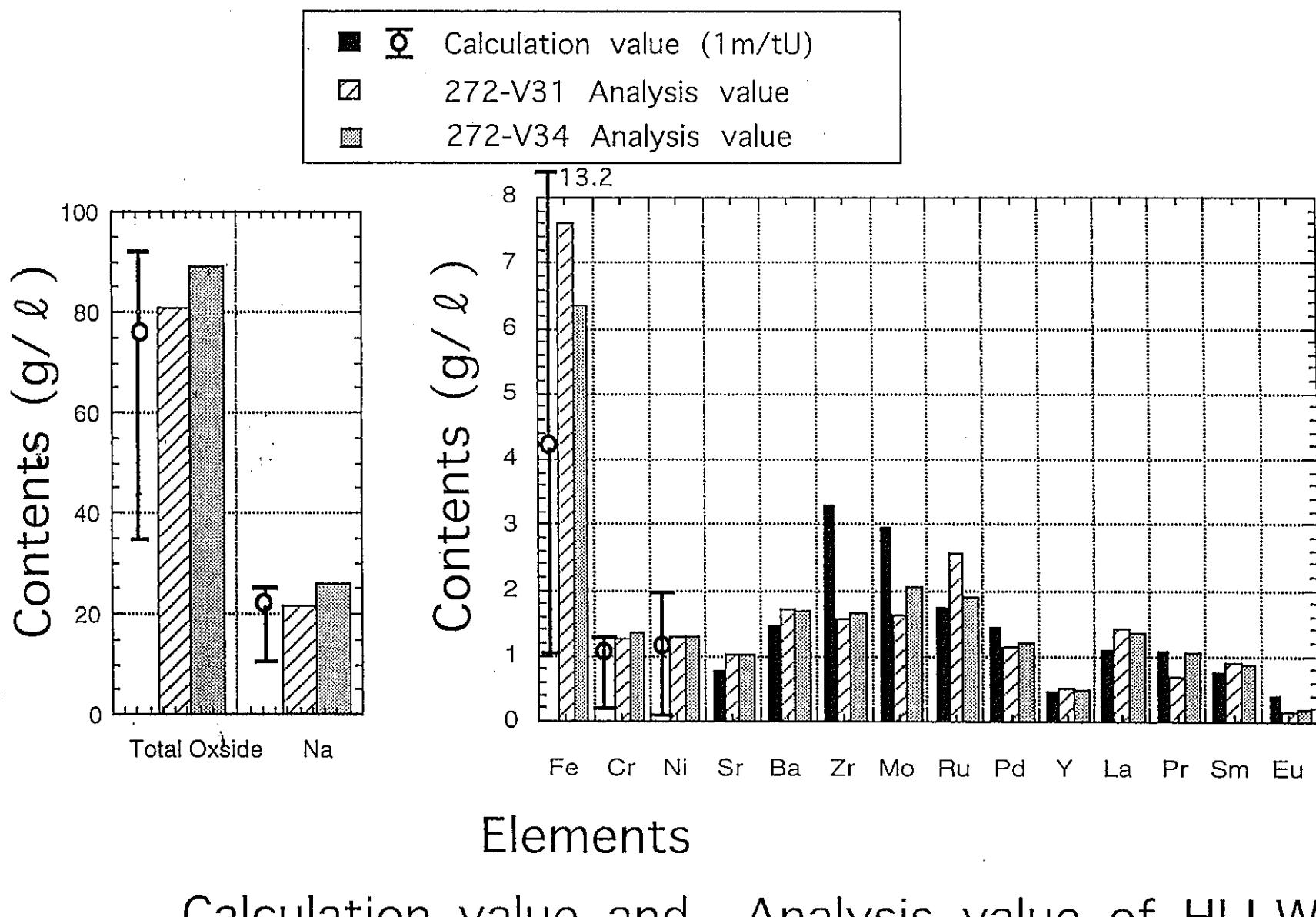
**PNC**

# Heading

1. Chemical Analysis of HLLW
2. Volume and Composition of Precipitate
3. Formation of Precipitate in HLLW

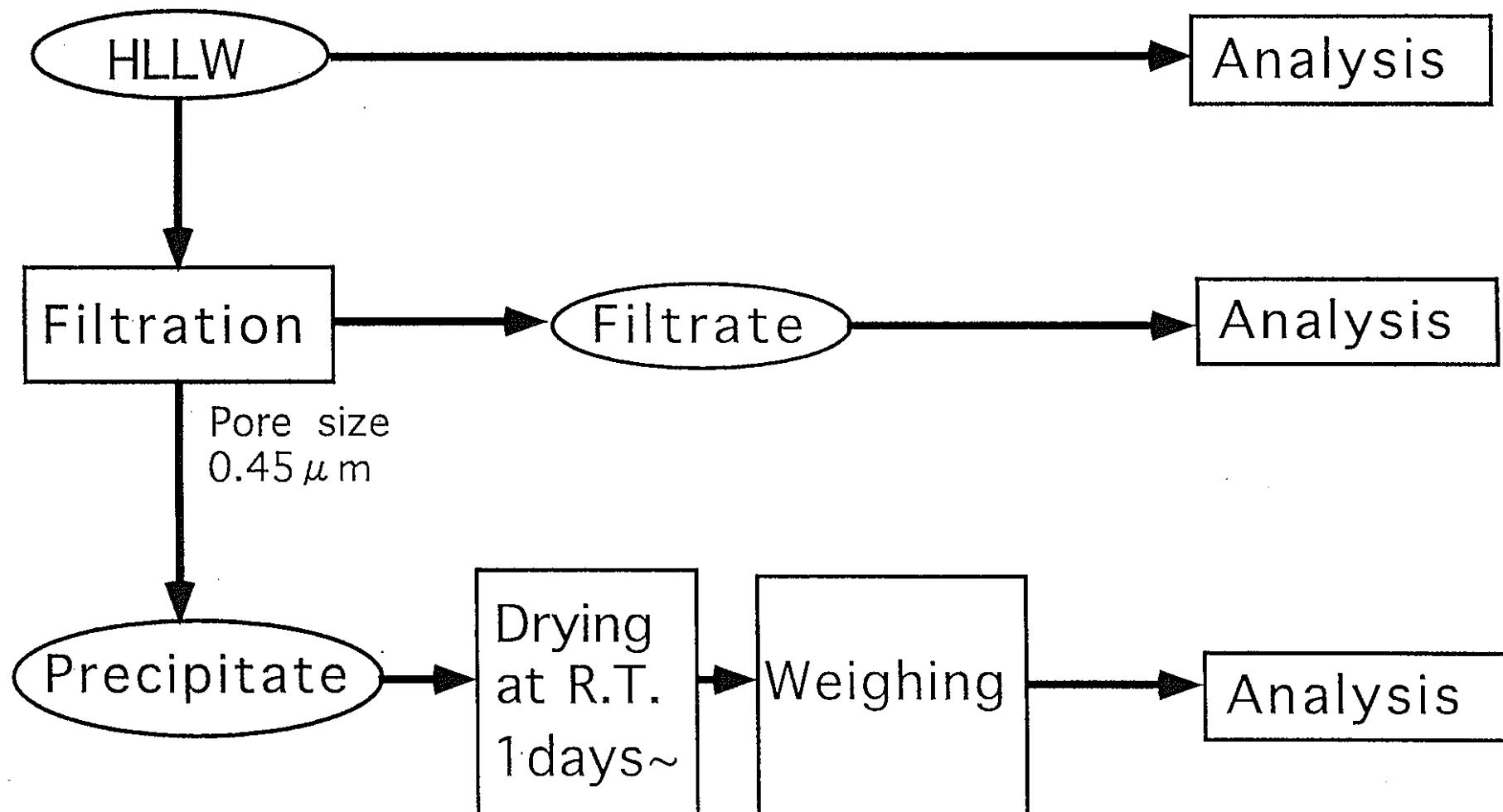
## Received HLLW at CPF

Times	HLLW Volume	Radio-activity	Remarks
1st	12 ℥	110 TBq	
2nd	15 ℥	190 TBq	
3rd	13 ℥	110 TBq	
4th	13 ℥	130 TBq	
5th	13 ℥	58 TBq	
6th	12 ℥	120 TBq	272-V31
7th	12 ℥	70 TBq	272-V31
8th	12 ℥	153 TBq	272-V34
9th	8 ℥	77 TBq	272-V34
10th	9 ℥	96 TBq	272-V31

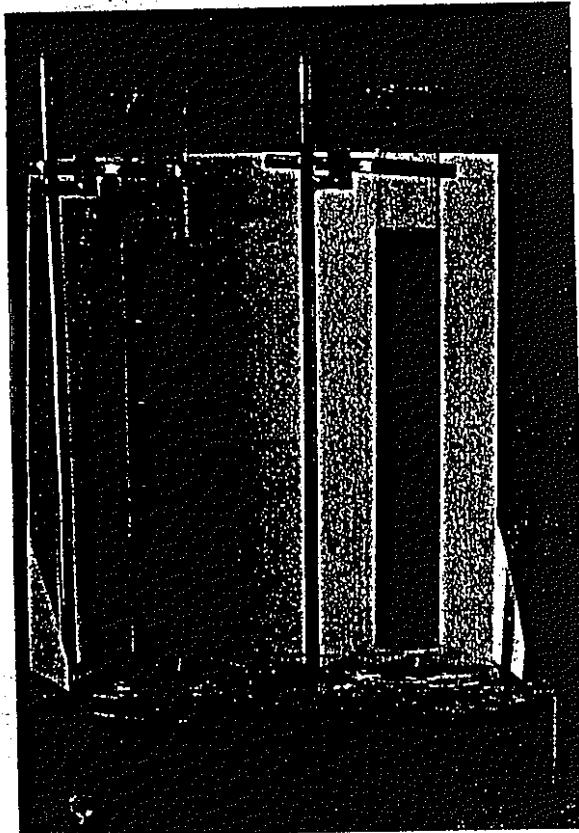


## 2. Volume and Composition of Precipitate

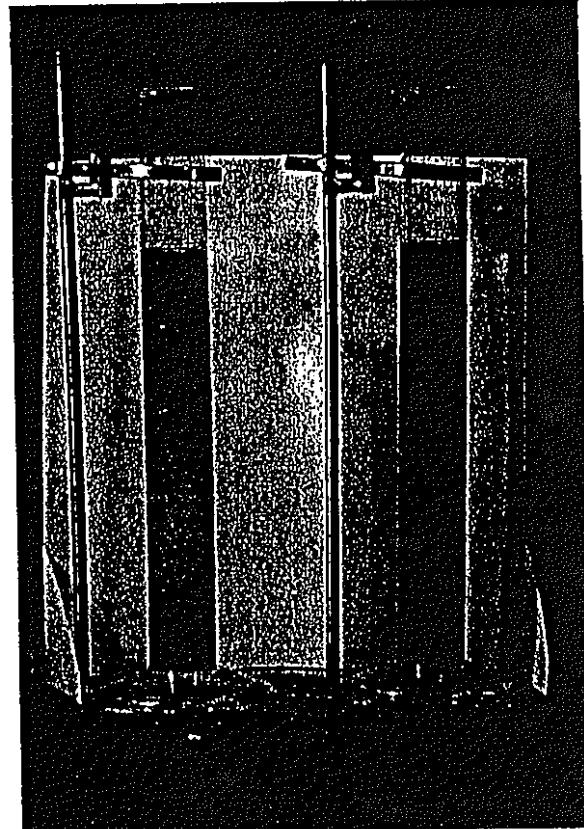
6



Filtration and measurements flow of  
10th Received HLLW (CPF)



0h

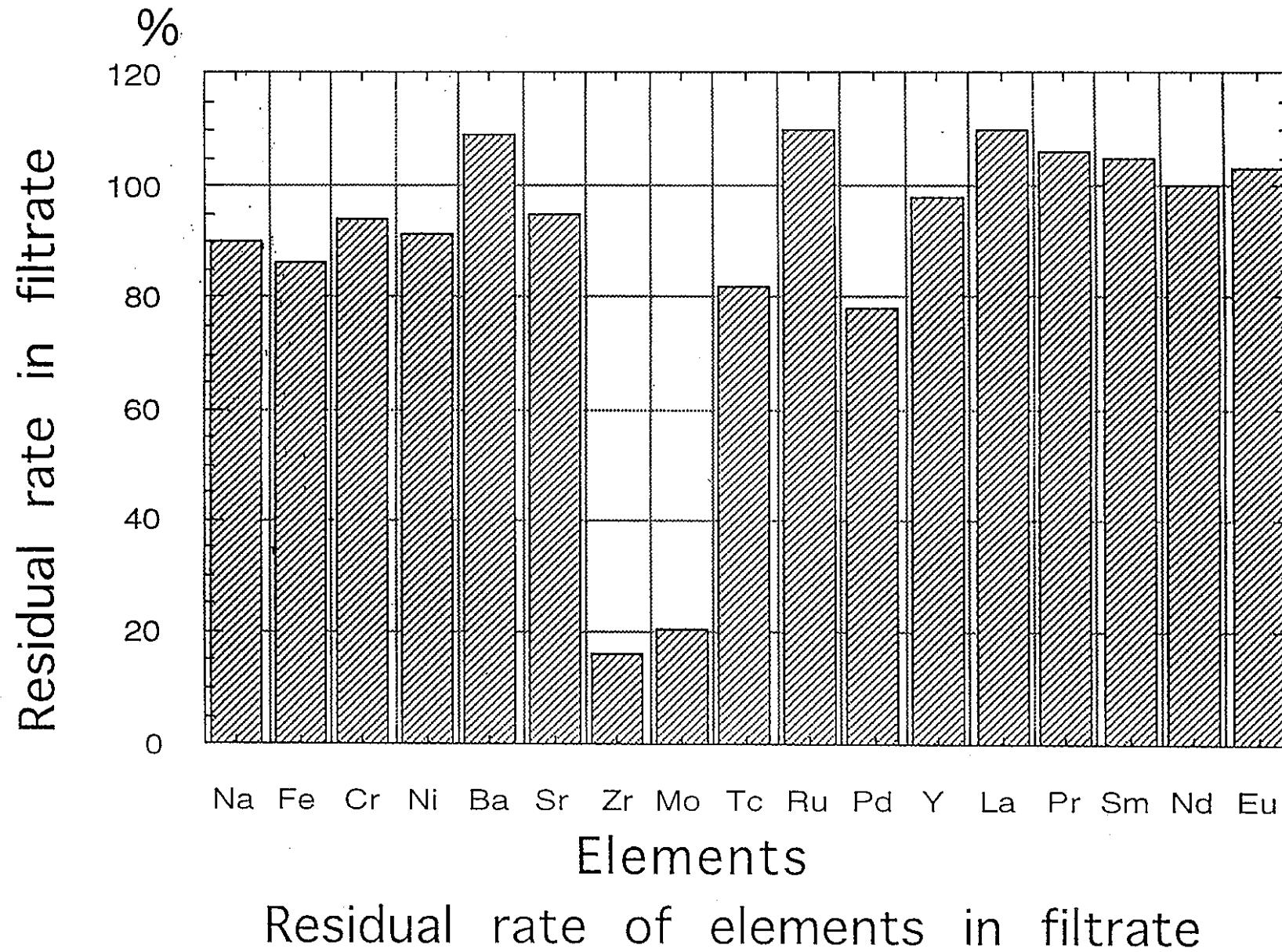


After 24h

HLLW Sedimentation Test at CPF Hot Cell

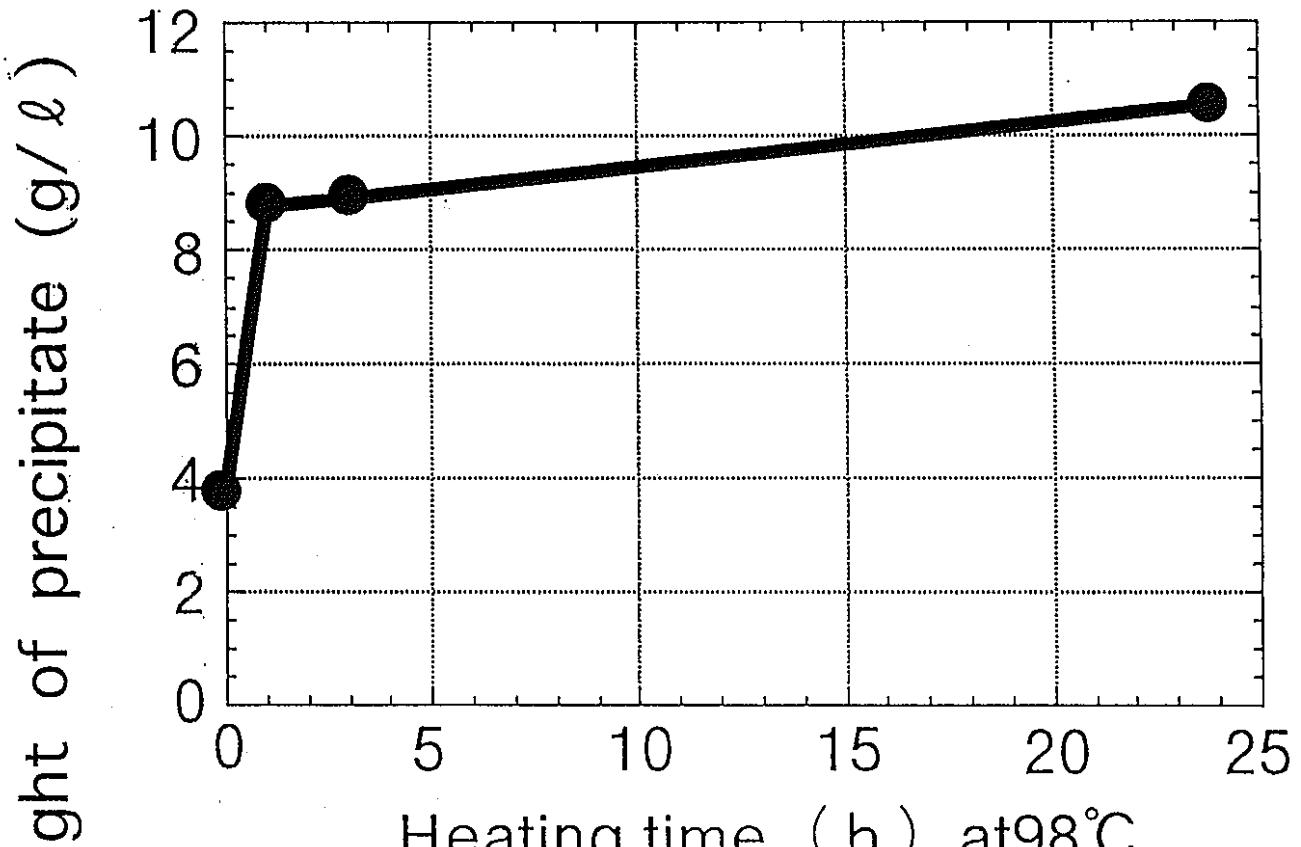
## Precipitate Volume of HLLW

HLLW	sedimentation Volume (after 24h)	Weight
CPF 10th	6.1m l / 50m l	11.6 g / l



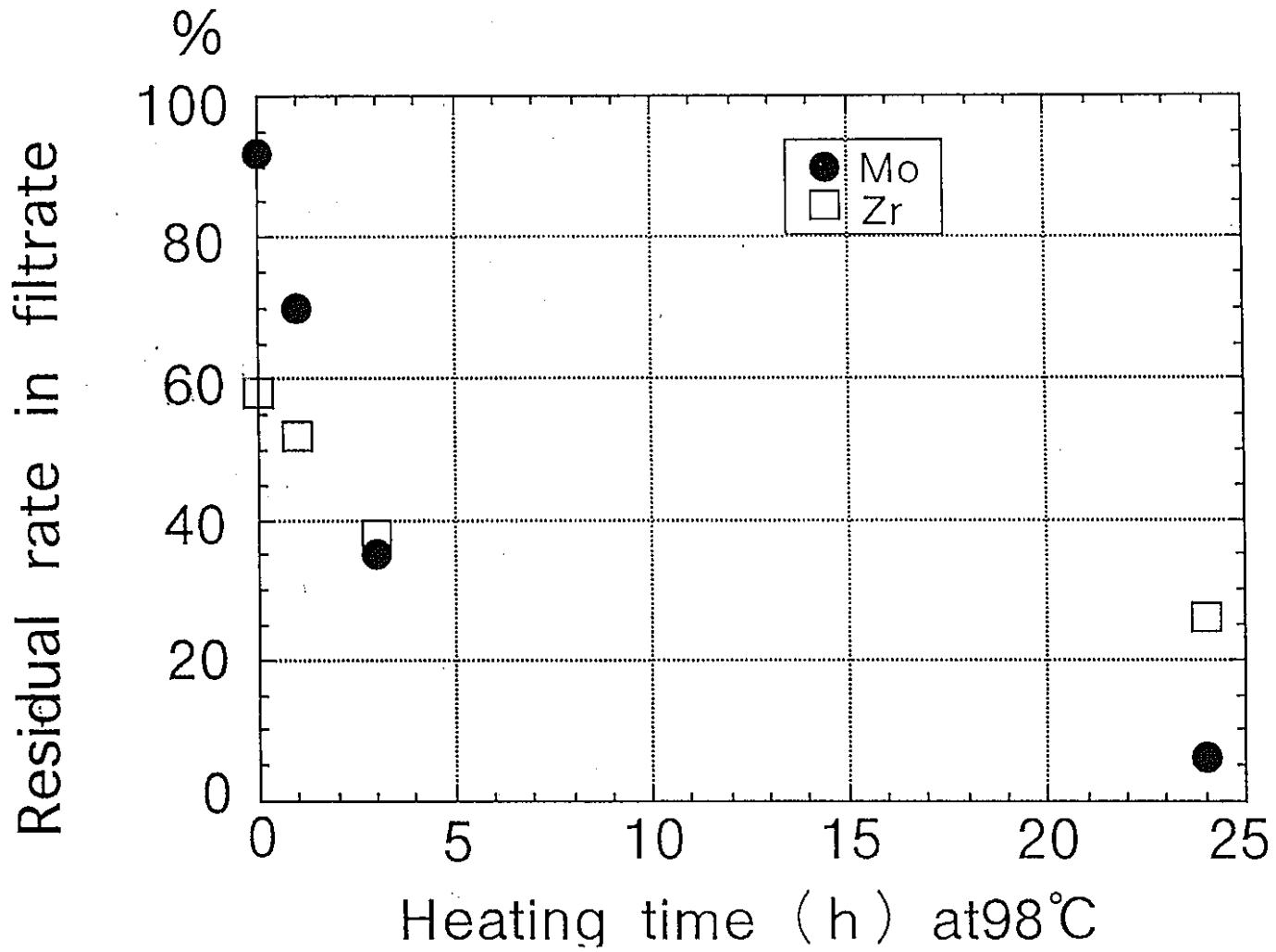
10

### 3. Formation of Precipitate in HLLW

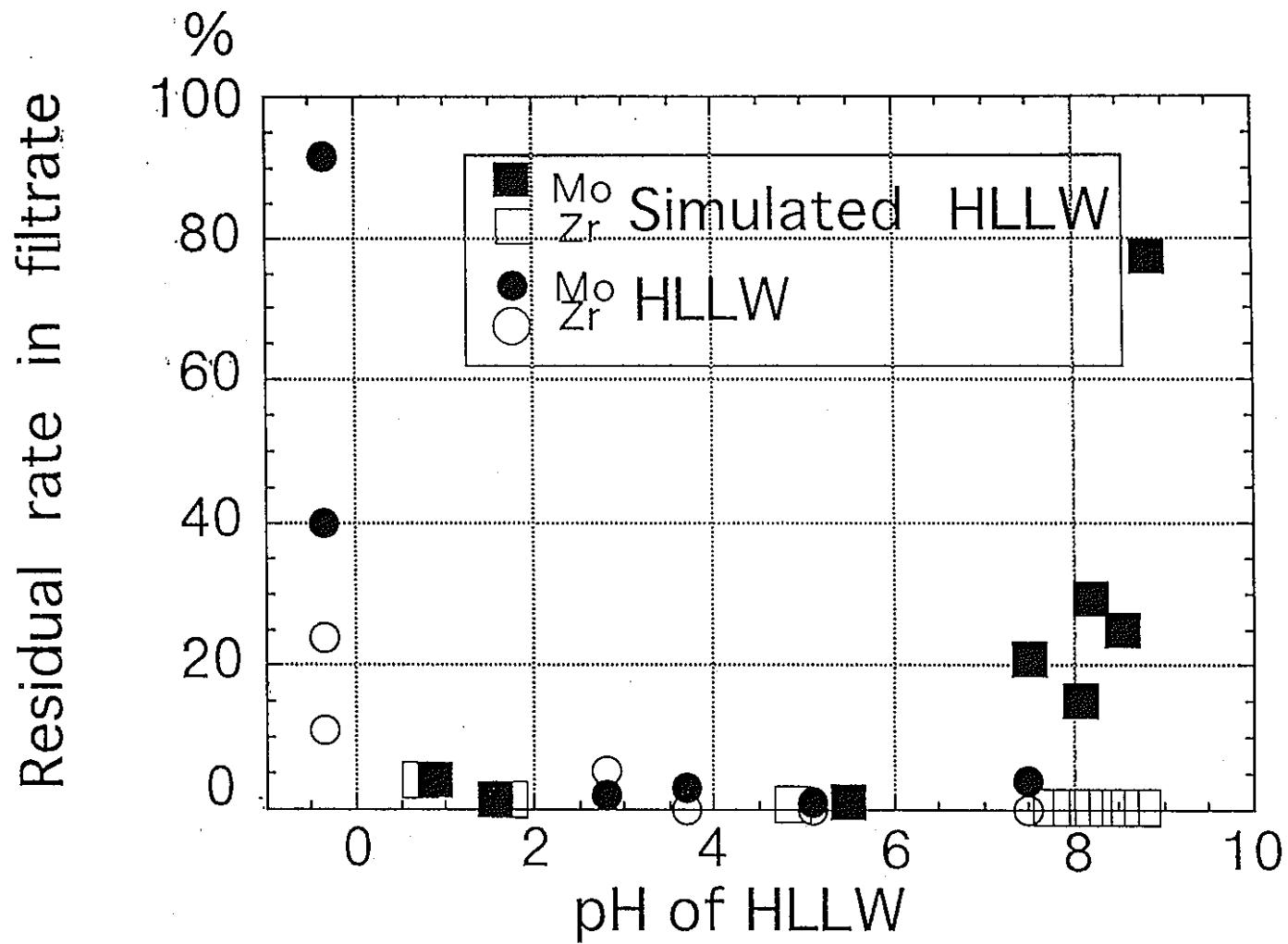


Effects of Heating time  
on Precipitation in HLLW  
(Simulated Waste)

12



Effects of Heating time  
on Mo and Zr Residual Rate in Filtrate  
(Simulated Waste)



Effects of pH on Mo and Zr Residual Rate in Filtrate

## Conclusion

1. HLLW composition is equivalent to calculation value.
2. Most part of Mo and Zr is precipitate in HLLW
3. Formation of precipitate was affected by both heating time and pH.  
Separation of precipitate from HLLW reduce Mo concentration in HLLW.

3 KfK 側の発表資料

3.1 Current status of radioactive waste management in GERMANY

20 LWR on line

23 GWe installed capacity

30-35% Electricity supply

4,000 tons of spent fuel

4,000 ÷ 5,000 m<sup>3</sup> of conditioned wastes/year

until 2000:

150,000 m<sup>3</sup> wastes with negligible heat-production ( $\approx 95\%$ )

3,000 m<sup>3</sup> wastes, heat-producing ( $\approx 15\%$ )

The Russian type reactors (WWER) are presently being assessed with respect to safe operation by the GRS (Gesellschaft für Reaktorsicherheit mbH, Köln).

There is at present no direct participation by public utilities from the old federal states in public utilities in the new federal states.

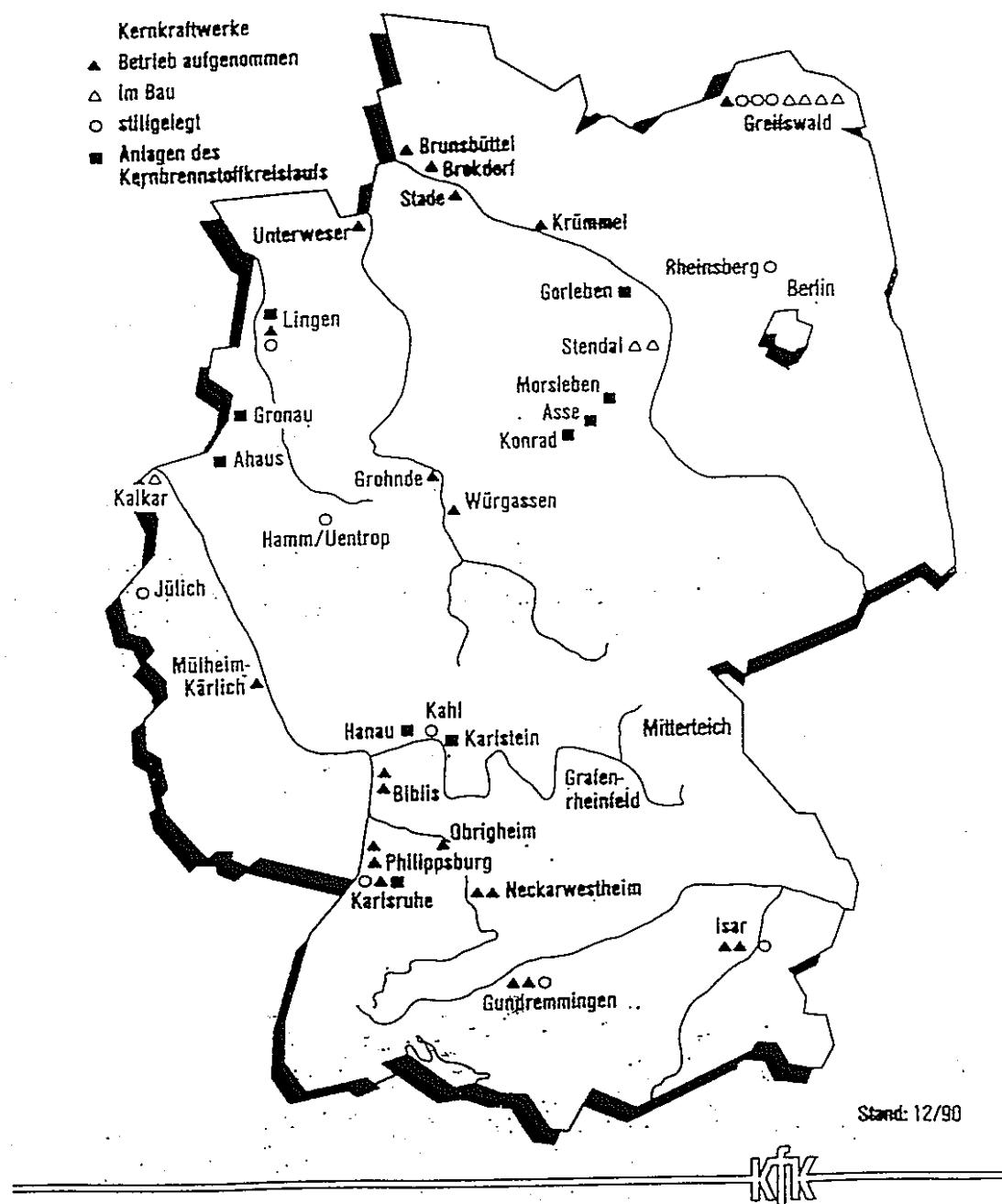


Fig. 1 Nuclear power plants and the facilities of the fuel cycle in the FRG

spent fuel with extremely high burnups or other spent fuel which does not meet the specifications of reprocessing.

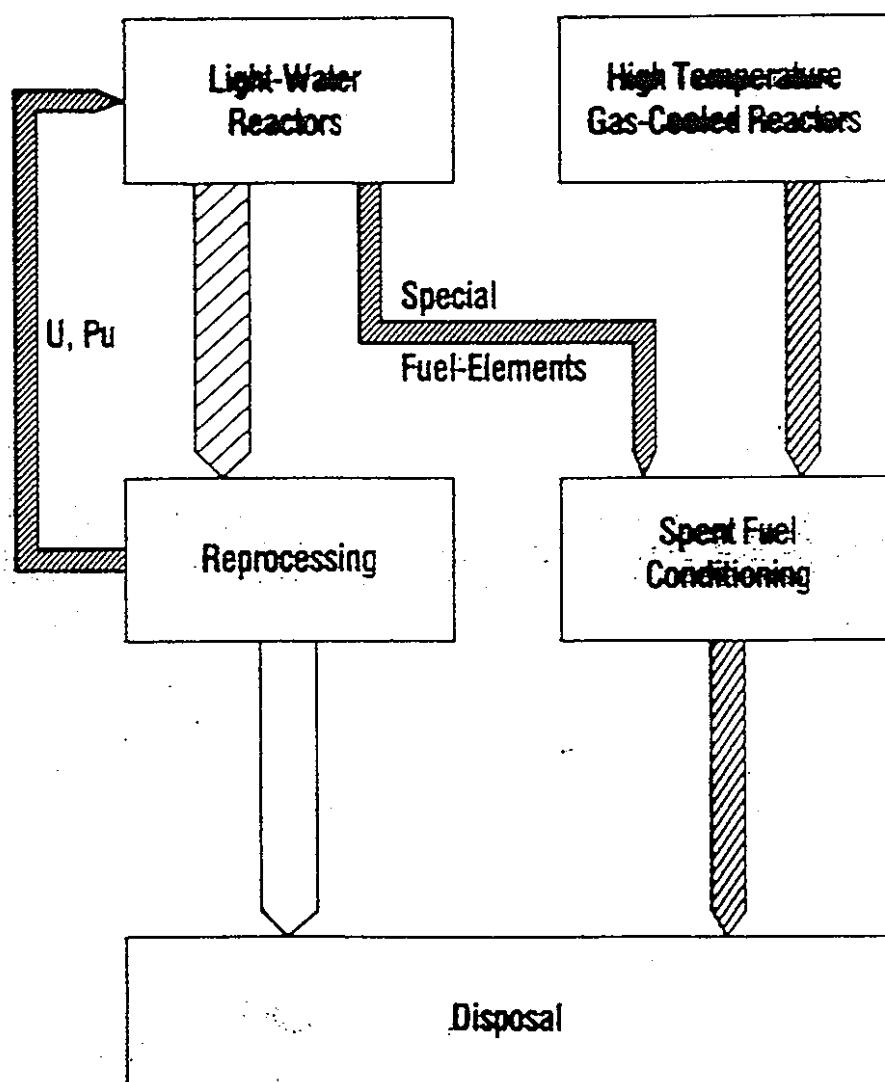


Fig. 2

## Future spent fuel strategy in the FRG

Decommissioning of the WAK Plant

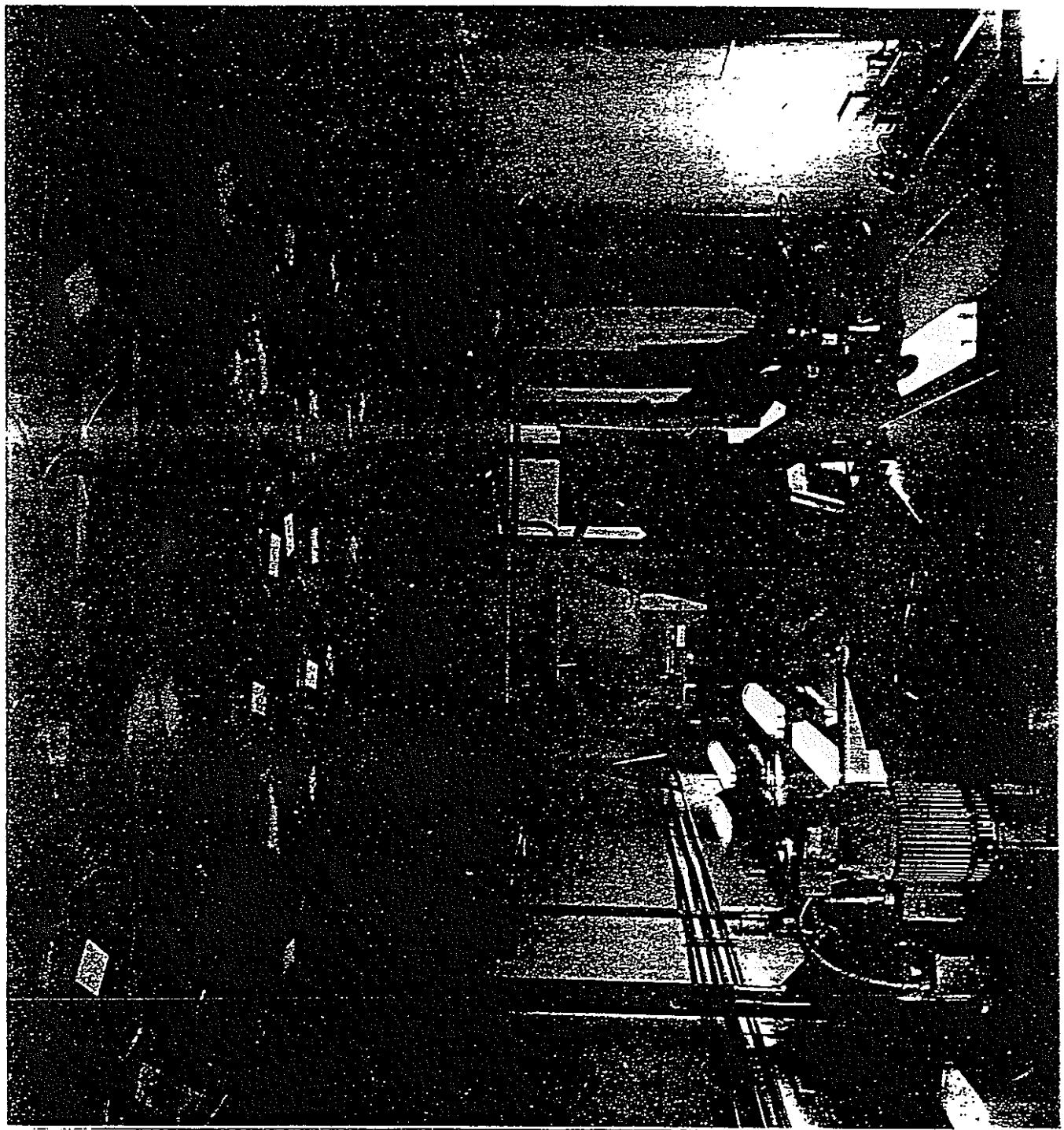
- Plant operation 1970-1991
- 88m<sup>3</sup> HAWC(24Million Ci)generated
- waste to be vitrified in PAMELA plant of Mai/Belgium
- Transport licensing applied, under progress
- Public hearing 1994
- Transportation by train, CASTOR casks
- 30 transportation events necessary
- Returned glass product to be stored in interim storage

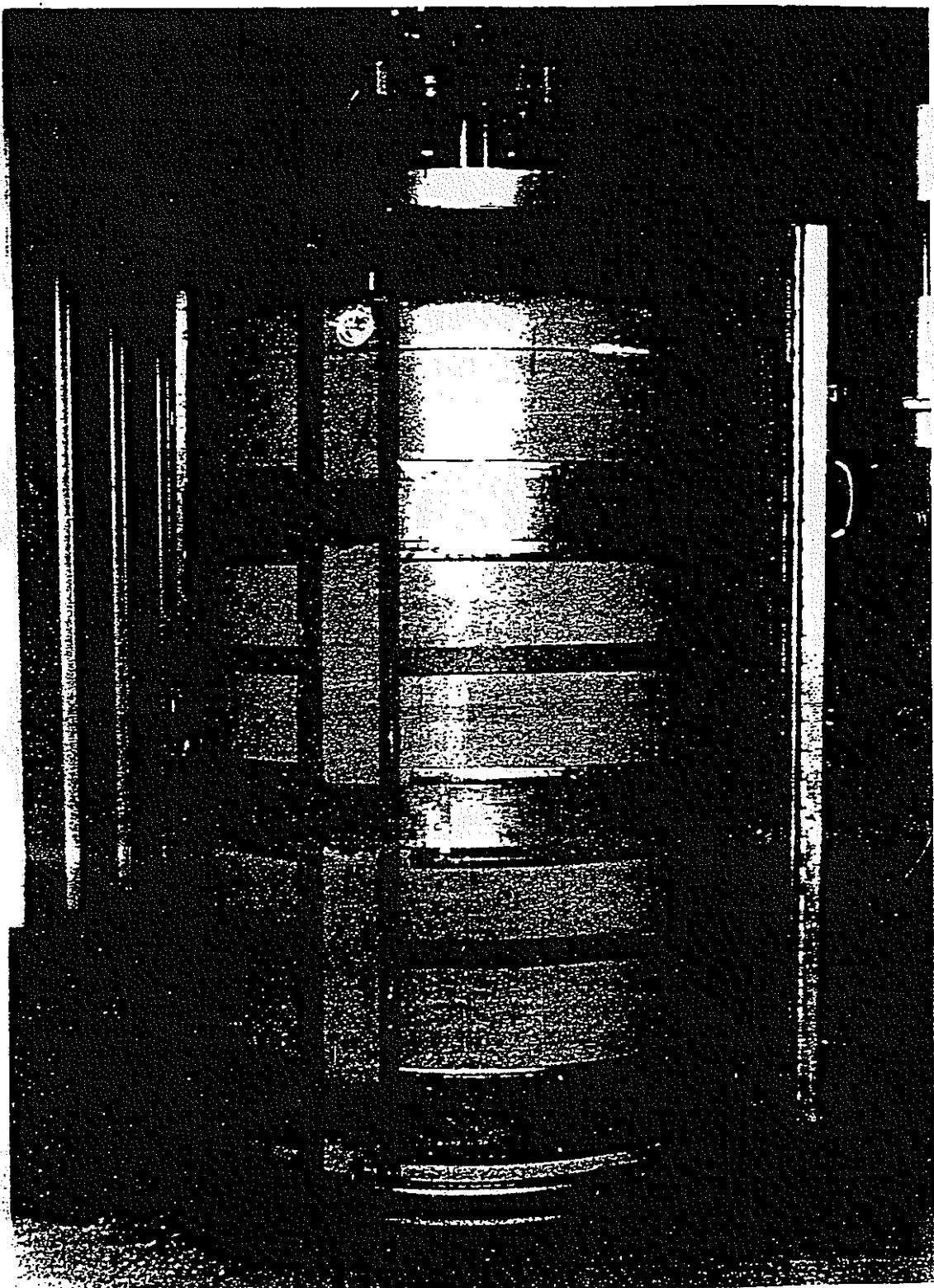
2005 Termination of decommissioning

1500 tons of metallic waste

3000 m<sup>3</sup> of construction materials waste

2.55 Billions DM estimated cost

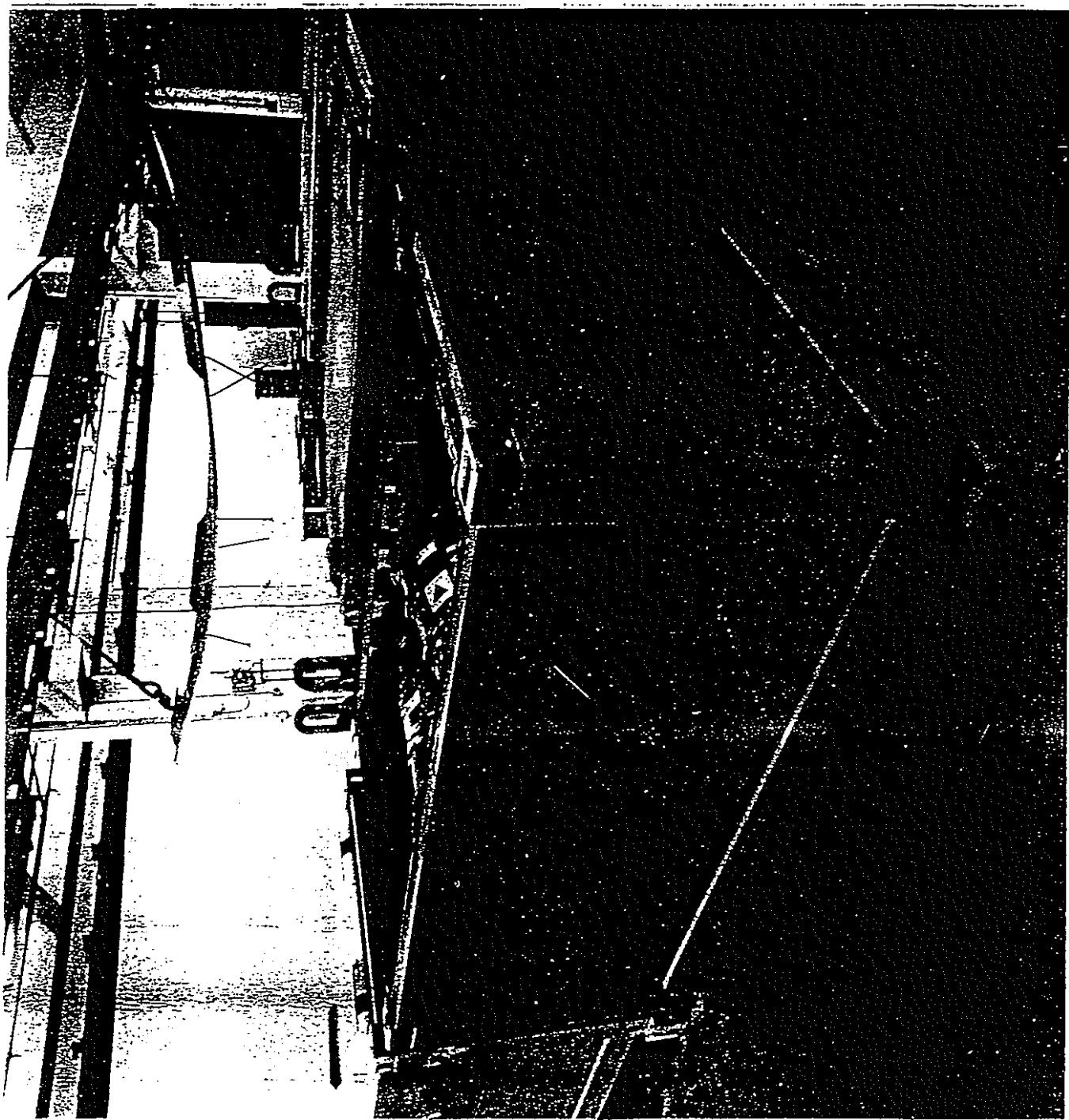


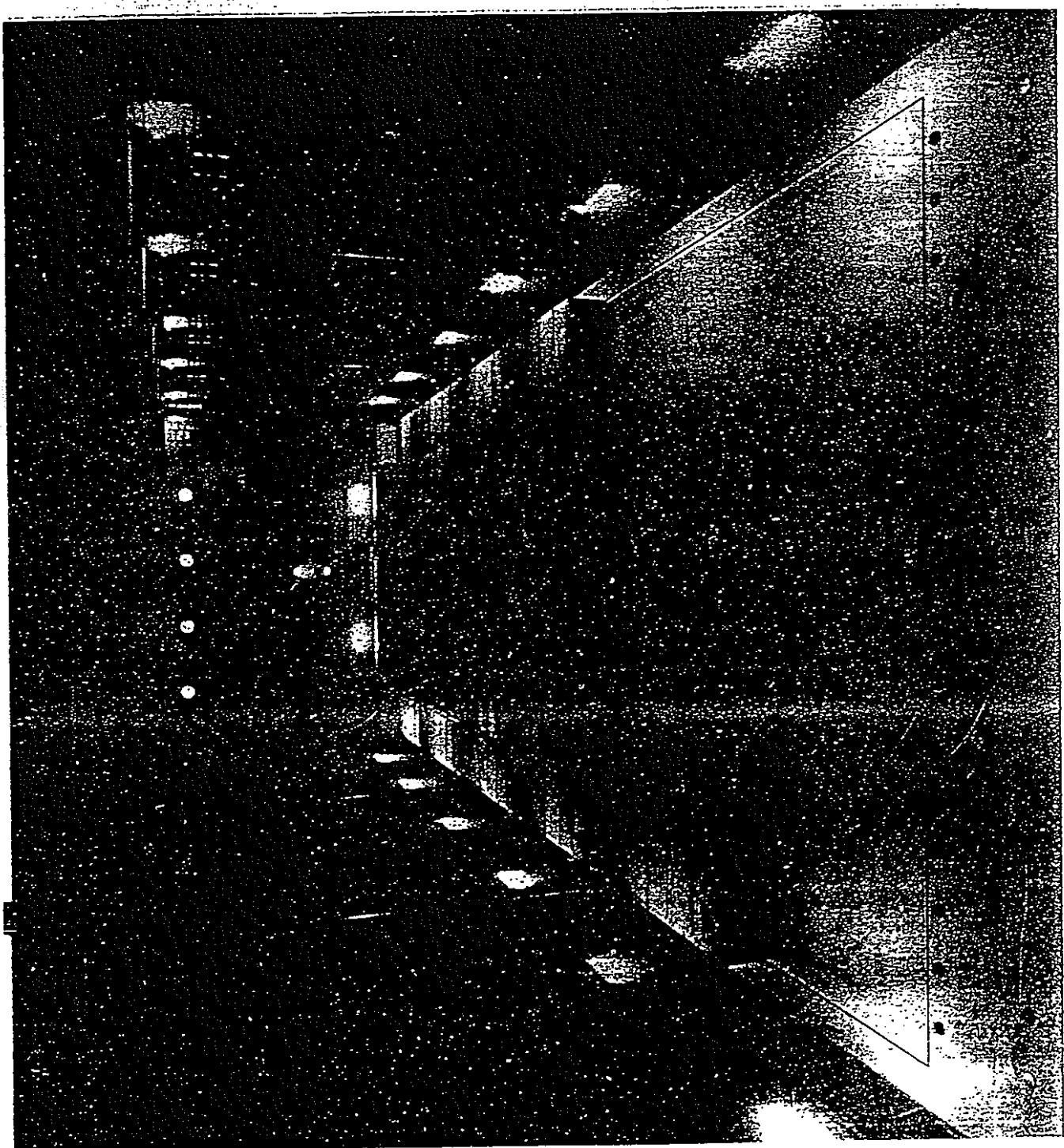


GNS

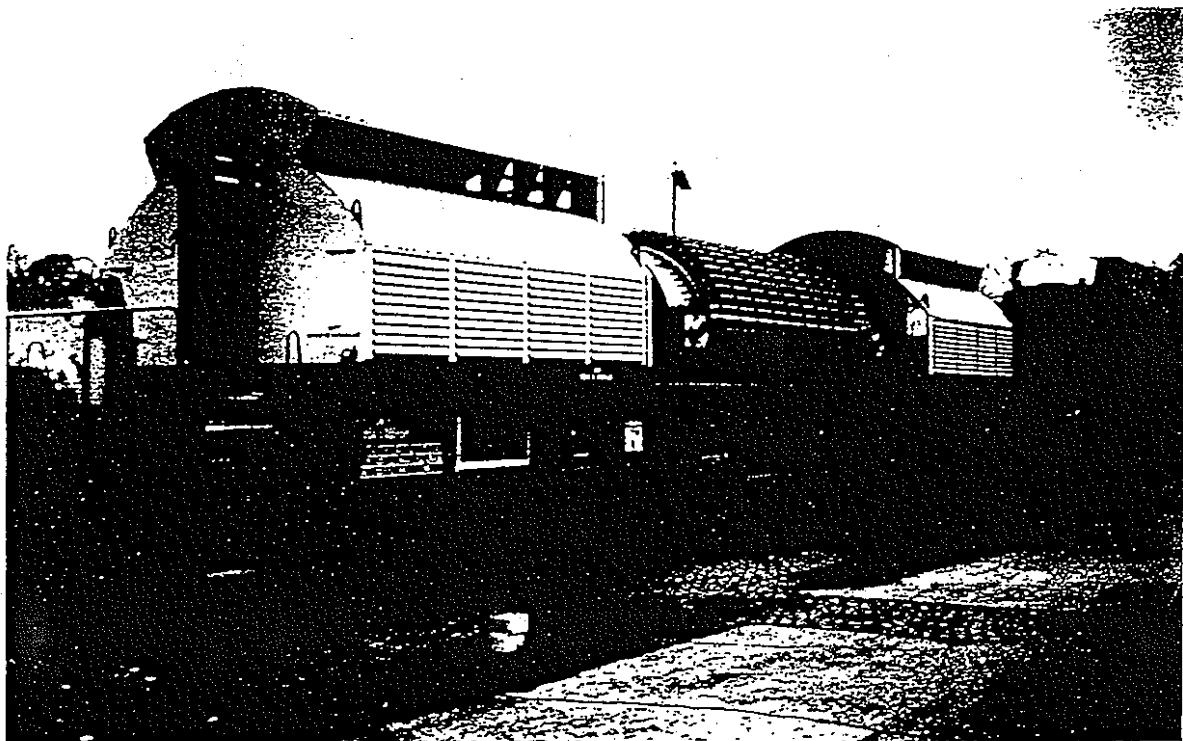
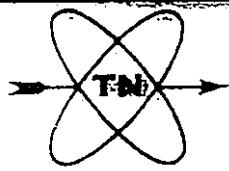
FAVORIT

Behälterheizung  
cask heating

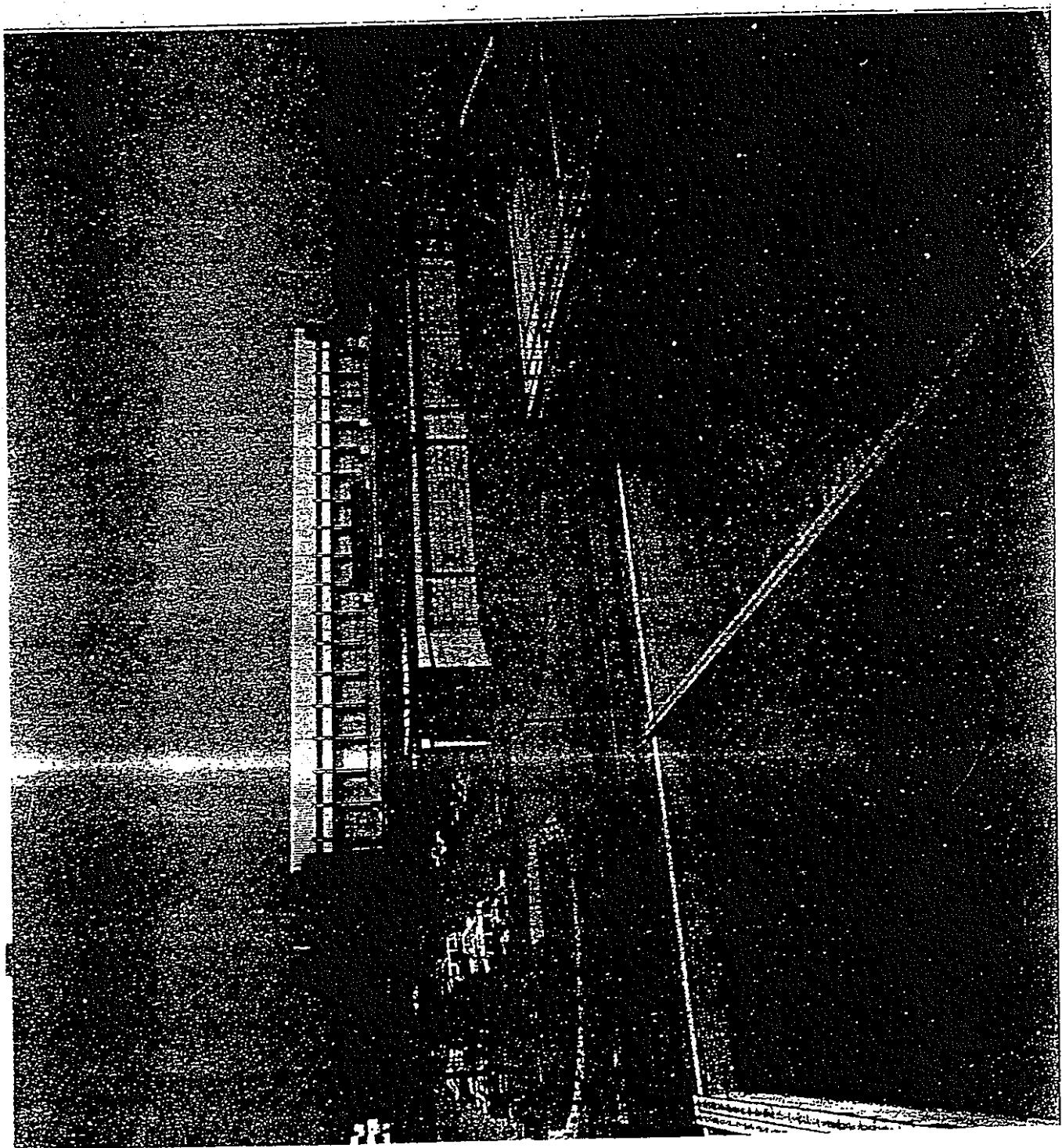




**TN 1300**



**Transportbehälter für bestr. Brennelemente**  
**Transport container for spent fuel elements**



Direct disposal of spent fuel

- additional management strategy for unreprocessable spent fuel elements
- amendment to the Atomic Energy Act to eliminate the priority of reprocessing

スケジュール

1979 Development of direct disposal techniques started

1986 License application for a pilot plant for spent fuel conditioning and encapsulation at Gorbeben

1996 Intended start-up of hot operation

- plant capacity : 35 tons/year spent fuel from LWR

- Two package options :

    Pollux-casks for disposal in drifts(reference)  
                for disposal in boreholes(back-up)

- Cask-dimensions : 5.5m×1.5m $\phi$ , 65tons.loaded)

Reprocessing of spent fuel

1985 Atomic Energy Act : Reprocessing as reference strategy

1989 German plants for industrial reprocessing(wackersdorf)  
abandoned

1990 Decision to reprocess spent fuel abroad

Contract with BNFL/COGEMA covering 500t/year reprocessing capacity

1994 start of return of waste glass from France

Year 2000 : 887tons of spent fuel reprocessed by BNFL/COGEMA

4200 canisters(150 l) of glass product

After 2000: 3200 canisters, optional

Waste disposal

Interim storage: Gorbeben, Aarhus (waste glass)

Mitterteich (NPP wastes)

Gorbeben repository

- salt dome, investigated since 1979
- suitable for all types of waste, especially heat-generating ones
- Above-ground exploration almost complete
- underground exploration still in progress
- Expected to be available in 2005-2010
- cost estimate for characterization, design and construction  
    2.6 billion DM

Konrad repository

- Former iron mine, to be used for non-heat generating waste
- $10^6 \text{ m}^3$  disposal volume (40 years utilization)
- Licensing procedure not yet terminated
- Start-up expected for mid-nineties
- costs for design, 380 million DM

Moersleben disposal facility

- used for disposal since 1979
- About 12000  $\text{m}^3$  liquid waste conditioned by in-situ solidification
- 4600  $\text{m}^3$  solid waste packages
- presently safety evaluation underway
- Will be used only to accommodate former GDR waste

Vitrification of the HAWC-WAK

Main operation data

88m<sup>3</sup> of HAWC(incl.dilution by transfer etc.)

9.6×10<sup>17</sup>Bq total activity, main radionuclides

Cs-137 and Sr-90 with daughters, 95%

About 400kg noble metal(commercial-like HAWC)

28ℓ/hr Melter feed rate (incl.10%recycle)

5 months operation time(100% availability)

50 tons of glass product

125 glass canisters(400kg,150ℓ )

16wt% waste oxides loading

Melter technology

•Melter type K-6', noble metals-compatible

•designed and constructed by KfK-INE

•Tested in the VA-WAK facility 1990-93

Vitrification schedule

•Transportation of the HAWC-WAK to Mol 1997/98

•1998 Start of vitrification campaign

3.2 Conceptual study for the conditioning of the HAWC-WAK waste  
and on-site vitrification



## **Basic data (theoret. blend of tanks)**

### **Total quantities and main data**

Total feed volume	86 m <sup>3</sup>
Total activity	9.6*10 <sup>17</sup> Bq
Specific activity	1.1*10 <sup>13</sup> Bq/l
Specific decay heat	0.9 W/l
Glass production	49.5 to
Total spec. activity product	1.9*10 <sup>13</sup> Bq/kg
Specific heat in product	1.6 W/kg

### **Material data**

Oxide yield	92 g/l
Waste loading	16 wt-%
Quantity of product per liter HAWC	0.575 kg

### **Main material streams**

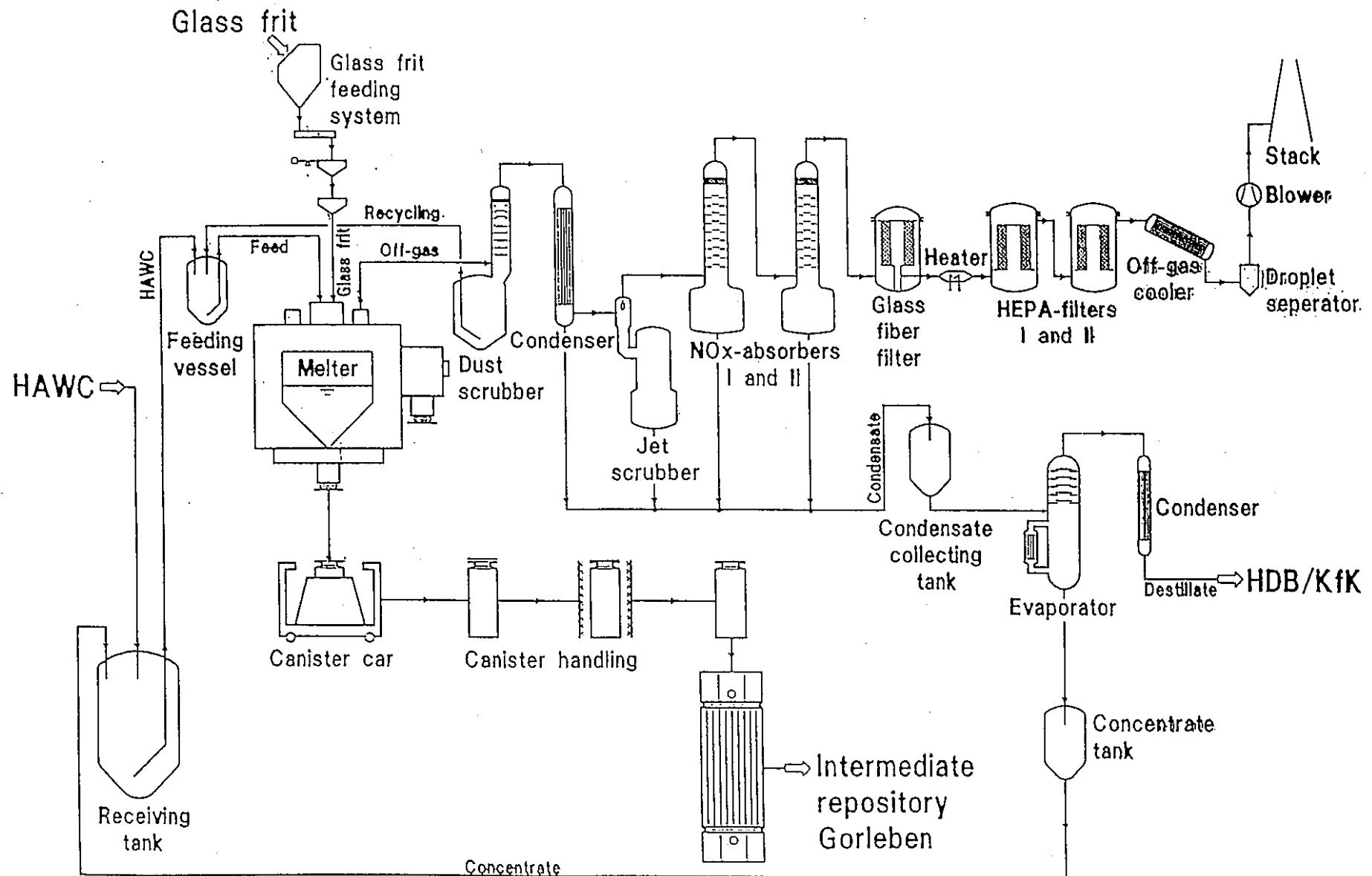
Throughput of HAWC	25 l/h
Throughput of Feed	28 l/h
Glass production rate	14.4 kg/h
Canister production rate	1 canister in 28 h

## Canister data

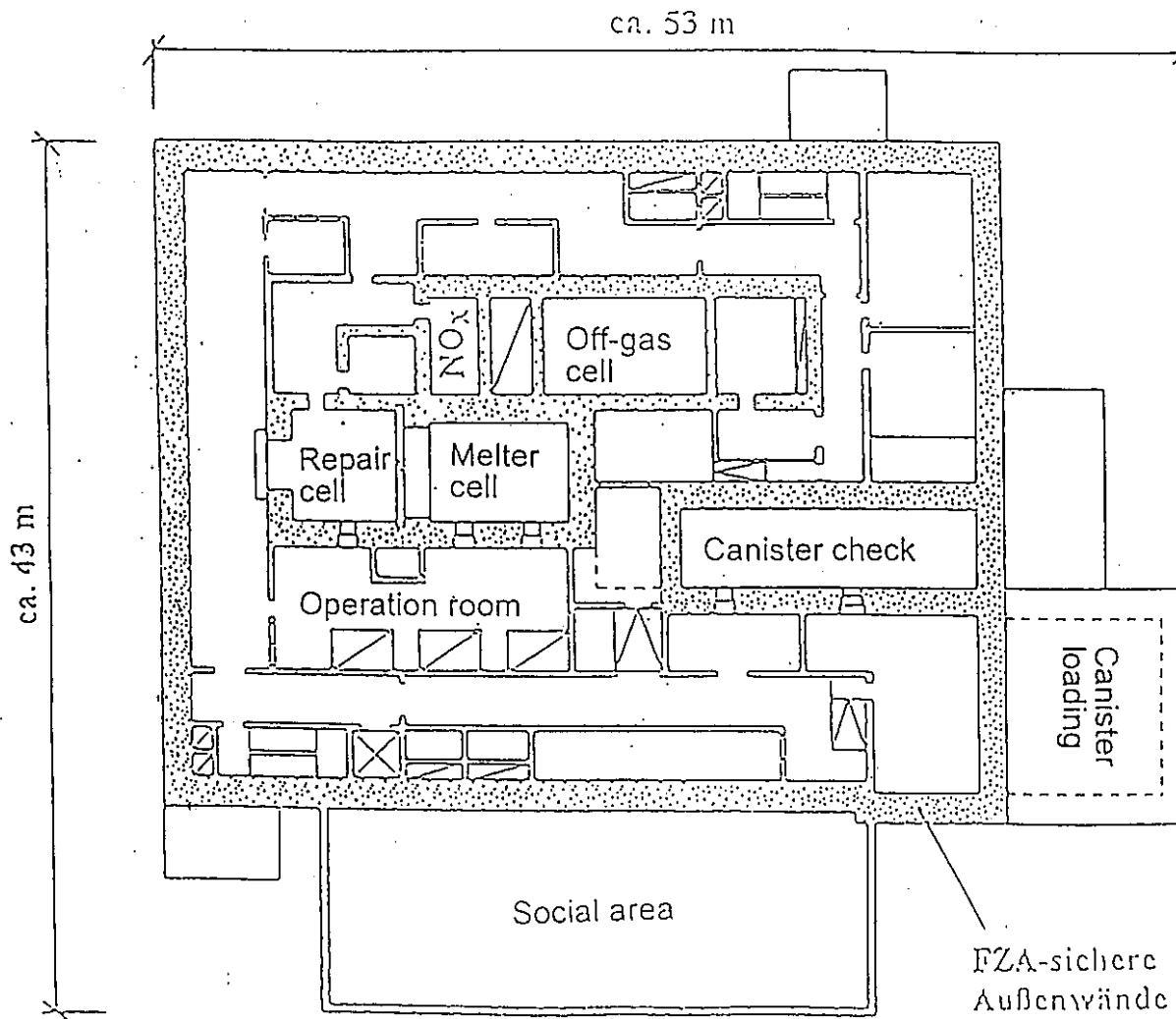
Canister size	dia. 430*130 mm height
Capacity	400 kg
Total activity	$7.8 \times 10^{15}$ Bq
Main radioisotope	Cs-137/Ba-137 m, 55 %
Number of canisters	124
Volume of HAWC pressed for one canister	$\approx 700$ l

## Operational data

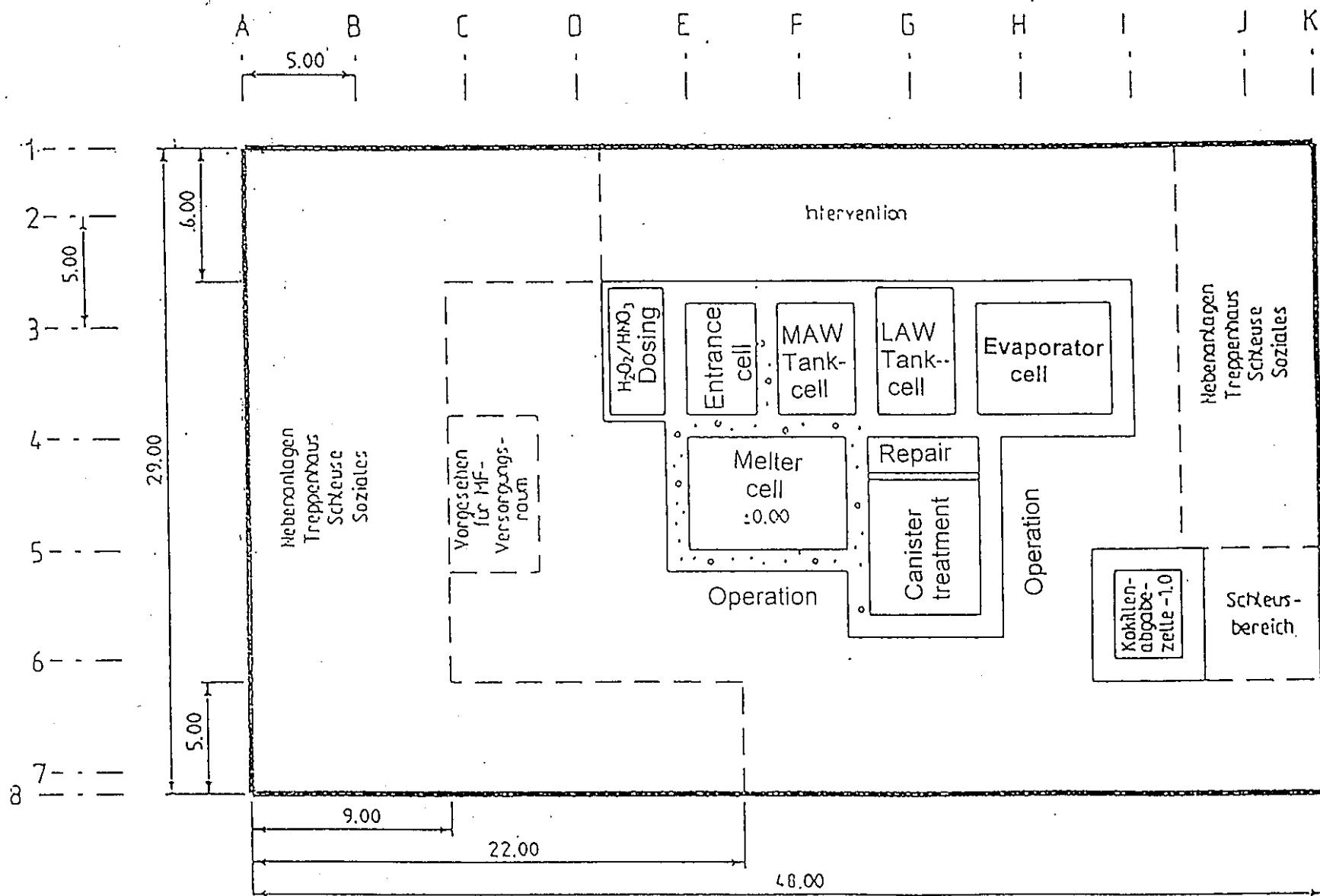
Total operation time (25 l/h, 100% availability)	143 days ( $\approx$ 5 months)
Operation mode	continuous
Secondary liquid waste, total	$\approx 70$ m <sup>3</sup> condensate $\approx 25$ m <sup>3</sup> scrub solution
Quantity of evaporator concentrate	$\approx 3$ m <sup>3</sup> (concentration factor of 30)



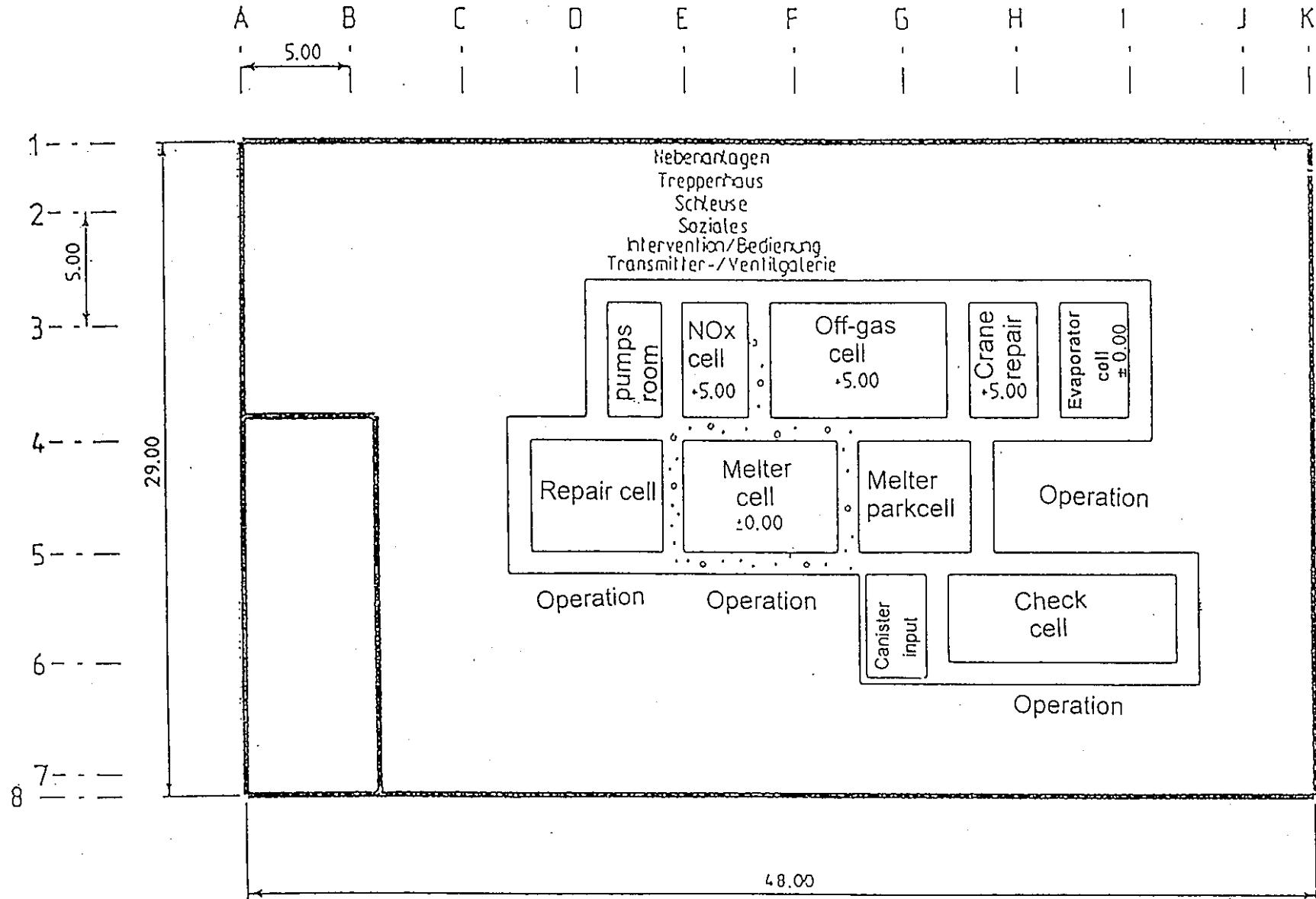
Basic flowsheet of the HAWC-WAK vitrification plant on site



Possible structure of the on-site vitrification plant

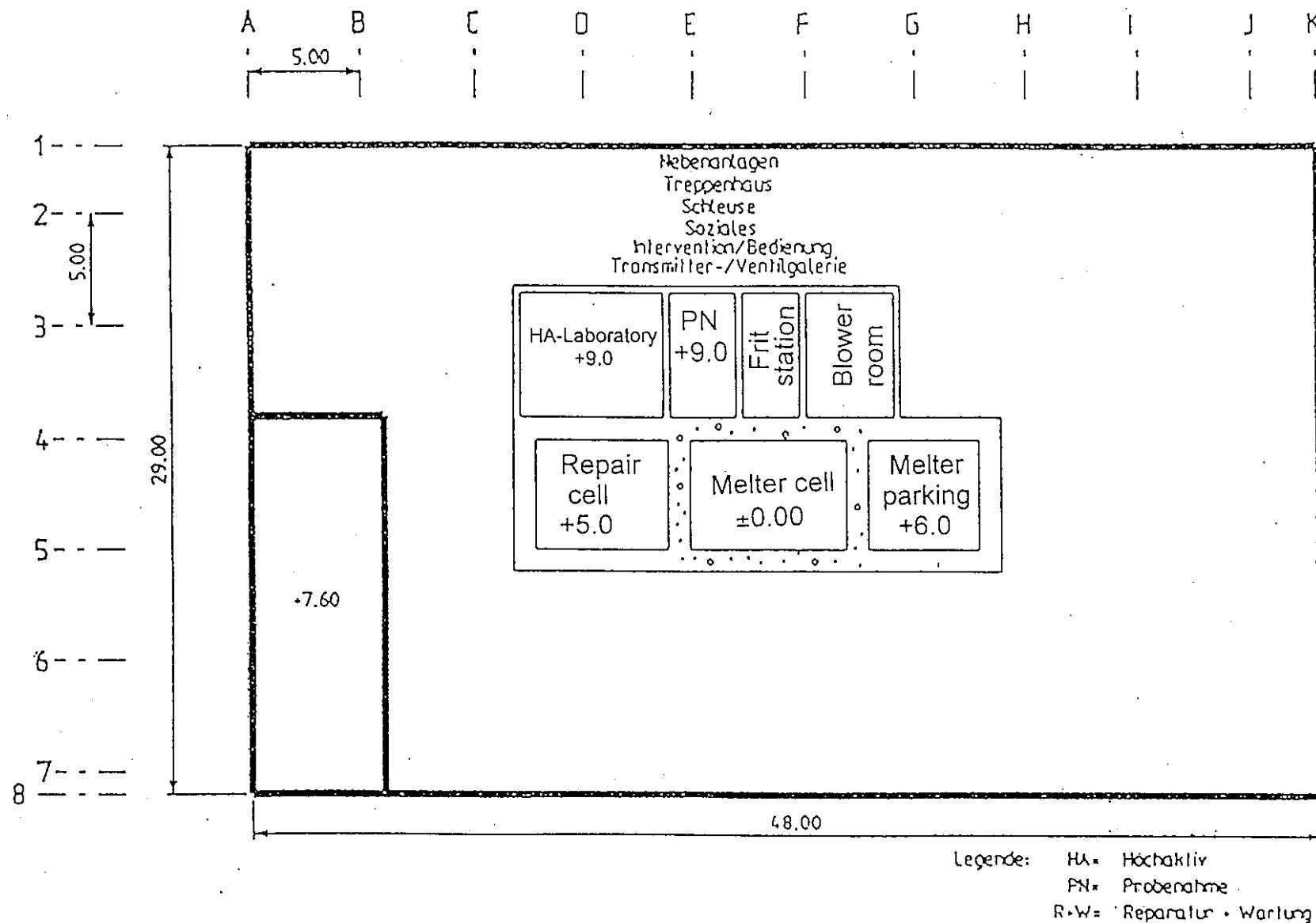


Cell arrangement, plan view, level +0m



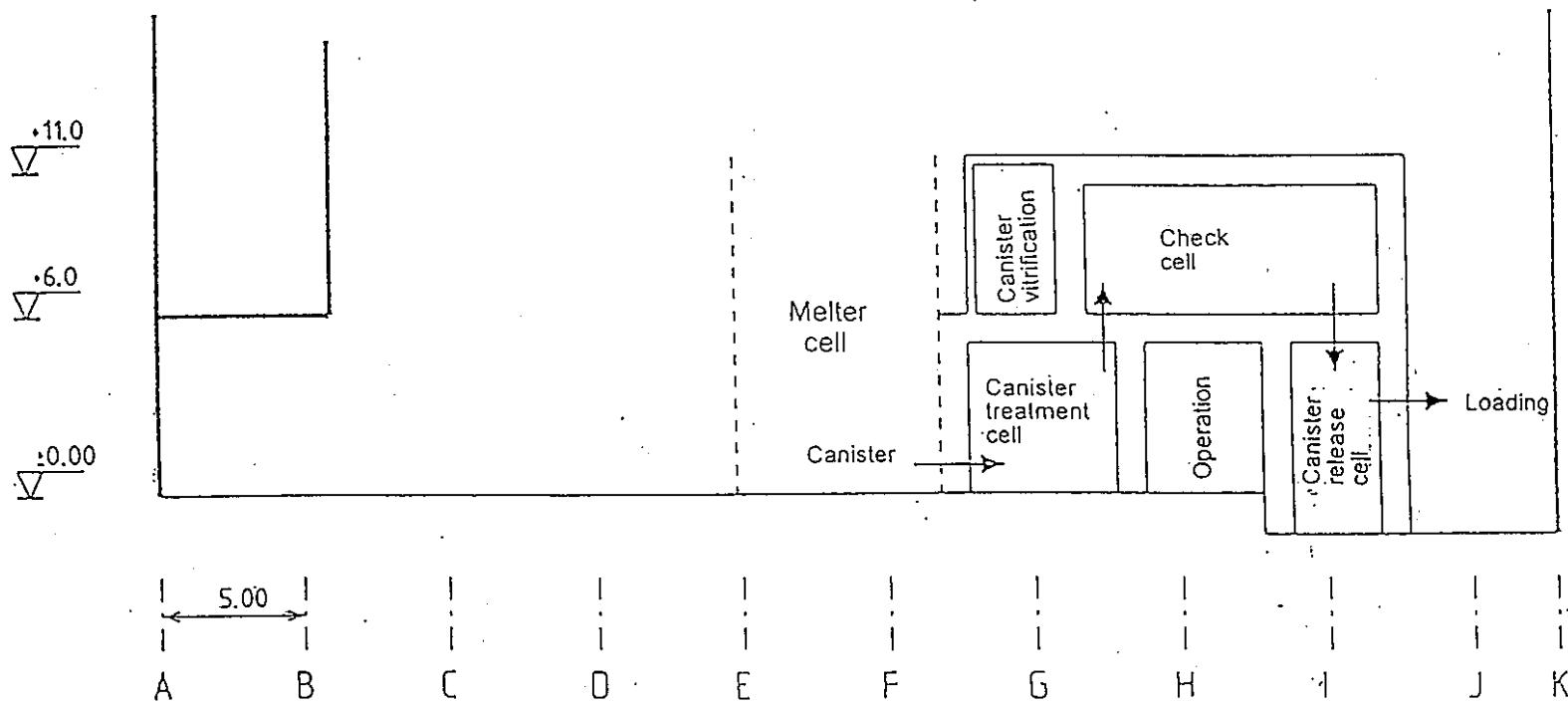
Cell arrangement, plan view, level +6m

-219-

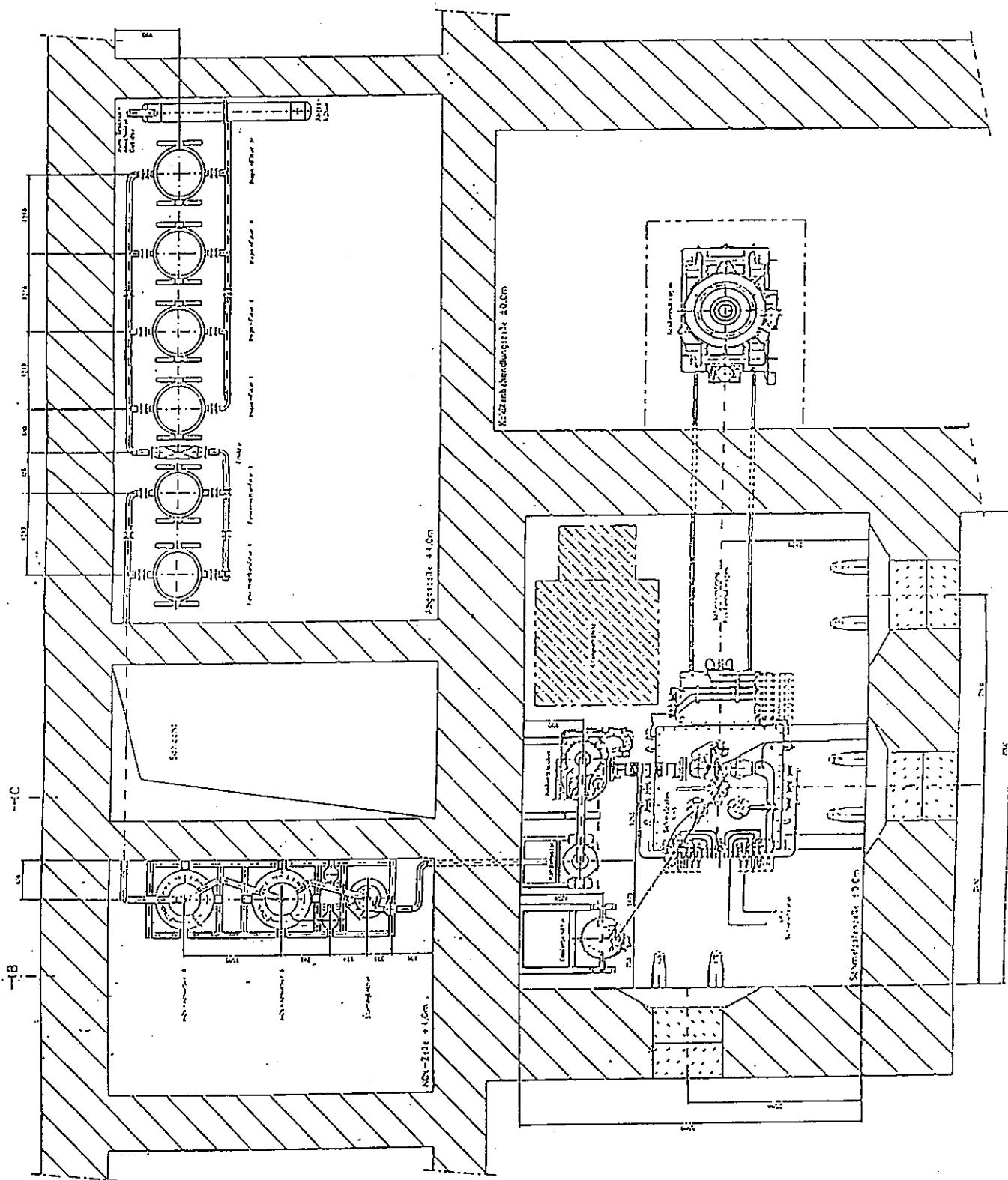


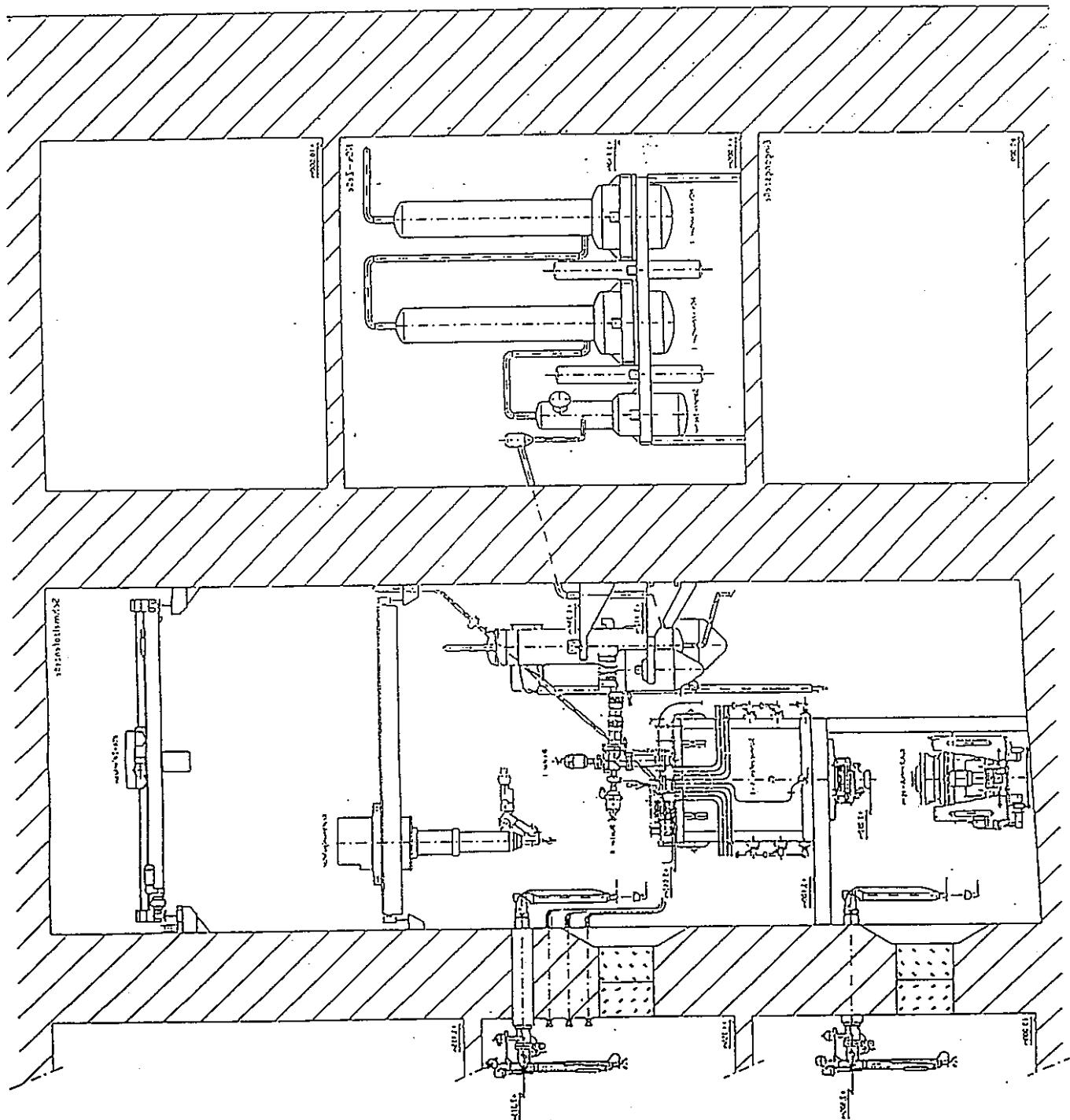
Cell arrangement, plan view, level +11 m

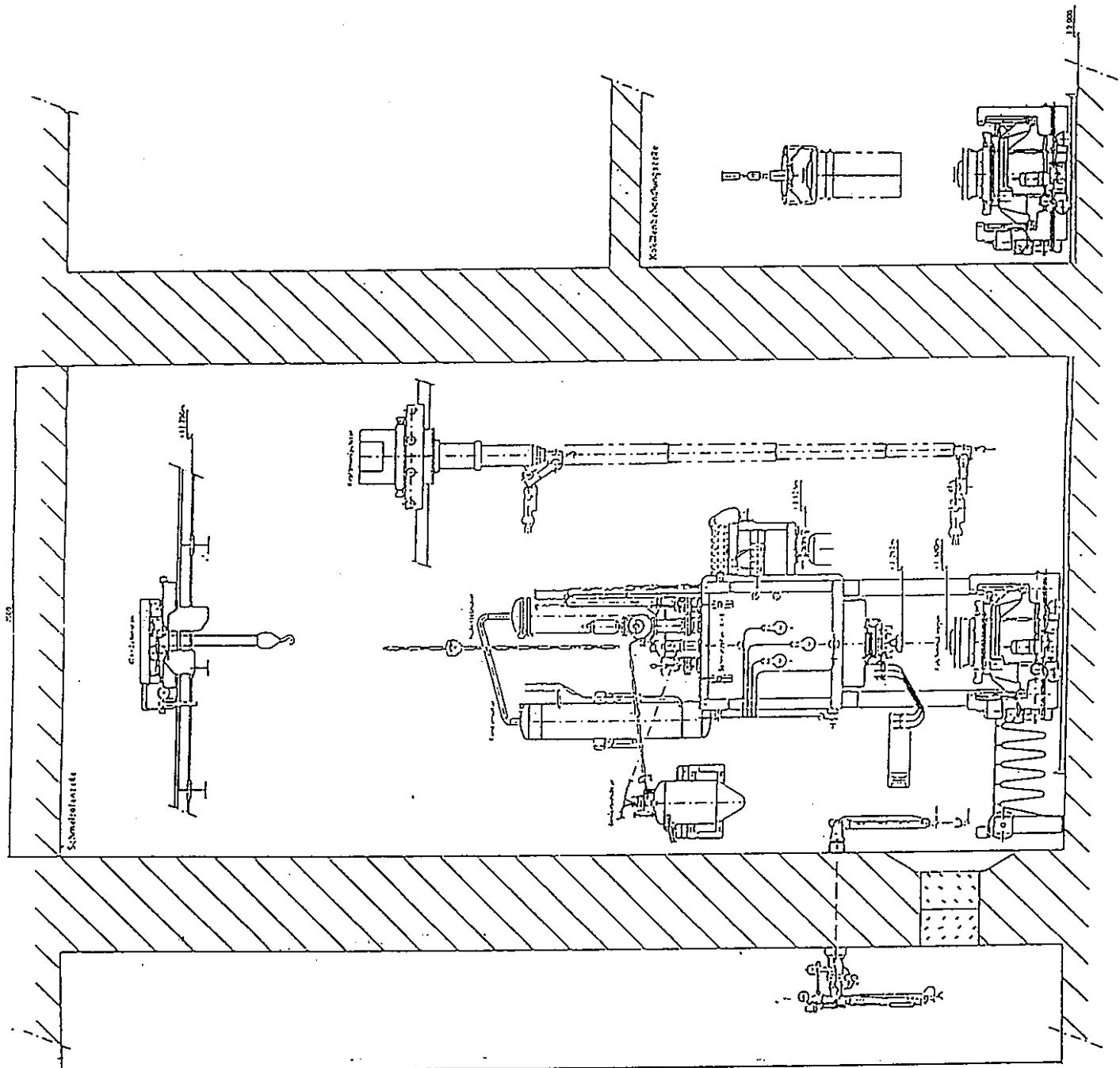
-220-



Cell arrangement, longitudinal cut canister handling / loading







## HAWC-Transfer

- Transport cask (CASTOR)
- Tube channel (underground / above ground)
- Direct transfer

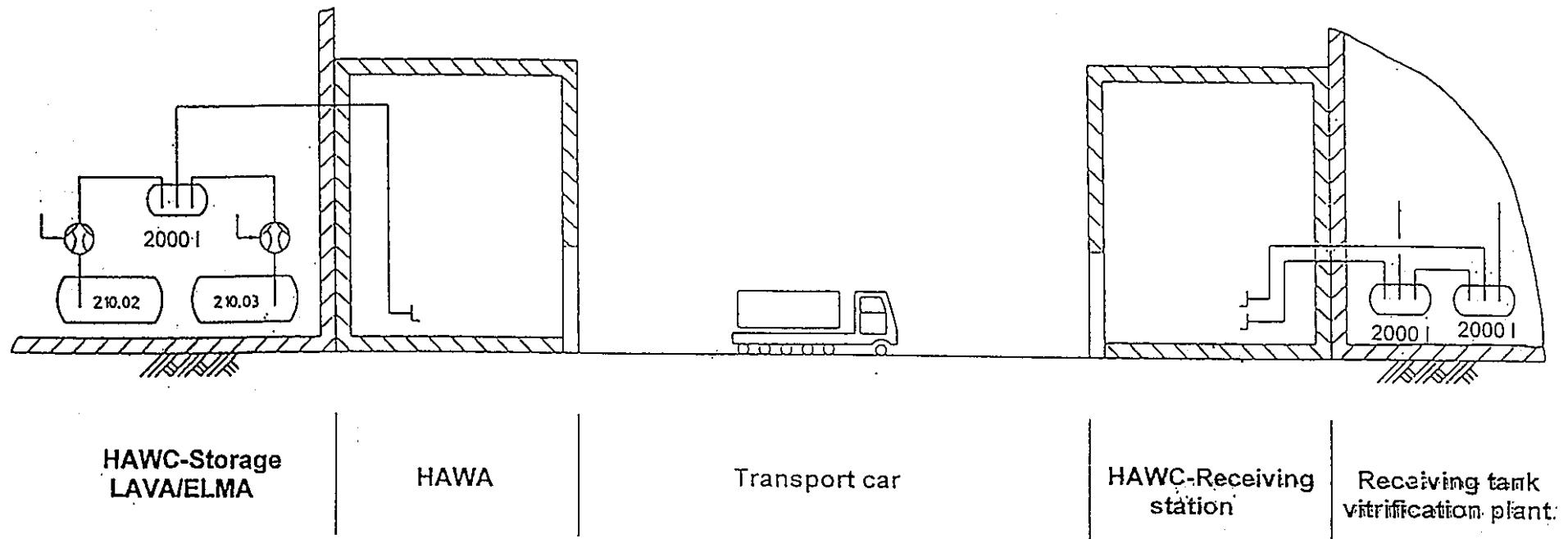
## Location

- Installation inside existing WAK / KfK buildings
- New building on WAK site / KfK site

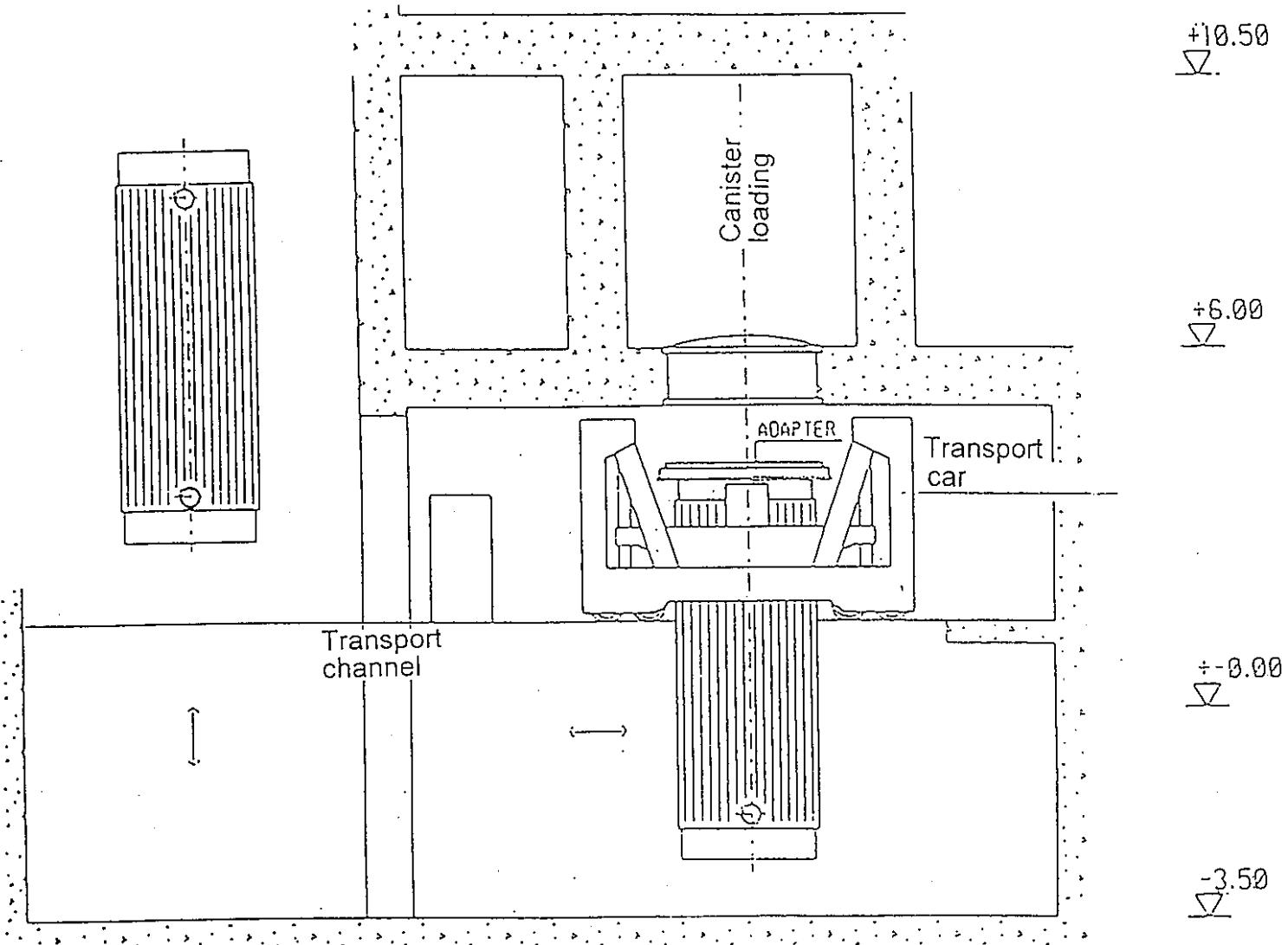
## Canister storage

- Direct storage inside existing WAK / KfK buildings
- Direct storage inside new WAK / KfK buildings
- Temporary storage in CASTOR on site
- CASTOR transport to Gorleben

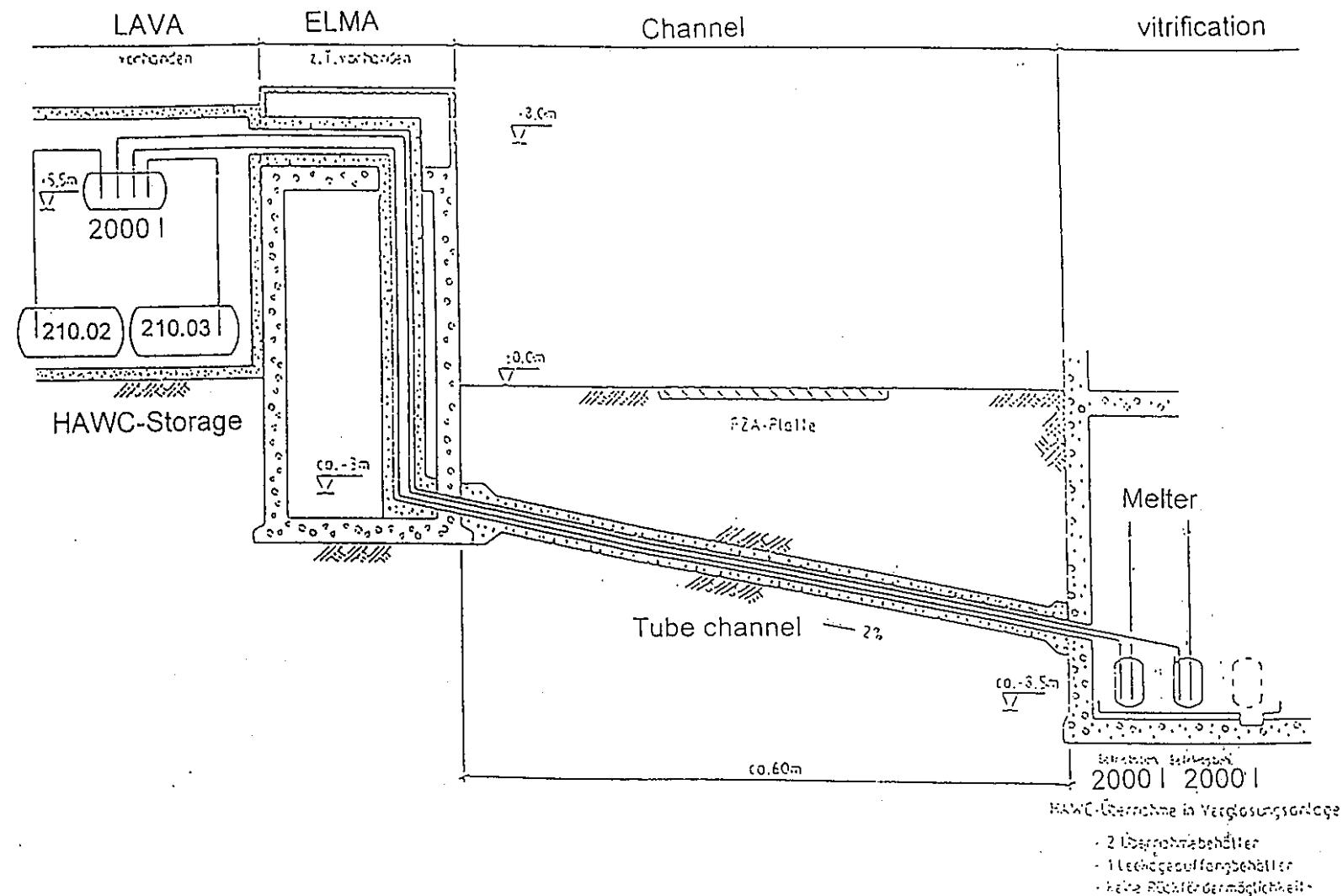
## **Variants considered for the vitrification plant**



**HAWC transfer by transport car**



Canister loading device for CASTOR



Underground HAWC transfer channel

## Time schedule for vitrification plant for on-site

Steps (6/93)	Months
1. Planing of construction, safety report, application	30
2. Examination of application	16
3. Participation of public	12
4. Permission of construction	14
5. Construction of building	18
6. Construction of facility	30
7. Cold test	6
8. Permit of operation	6
9. Vitrification operation	16
Total time in months	148
Total time in years	12

	<b>state of cost' 93</b>	<b>extrapolated</b>
1. Work of building owner	65 Mio DM	74 Mio DM
2. Planning costs	120 Mio DM	136 Mio DM
3. Cost of construction	242 Mio DM	340 Mio DM
4. Operation	36 Mio DM	60 Mio DM
5. Decommissioning, Removal	220 Mio DM	420 Mio DM
6. Glass canister, secondary waste	40 Mio DM	70 Mio DM
7. Operation HAWC storage tanks	340 Mio DM	450 Mio DM
Total costs	1.063 Mio DM	1.550 Mio DM

Costs calculation for the vitrification plant

3.3 Evaluation of the noble metals behavior, ESM feed  
preparation system

## Vitrification process performance

- Noble metals behavior in the K-G' melter
- ESM - project

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12<sup>th</sup> annual PNC/KFK meeting, Dec. 1993

- 234 -

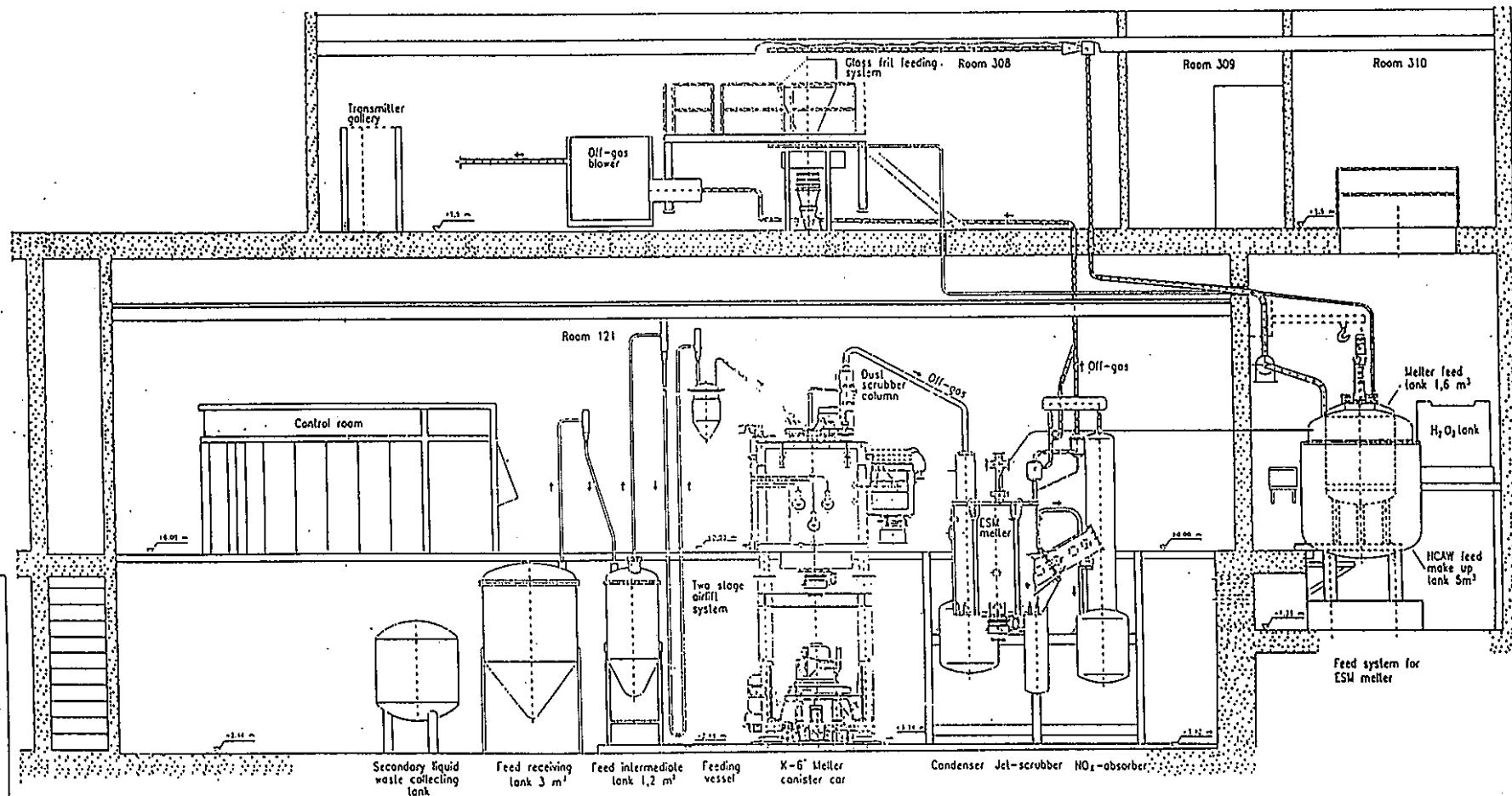


Fig. 4-1: Simplified sketch of the longitudinal cross section of the inactive test facility VA-WAK with the meter K-6' (integrated in this facility is the ESM meter on the right side of K-6' and its feeding system)

- 235 -

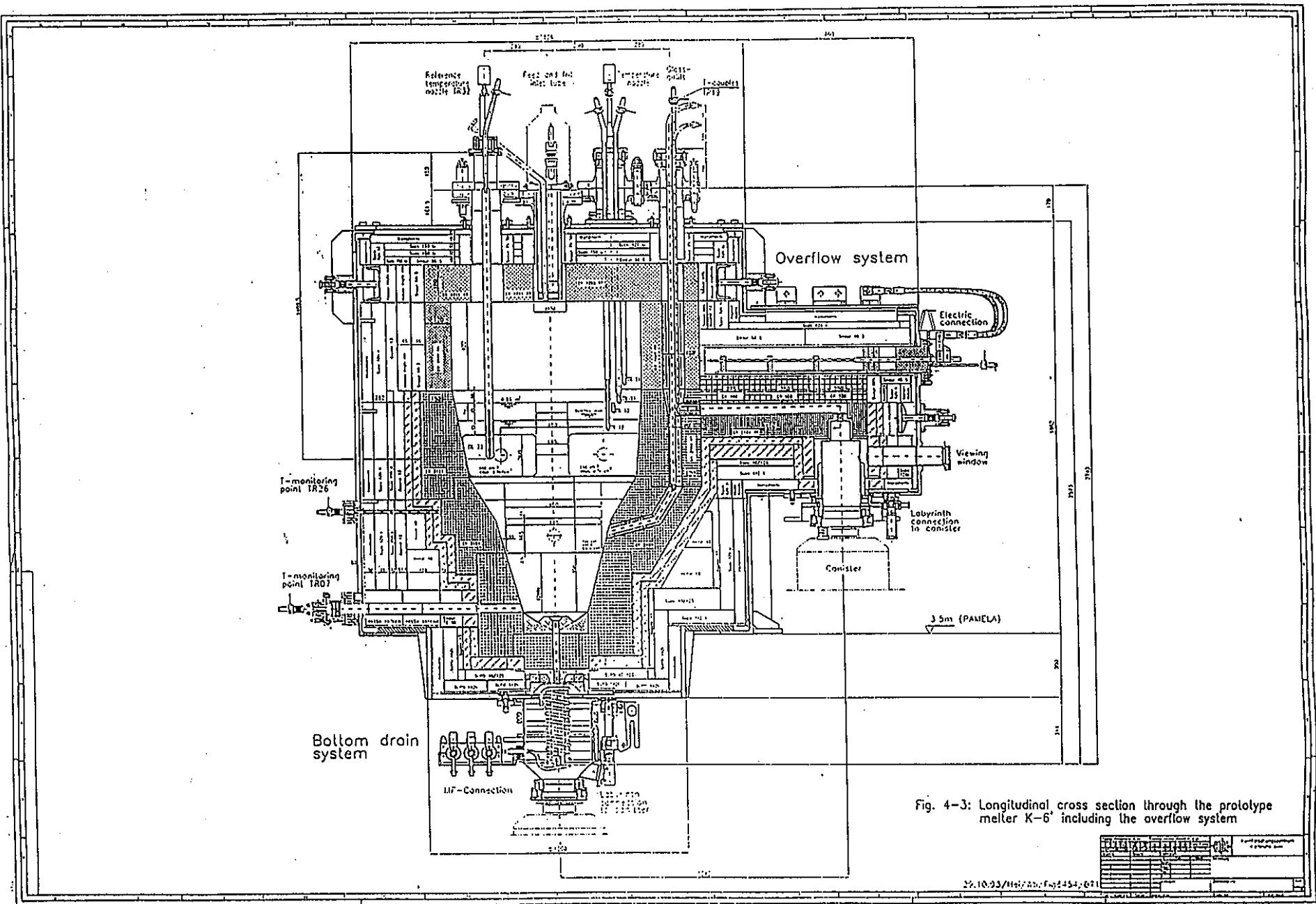


Fig. 4-3: Longitudinal cross section through the prototype melt K-6' including the overflow system

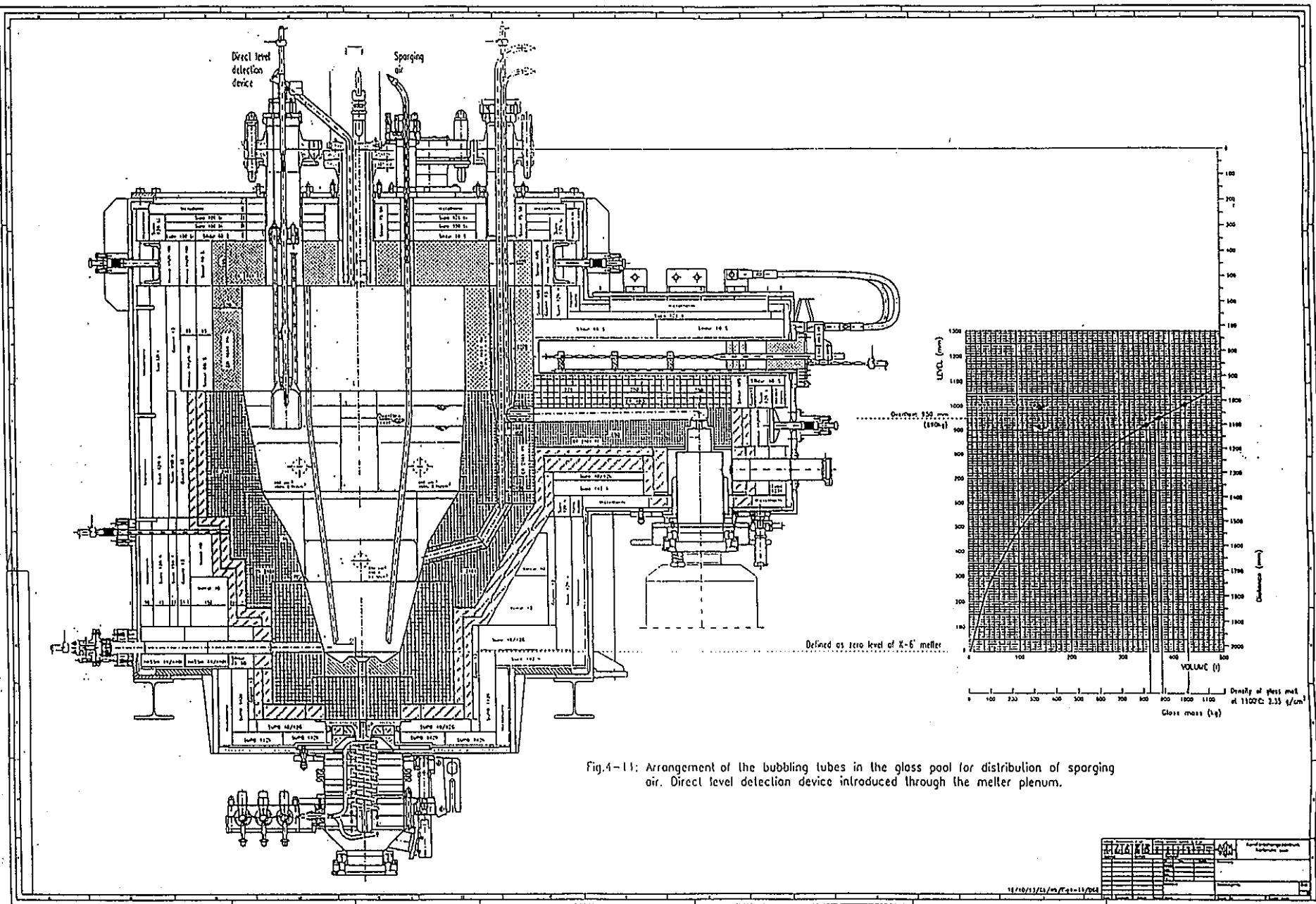


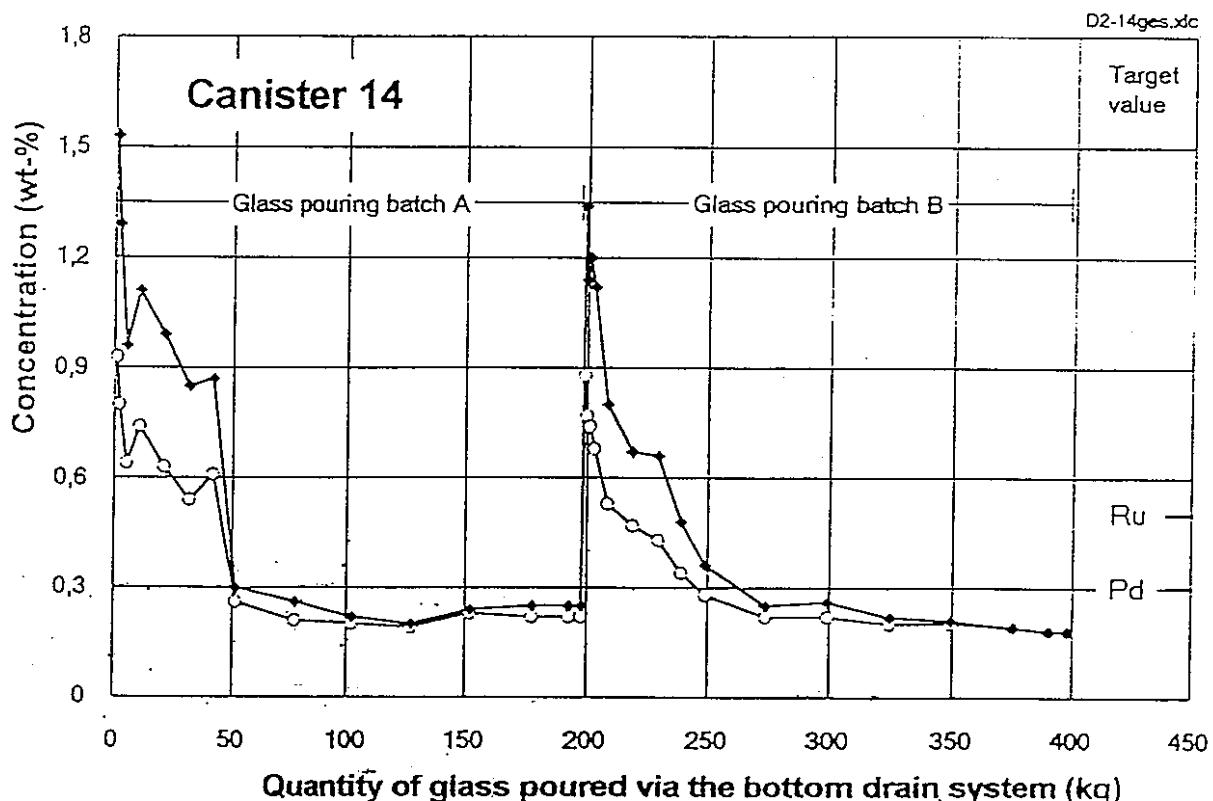
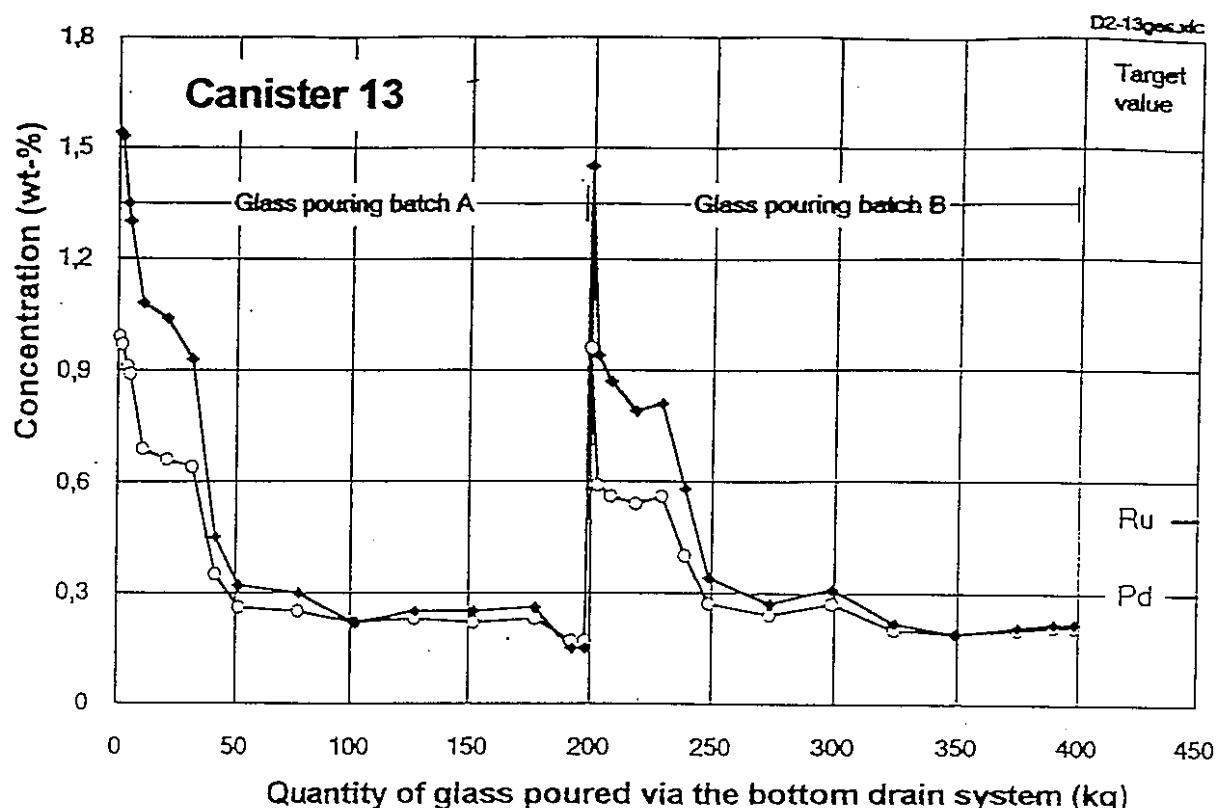
Fig.4-11: Arrangement of the bubbling tubes in the glass pool for distribution of sparging air. Direct level detection device introduced through the melter plenum.

# Vitrification runs with the K-6<sup>1</sup> melter

	D 1	D 2	D 3	D 4	D 5	D 6
Time of operation:	May 25-June 4, 1990	June 4-24, 1990	April 13-18, 1991	June 5-26, 1991 (42 h idling)	Feb. 17-March 28, 1992 (336 h idling)	March 25-July 1, 1993 (286 h idling)
Feed type	HAWC-WAK <u>no noble metals</u>	HAWC-WAK Tank 210.02 2.91 % Ru 1.45 % Pd	-	HAWC-WAK Tank 210.02 2.91 % Ru 1.45 % Pd	HAWC-WAK Blend of Tank 210.02 + 210.03 2.28 % Ru 1.32 % Pd	HAWC-WAK Blend of Tank 210.02 + 210.03 + undissolved 2 r 2.29 % Ru 1.32 % Pd
Total volume, m <sup>3</sup>	5	10.3 (30 kg Ru) (18 kg Pd)	- glass product feeding	9.6 (28 kg Ru) (17 kg Pd)	15.4 (35 kg Ru) (20 kg Pd)	15.9 (36 kg Ru) (21 kg Pd)
glass frit	SWA 752	SWA 752	-	SWA 752	GG-WAK 1	GG-WAK 1
glass loading, wt.-%	13	14 (0.5 wt.-% Ru) (0.3 wt.-% Pd)	-	13.9 (0.5 wt.-% Ru) (0.3 wt.-% Pd)	14.3 (0.4 wt.-% Ru) (0.23 wt.-% Pd)	13.8 (0.38 wt.-% Ru) (0.22 wt.-% Pd)
glass product, to	2.5	5.9	0.8	5.6	8.7	9.3
number of pourings	15	30	9	28	42	47
glass pouring via	bottom drain	bottom drain	overflow drain	overflow drain	bottom drain	overflow drain

## Vitrification runs with the K-6' melter, continued

	D1	D2	D3	D4	D5	D6
Feed throughput, %/h	26-28	25	-	30 (max 35)	36	34
glass production rate, kg/h	15.4	13	14	15.4 (max 18.3)	18	14.9
pouring batch, kg	195	195	100	195	195	195
glass production yield, %/e	585	580	-	580	594	594
total melter power, kw (average)	80	82	-	85	85	80
power skew (upper to lower elect.)	2:1	2:1	-	3:1	4:1	4:1
glass pouring rate, kg/h	100-120	100-120	-	100-120	100-120	100-130
time availability, %	100	100	-	98	100	100
bubbling air	no	no	yes (600% each tube)	yes (600, 400, 200 l/h each tube)	no	yes
Draughting of the melter	$\nearrow$ 2.2 kg Pa Pd			$\nearrow$ 3.4 kg Pa 2 kg Pd		



U15.2-10

Fig. 6-7g : Noble metal concentrations in glass samples taken from pouring stream while filling canister 13 and 14 of the D2 test run

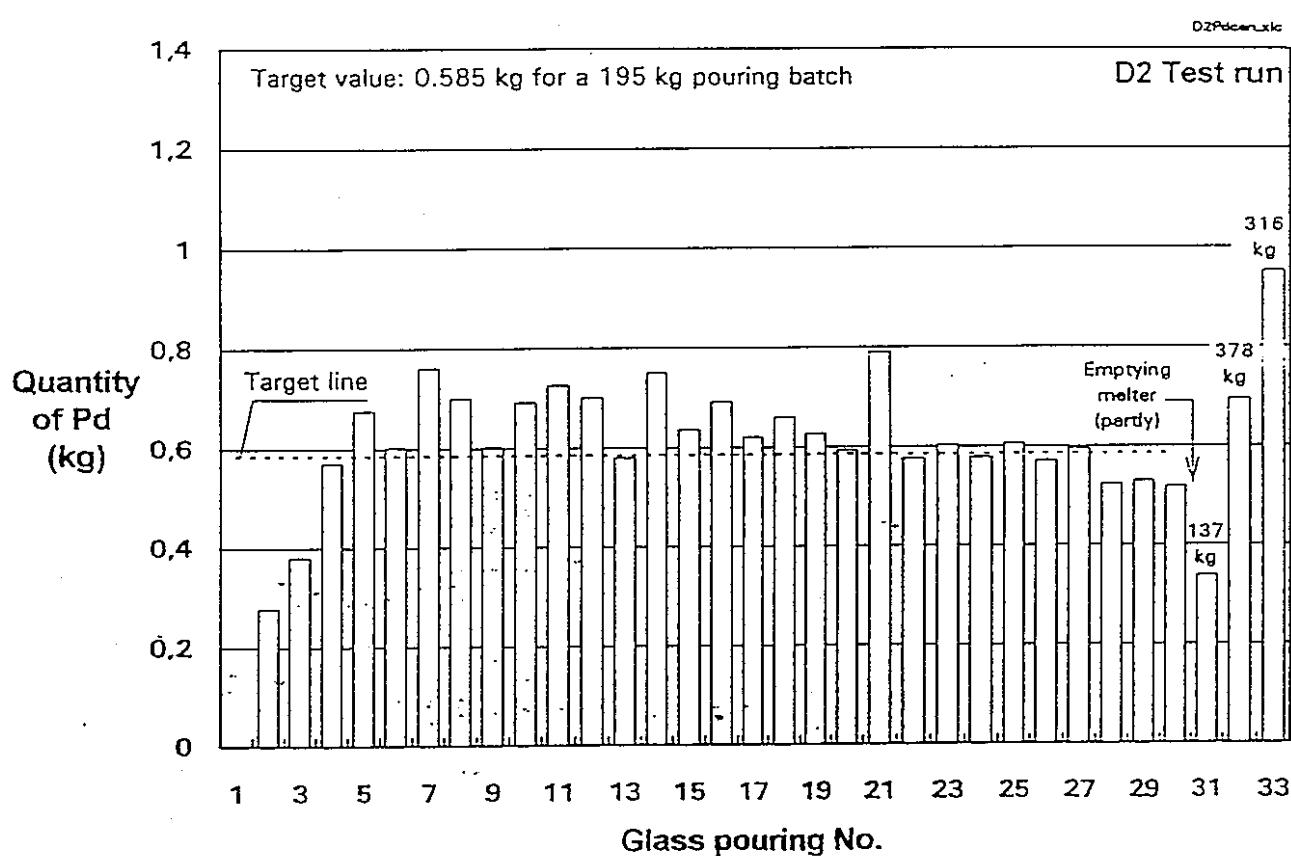
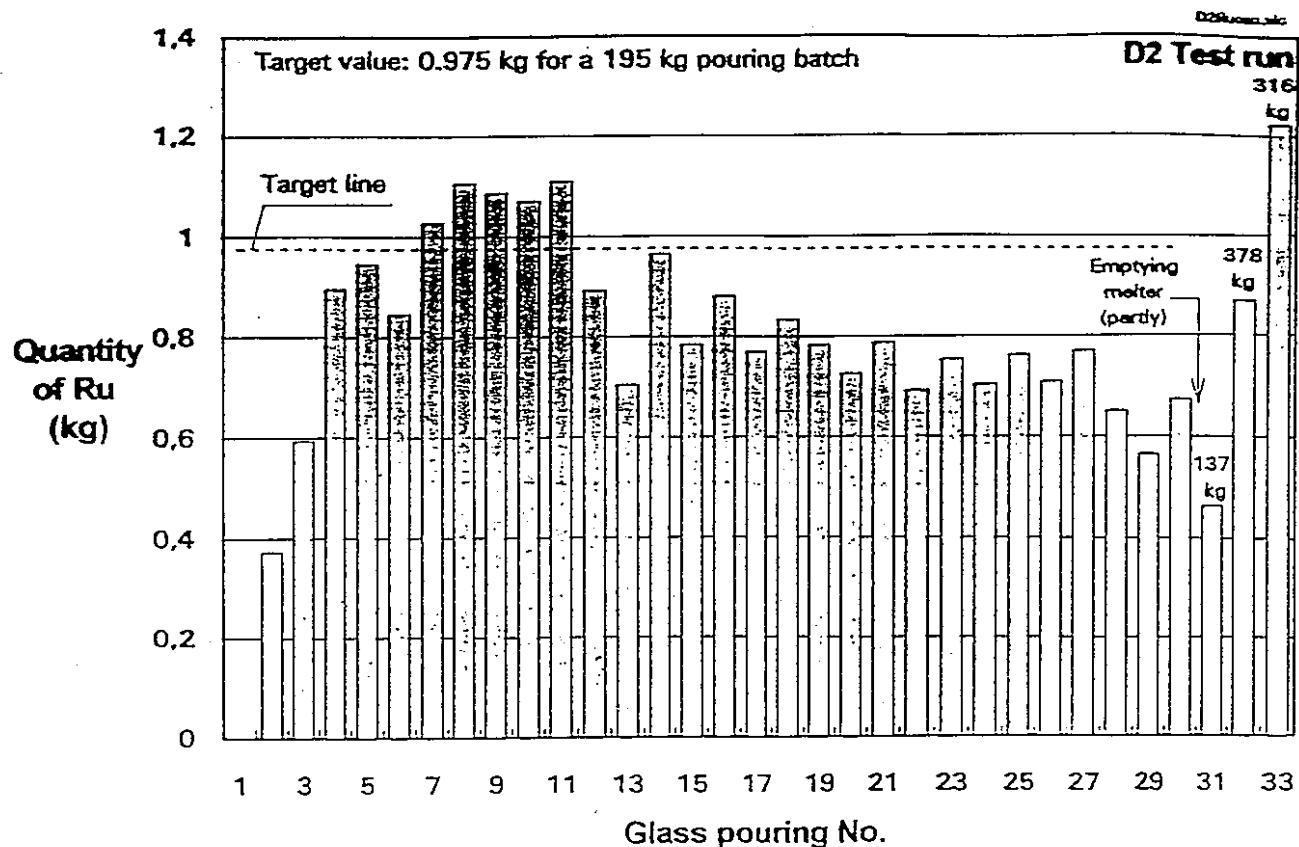


Fig. 6-8: Quantity of Ruthenium and Palladium, respectively, removed from melter by glass pouring via the bottom drain system. Test run D2; June 1990

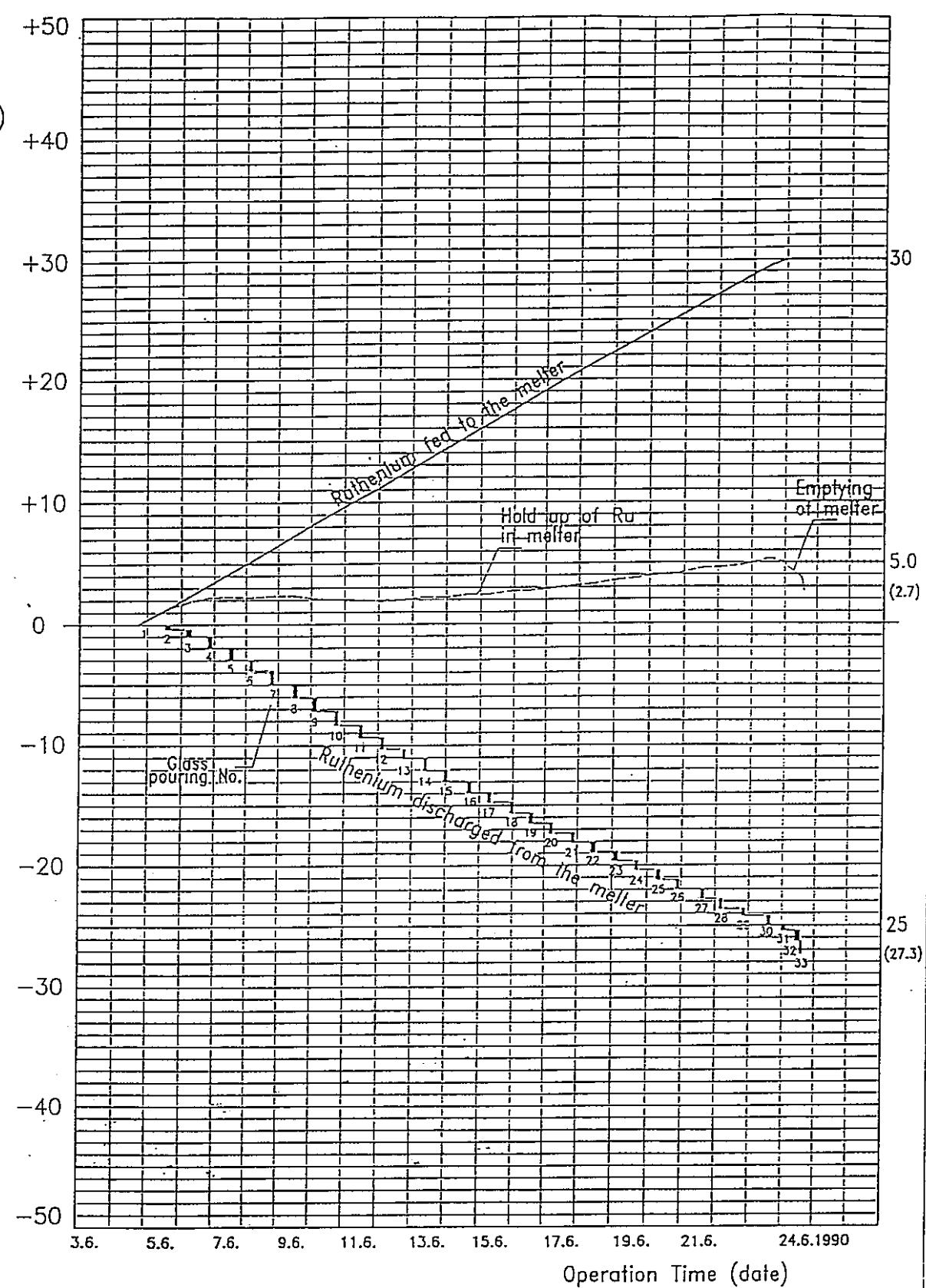


Fig.6-9a: Input and removal of Ruthenium from the K-6' melter during the long-term noble metals test run D2

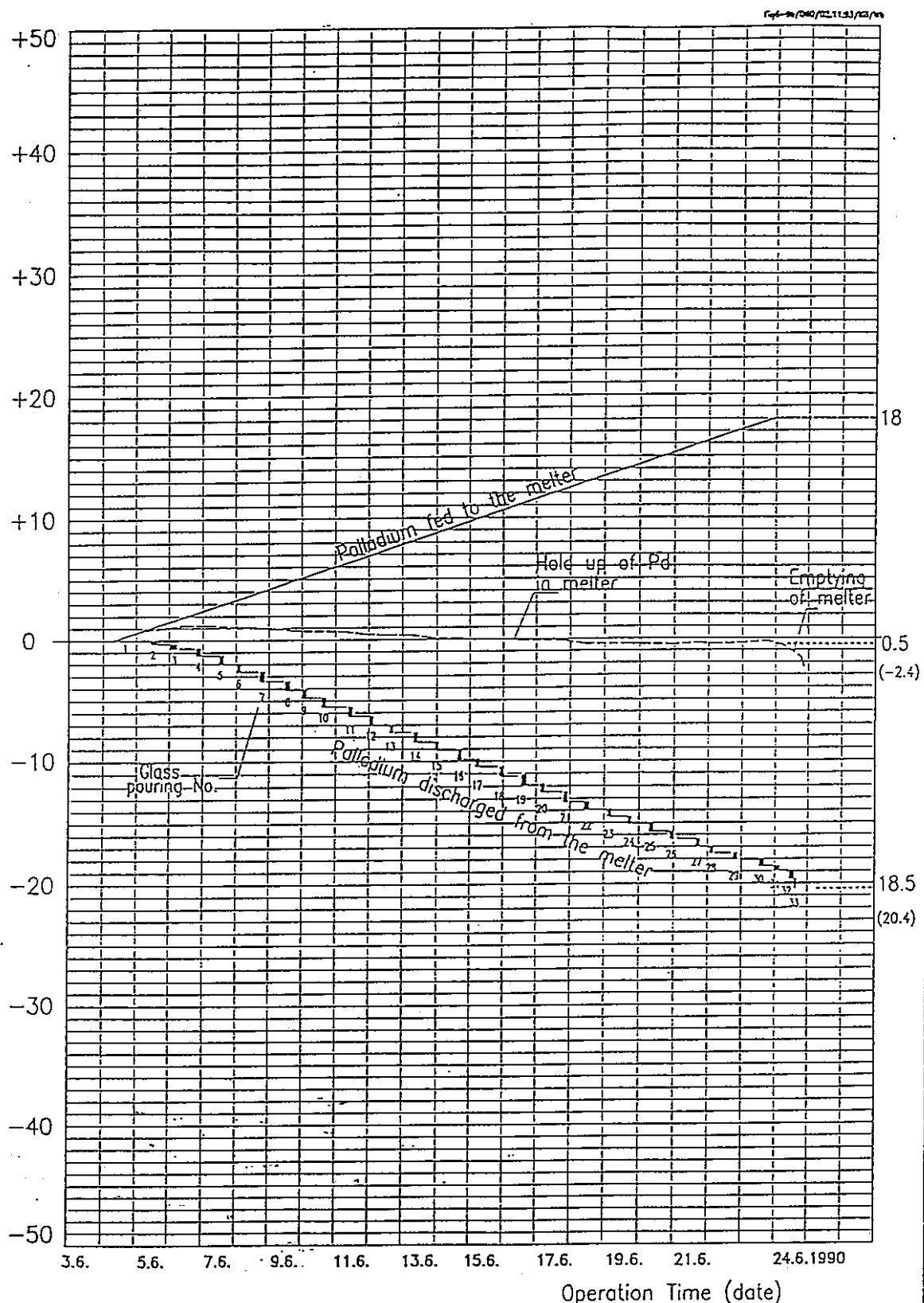


Fig.6-9b: Input and removal of Palladium from the K-6' melter during the long-term noble metals test run D2

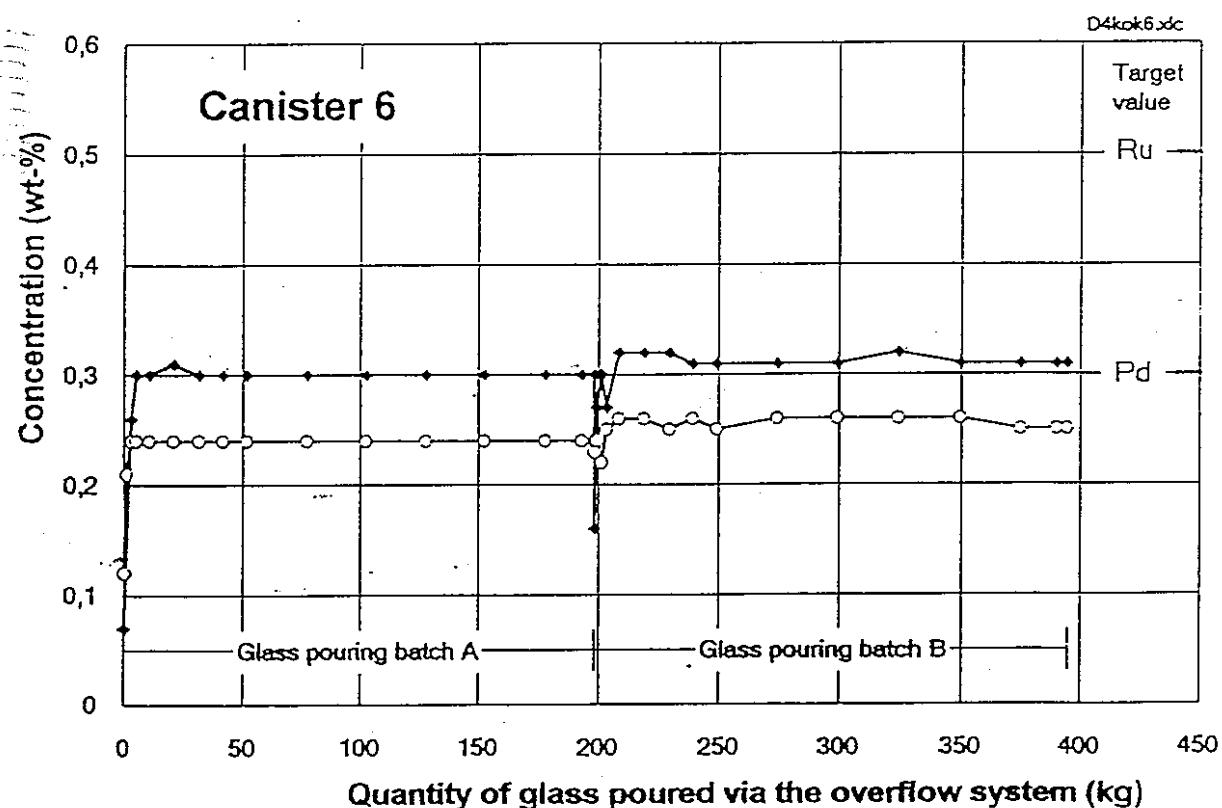
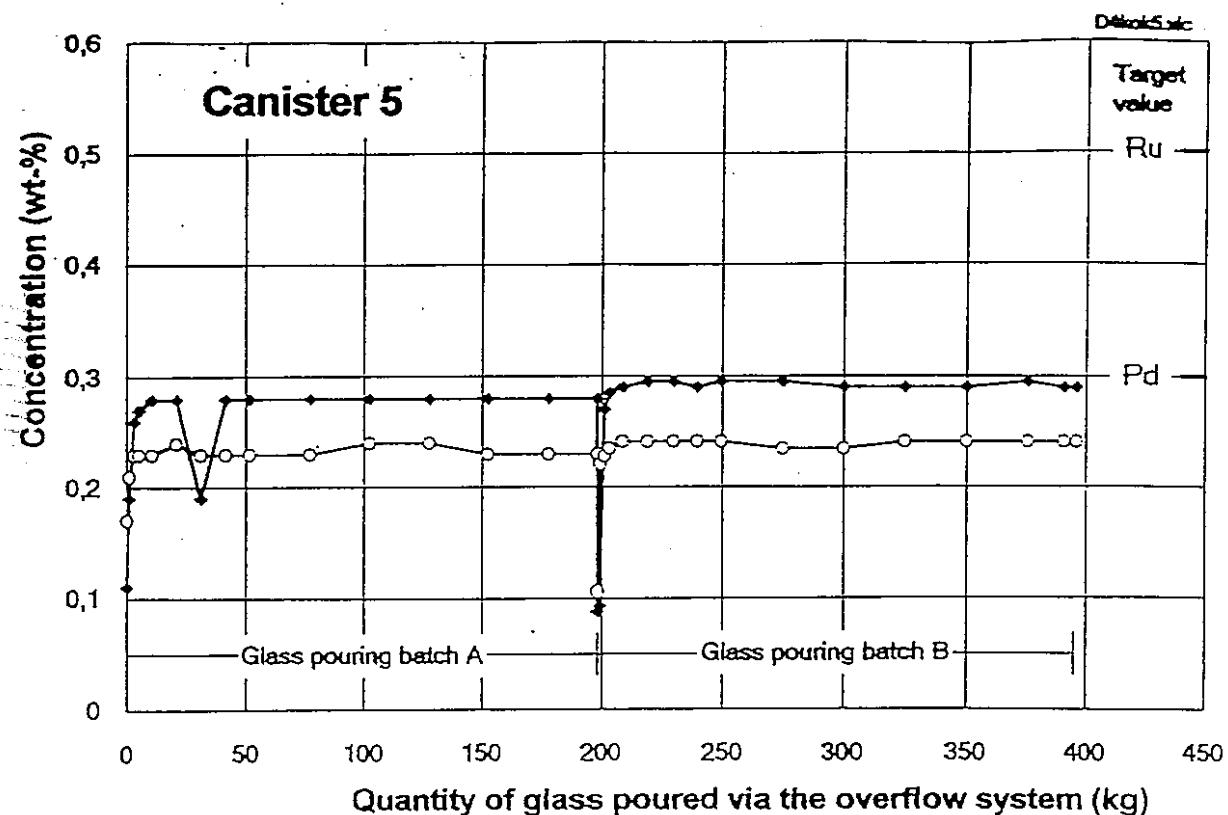


Fig. 6-17c: Noble metal concentrations in glass samples taken from pouring stream while filling canister 5 and 6 of the D4 test run

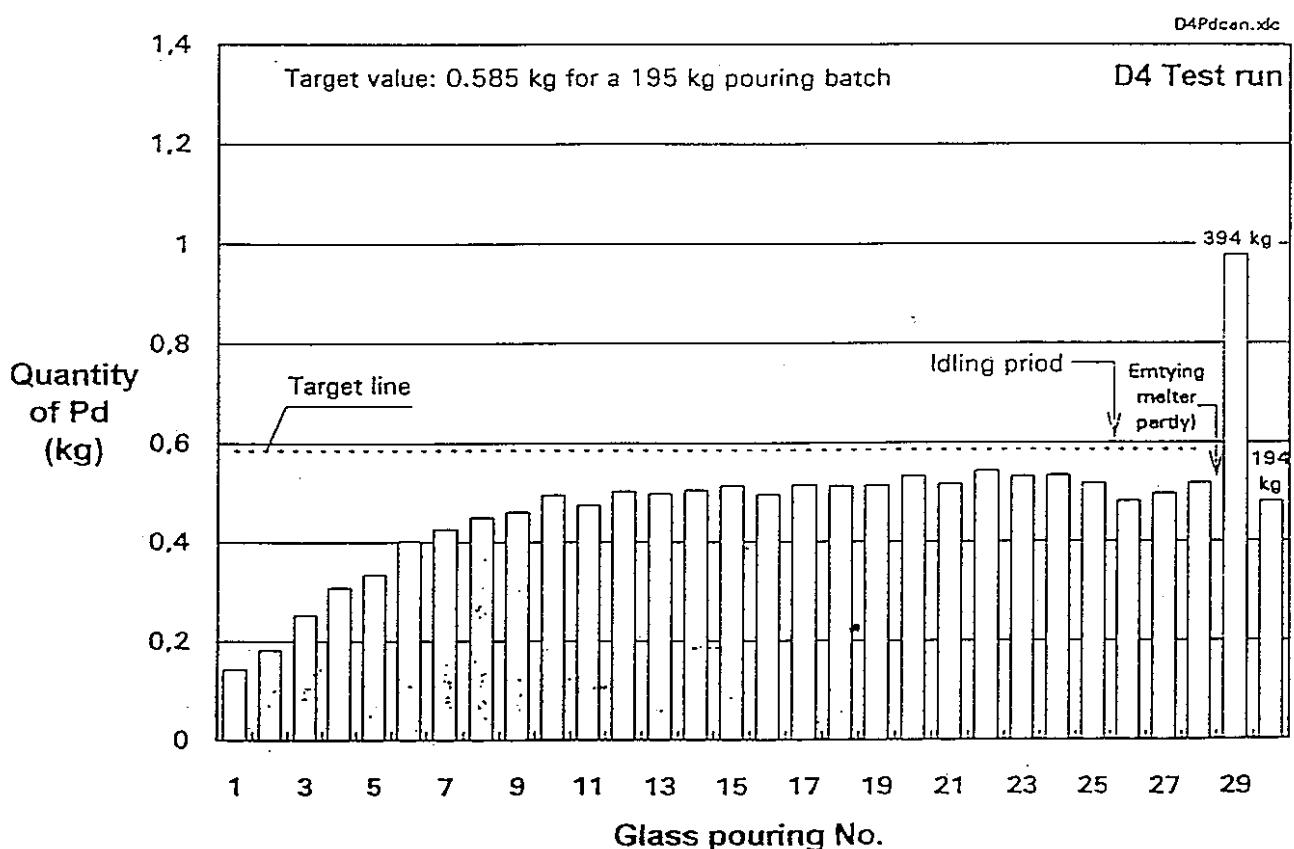
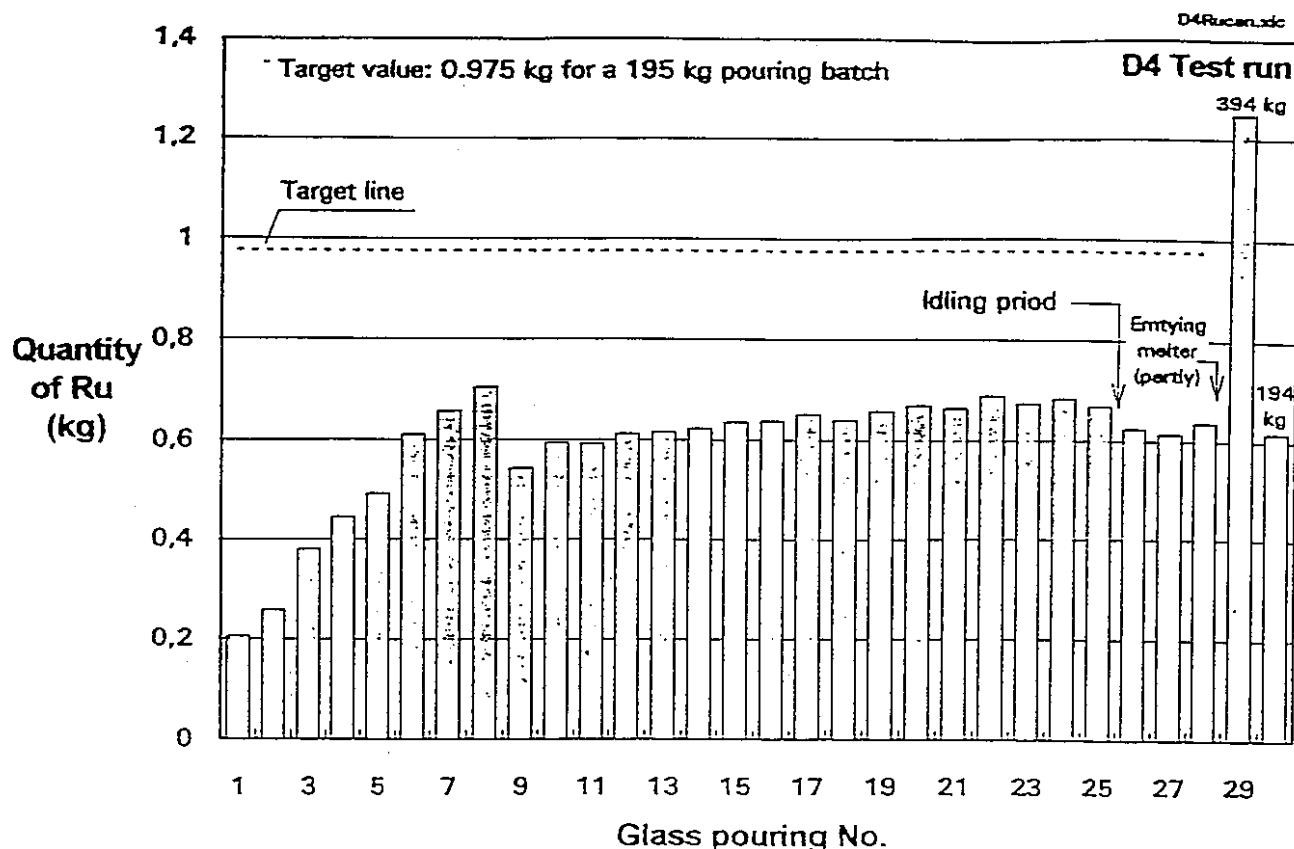


Fig. 6-18: Quantity of Ruthenium and Palladium, respectively, removed from melter by glass pouring via the overflow system. Test run D4, June 1991

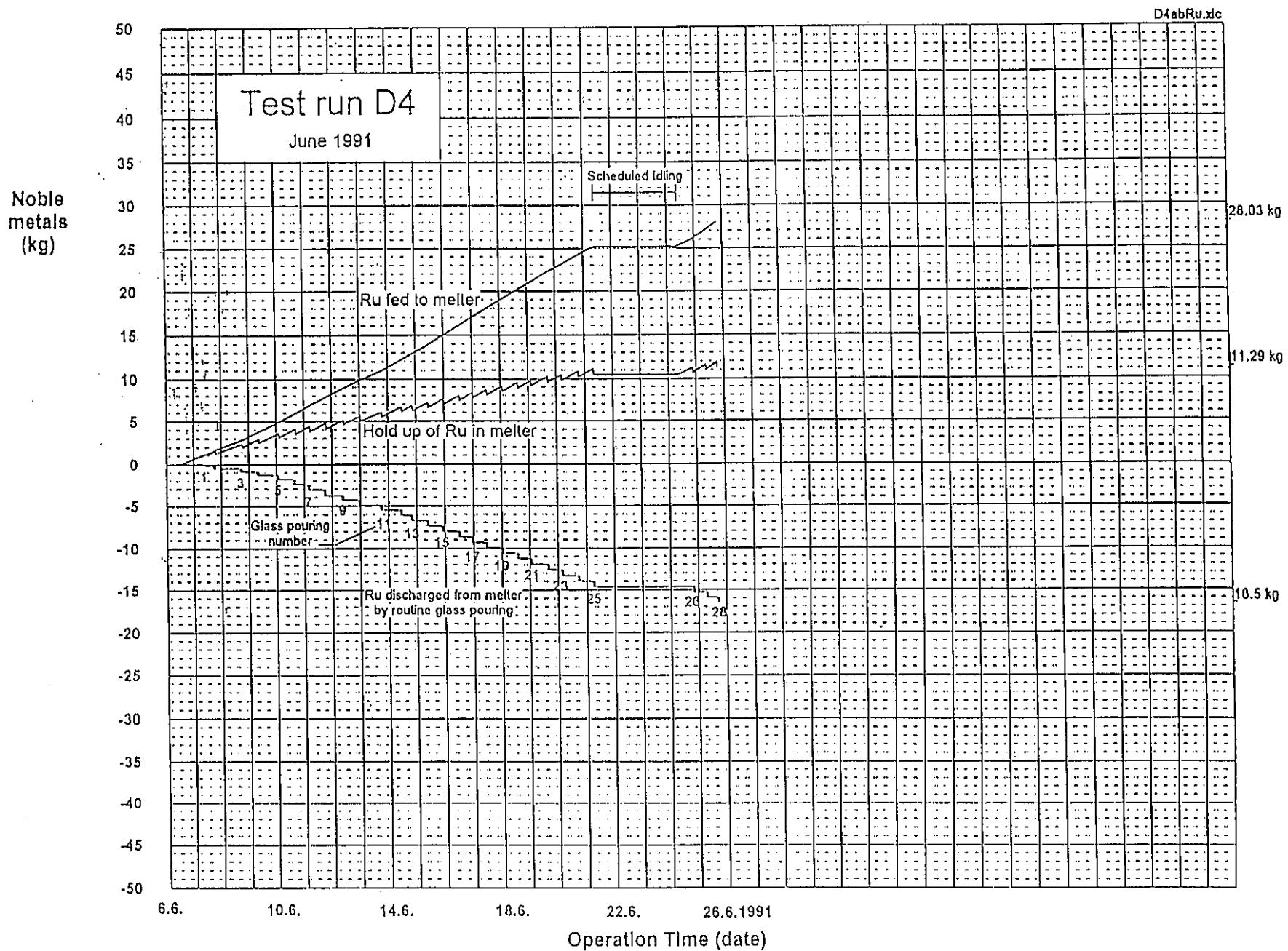


Fig. 6-19 : Input and removal of Ruthenium from the K-6' melter during the long-term noble metals test run D4

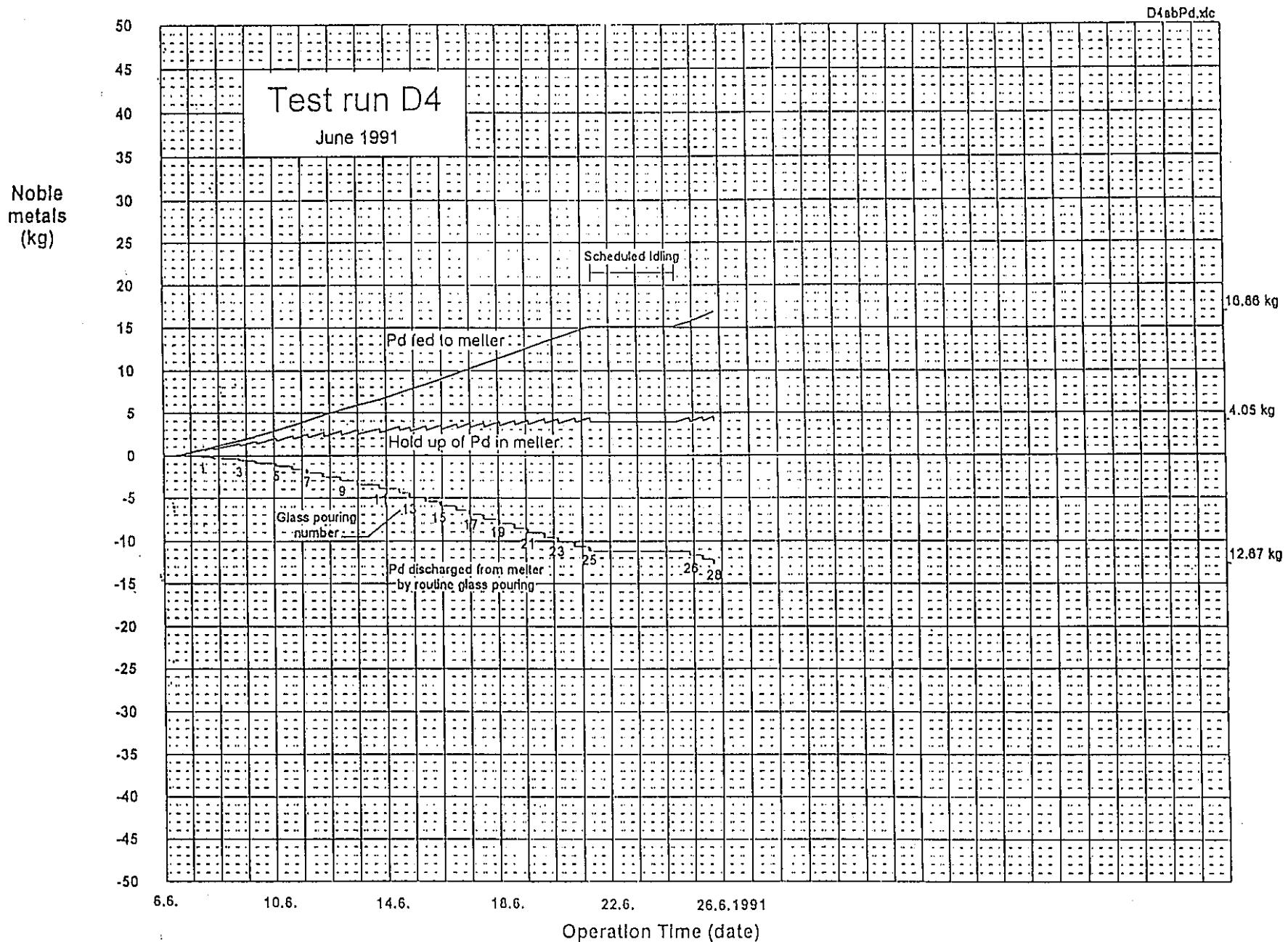


Fig. 6-20 : Input and removal of Palladium from the K-6' melter during the long-term noble metals test run D4

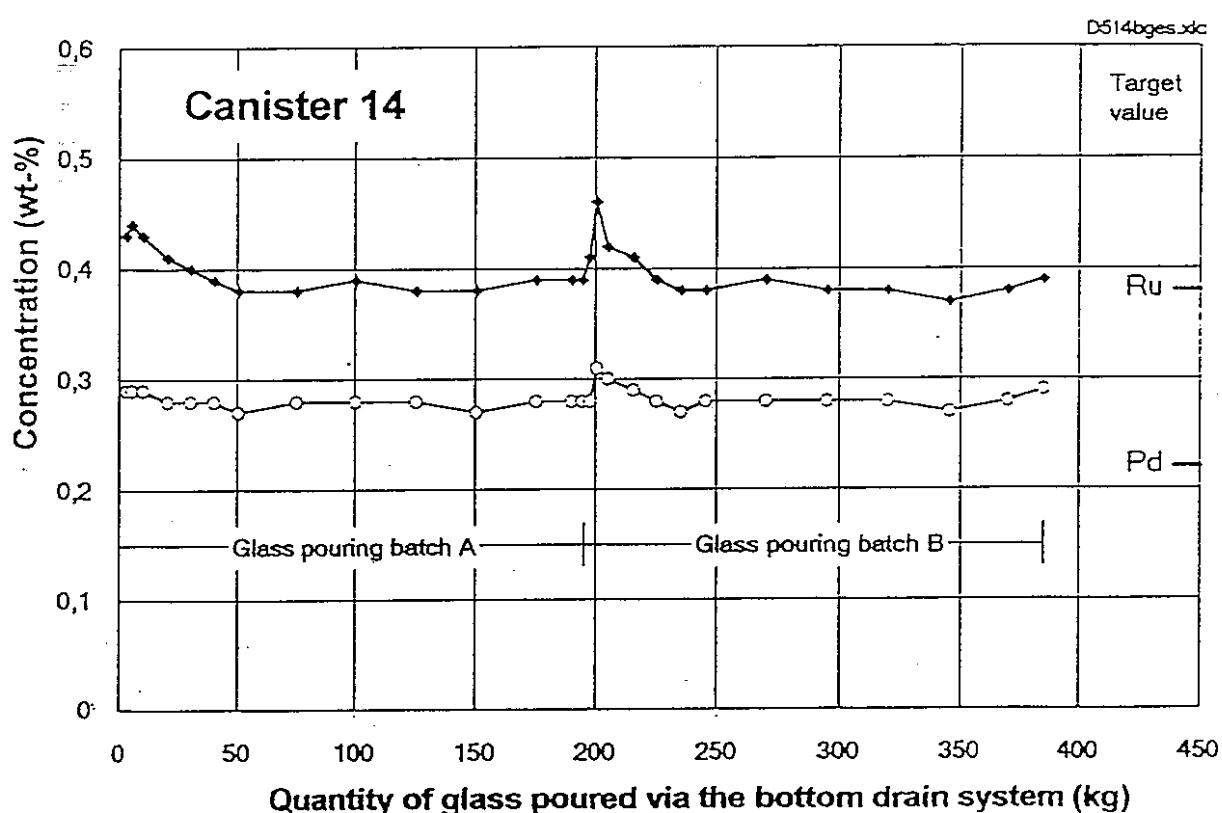
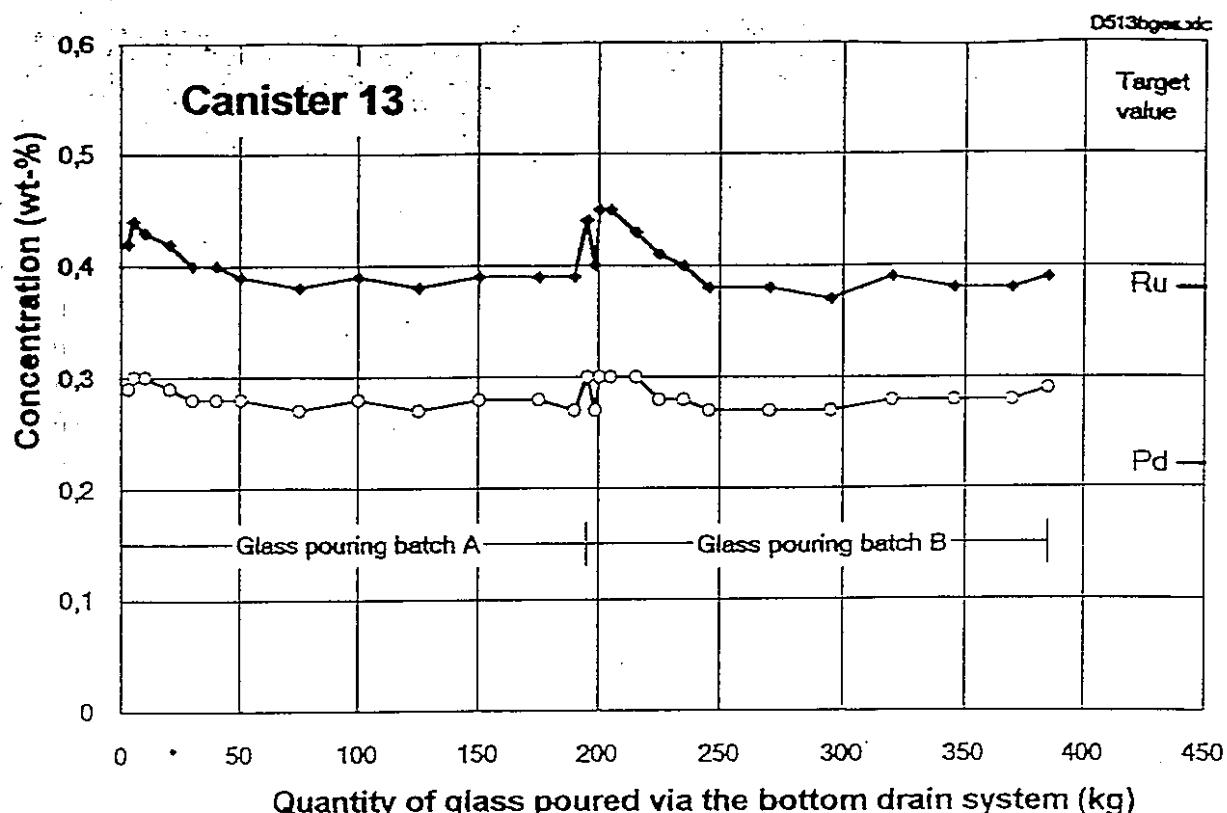


Fig. 7-7g : Noble metal concentrations in glass samples taken from pouring stream while filling canister 13 and 14 of the D5 test run

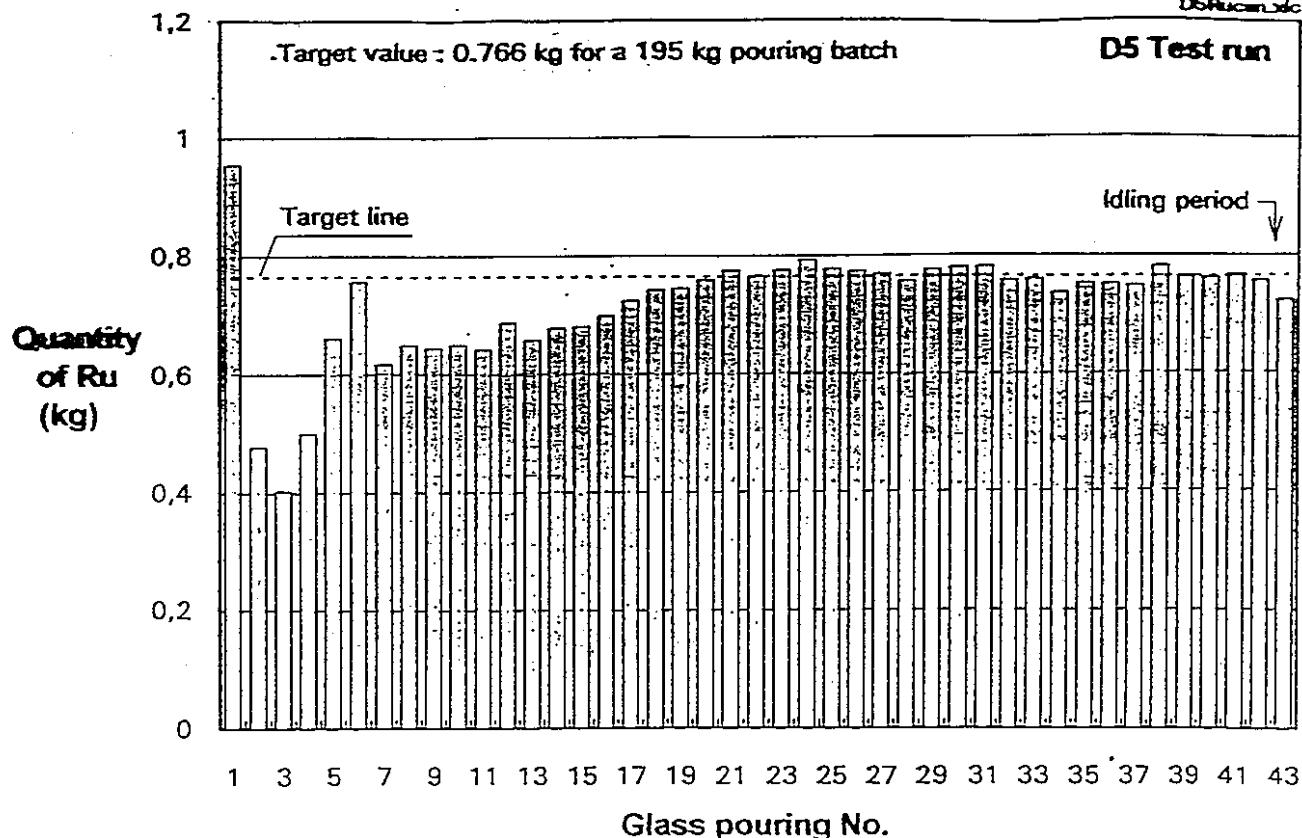


Fig. 7-8: Quantity of Ruthenium removed from melter by glass pouring via the bottom drain system.  
Test run D5, February/March 1992.

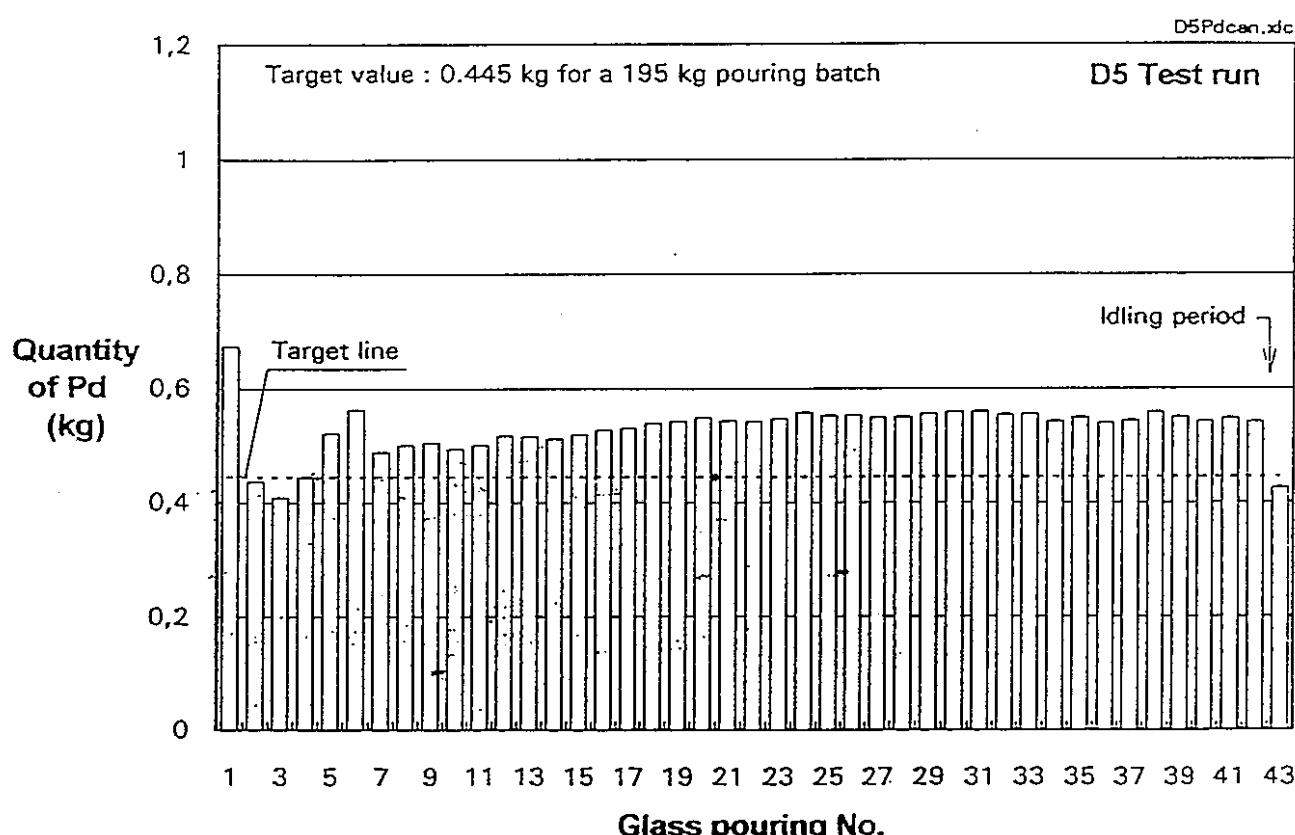


Fig. 7-9: Quantity of Palladium removed from melter by glass pouring via the bottom drain system.  
Test run D5, February/March 1992.

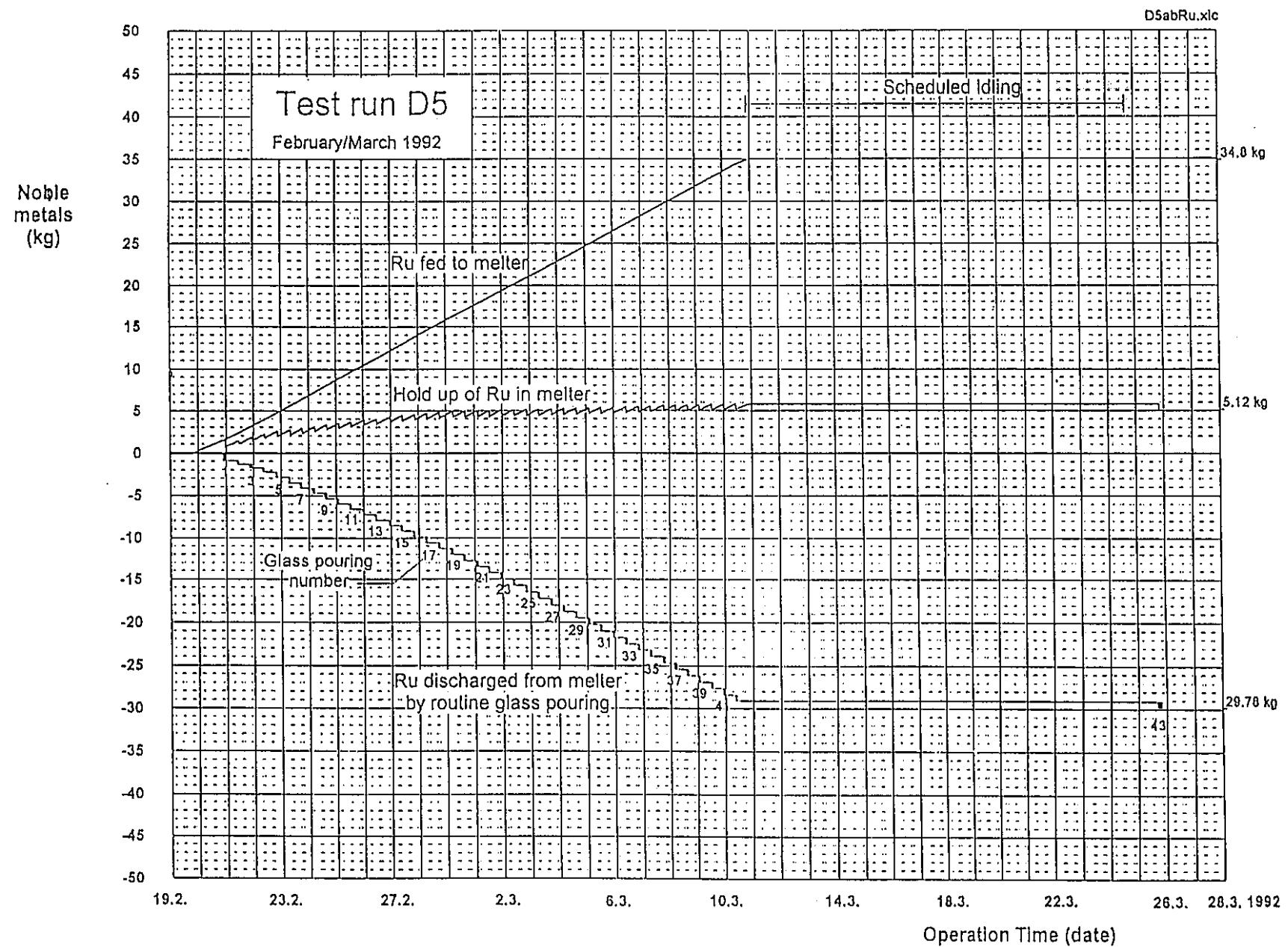


Fig. 7-10a : Input and removal of Ruthenium from the K-6' melter during the long-term noble metals test run D5

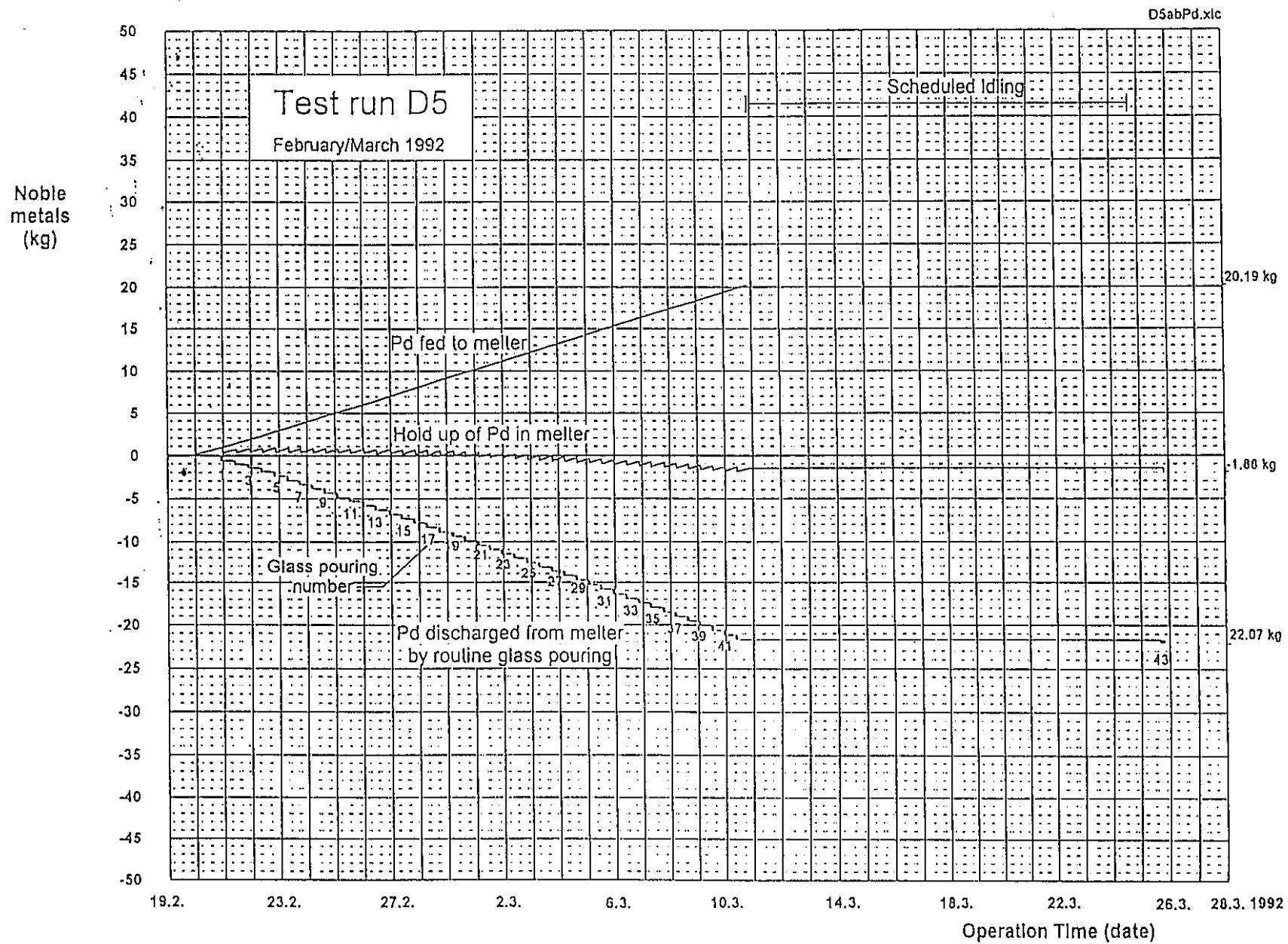


Fig. 7-10b : Input and removal of Palladium from the K-6' melter during the long-term noble metals test run D5

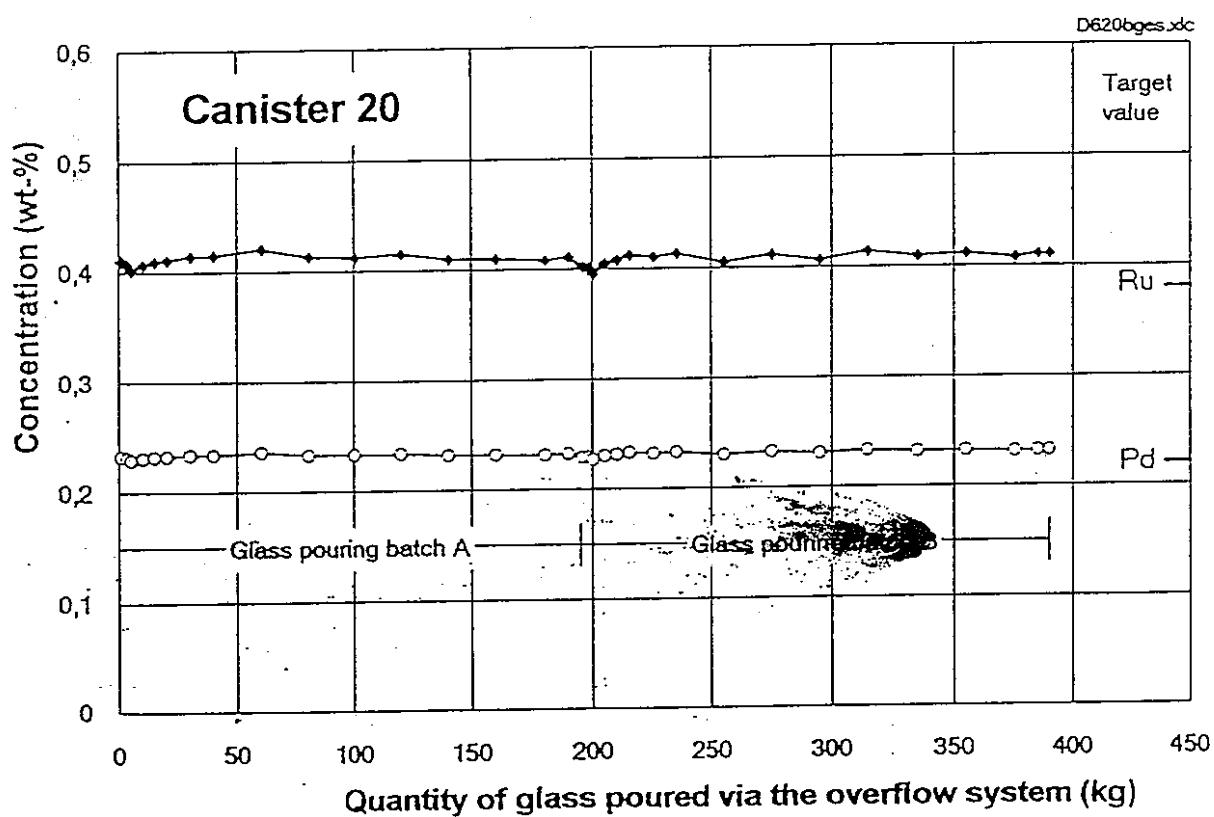
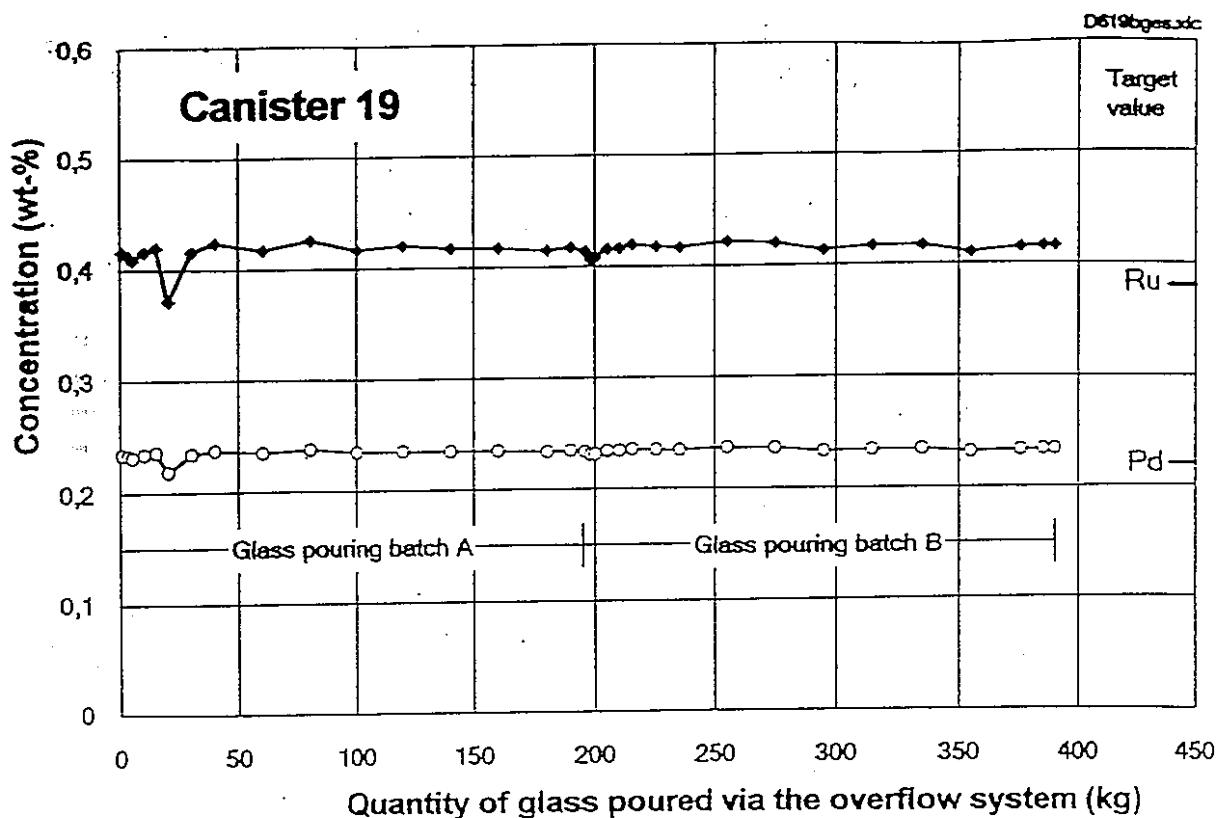


Fig. 7-22j : Noble metal concentrations in glass samples taken from pouring stream while filling canister 19 and 20 of the D6 test run

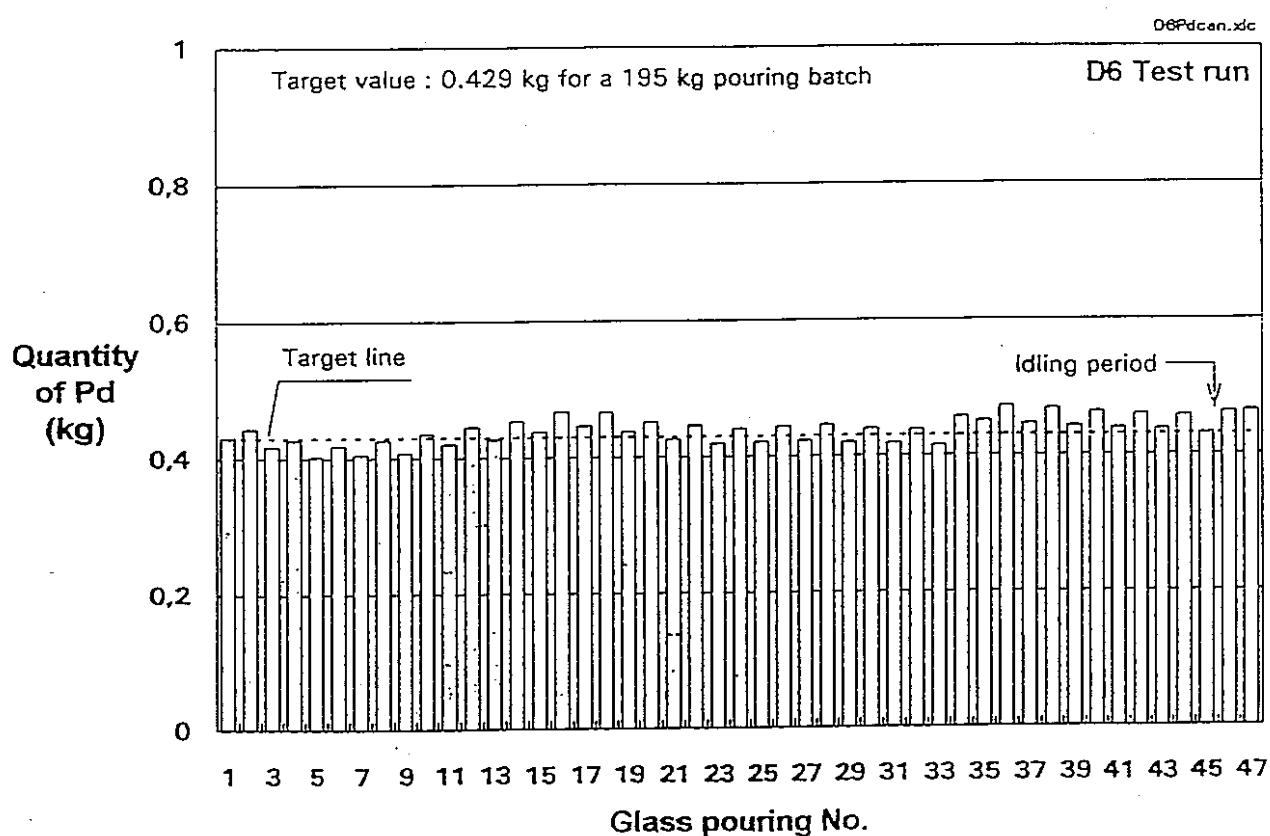
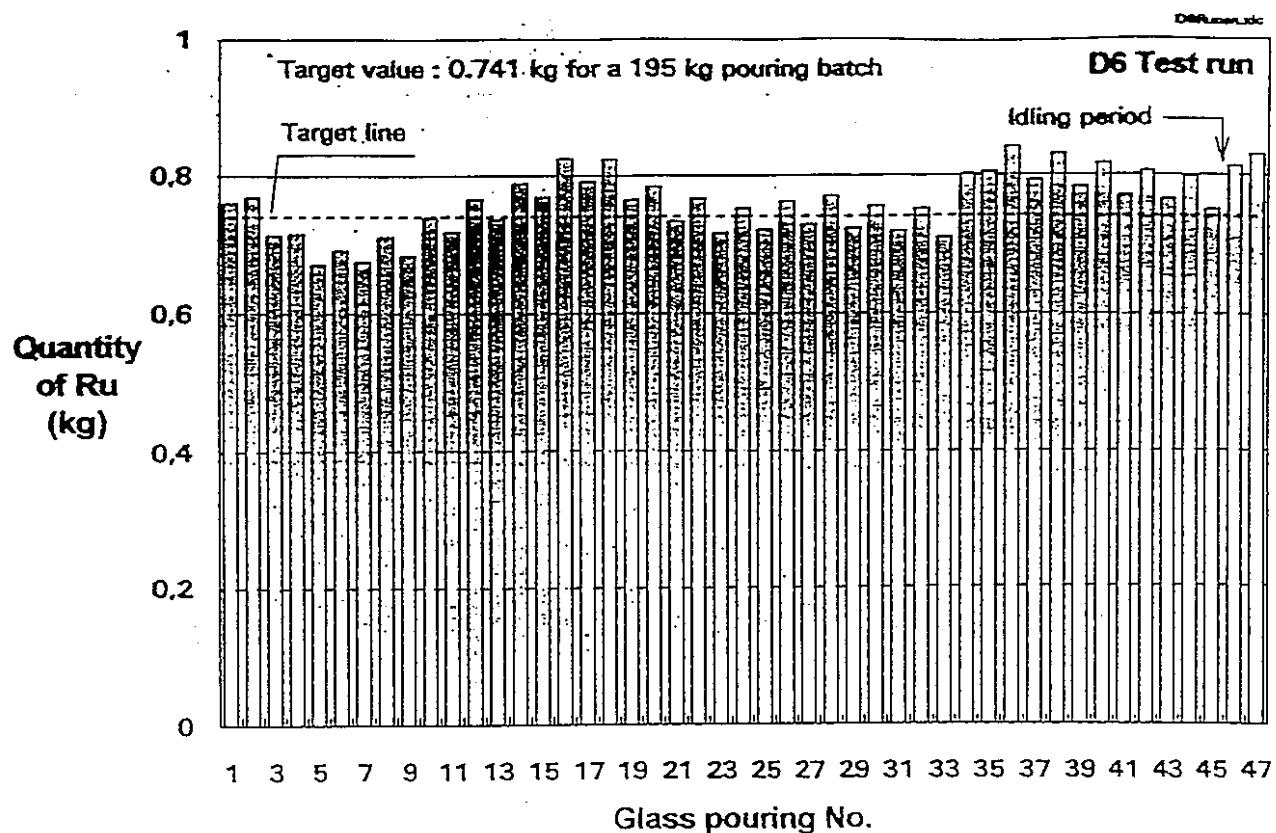


Fig. 7-23: Quantity of Ruthenium and Palladium, respectively, removed from melter by glass pouring via the overflow system. Test run D6 May/June 1993

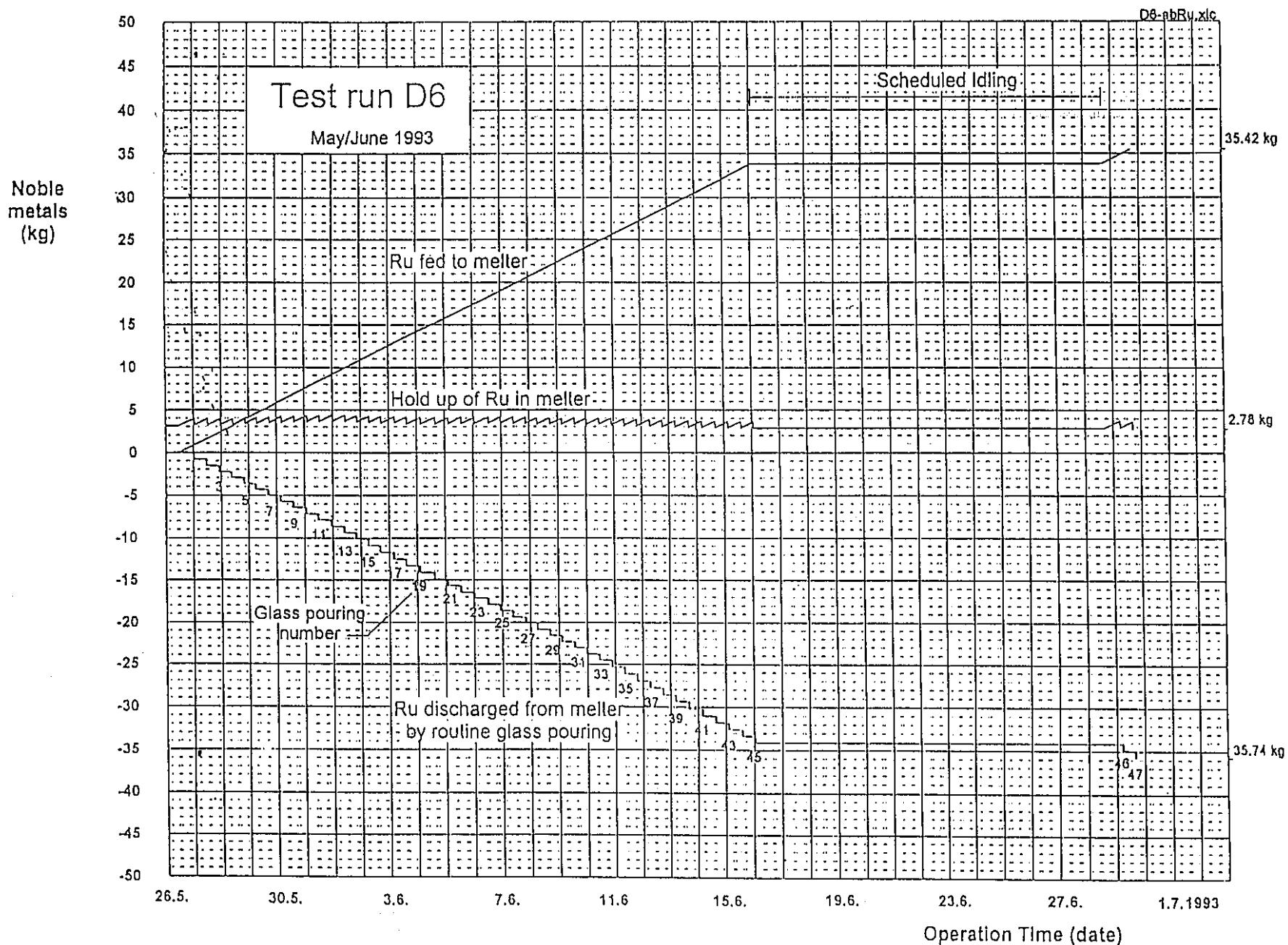


Fig. 7-24: Input and removal of Ruthenium from the K-6' melter during the long-term noble metals test run D6

D6-abPd.xls

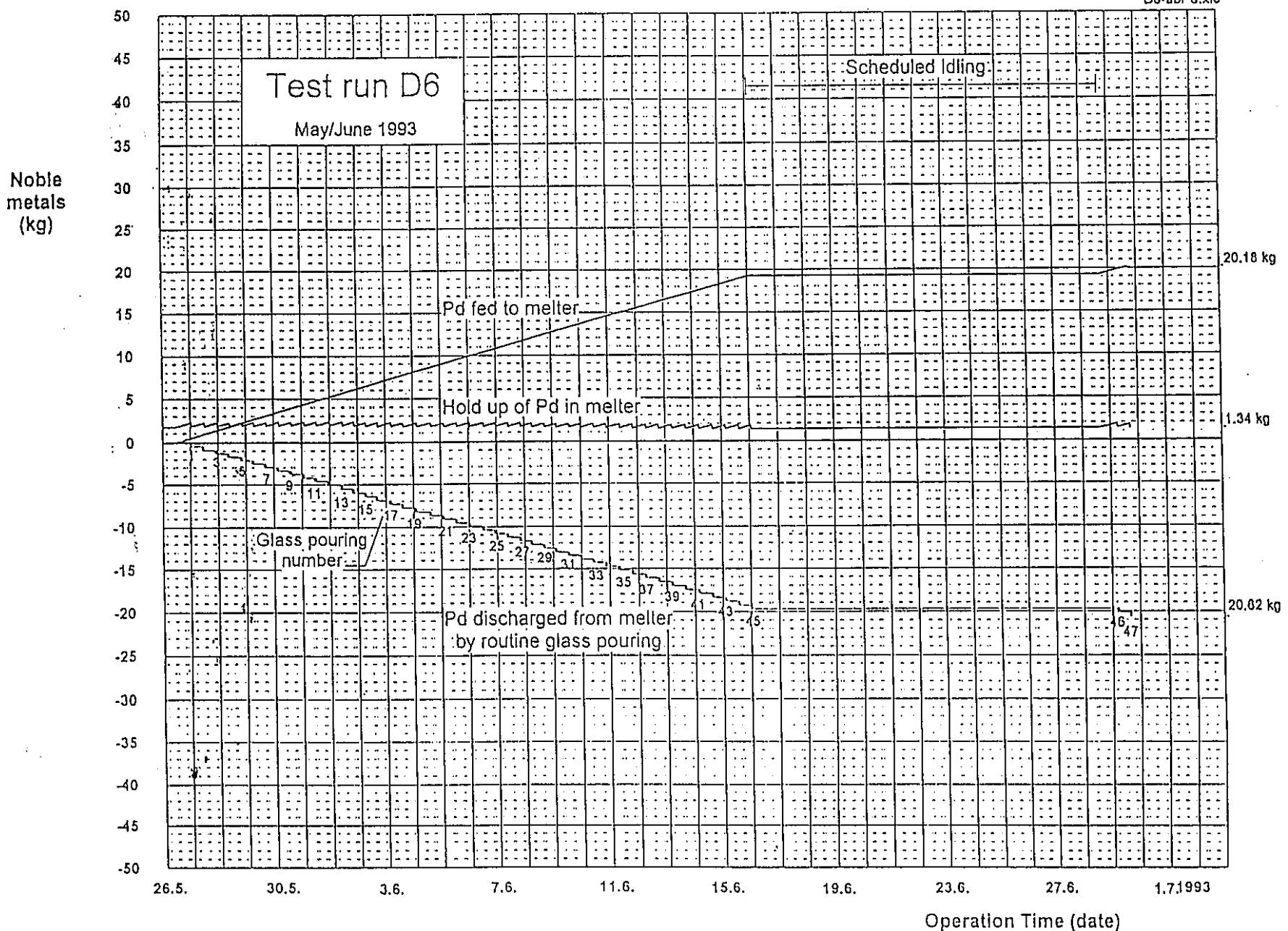


Fig. 7- 25 : Input and removal of Palladium from the K-6' melter during the long-term noble metals test run D6

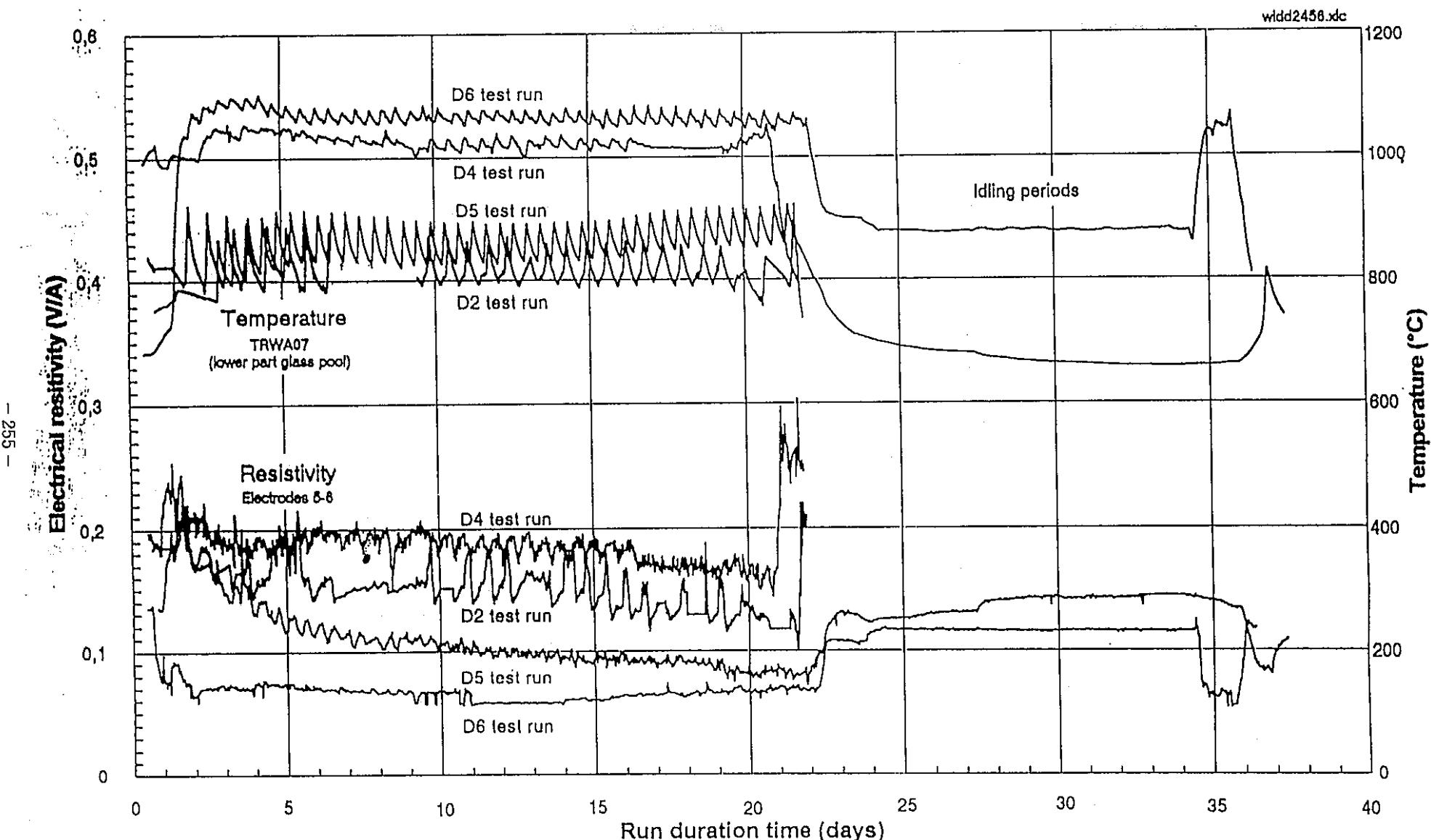
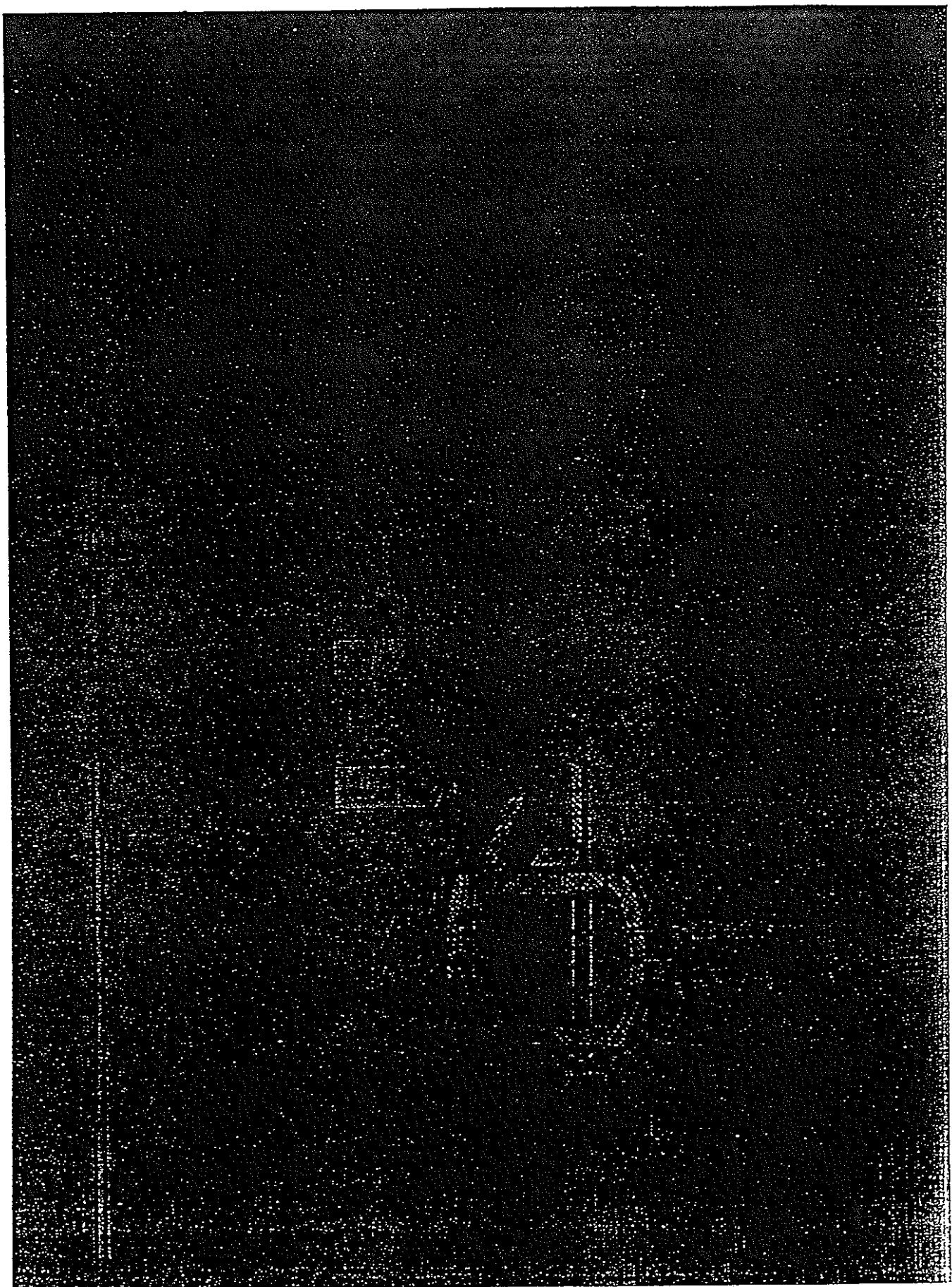
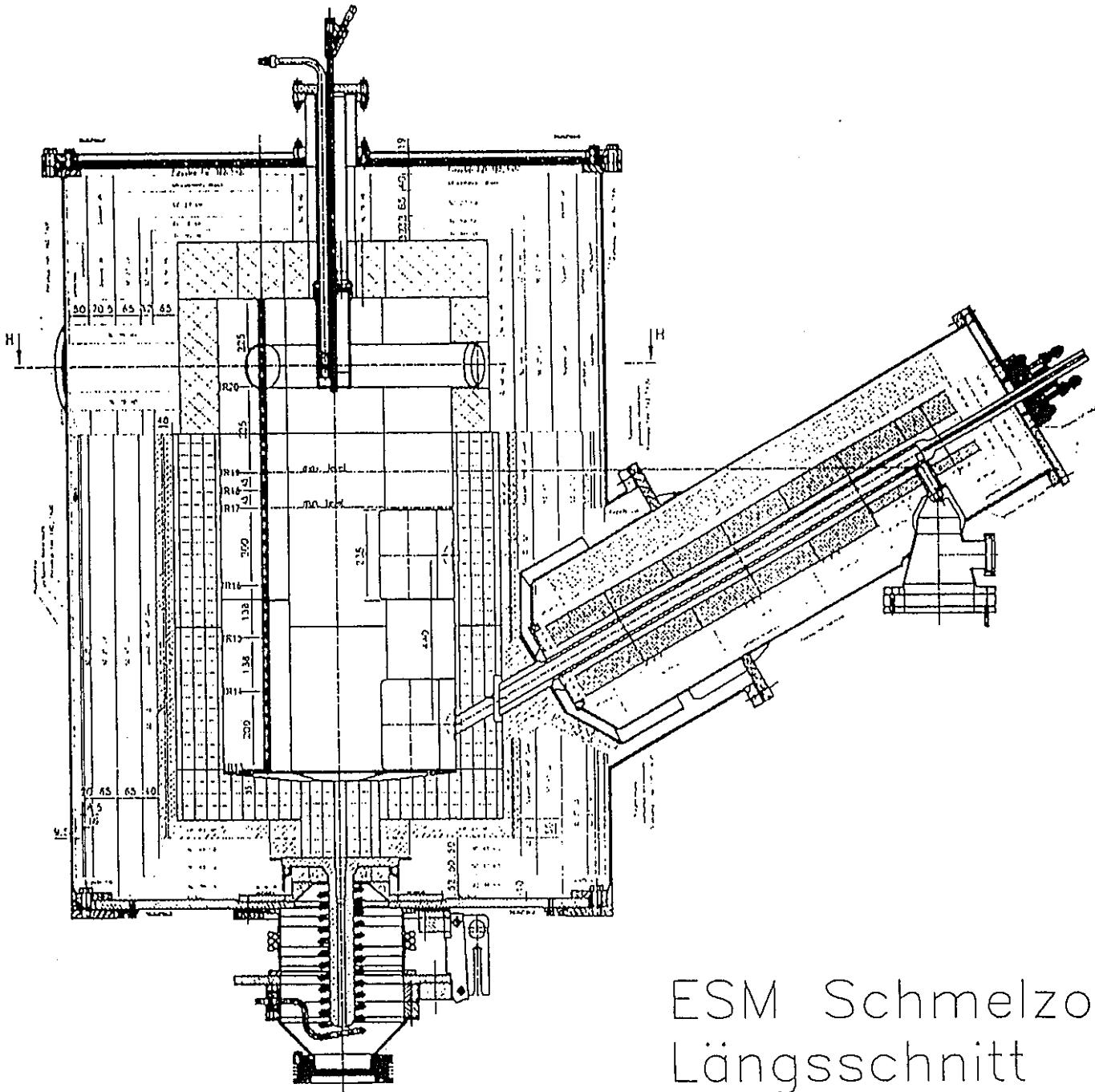


Fig. 7-28: Electrical resistivity curves of the test runs D2, D4, D5 and D6 obtained for the electrode pair 5-6.  
Included are key temperature curves for the lower part of the glass pool.

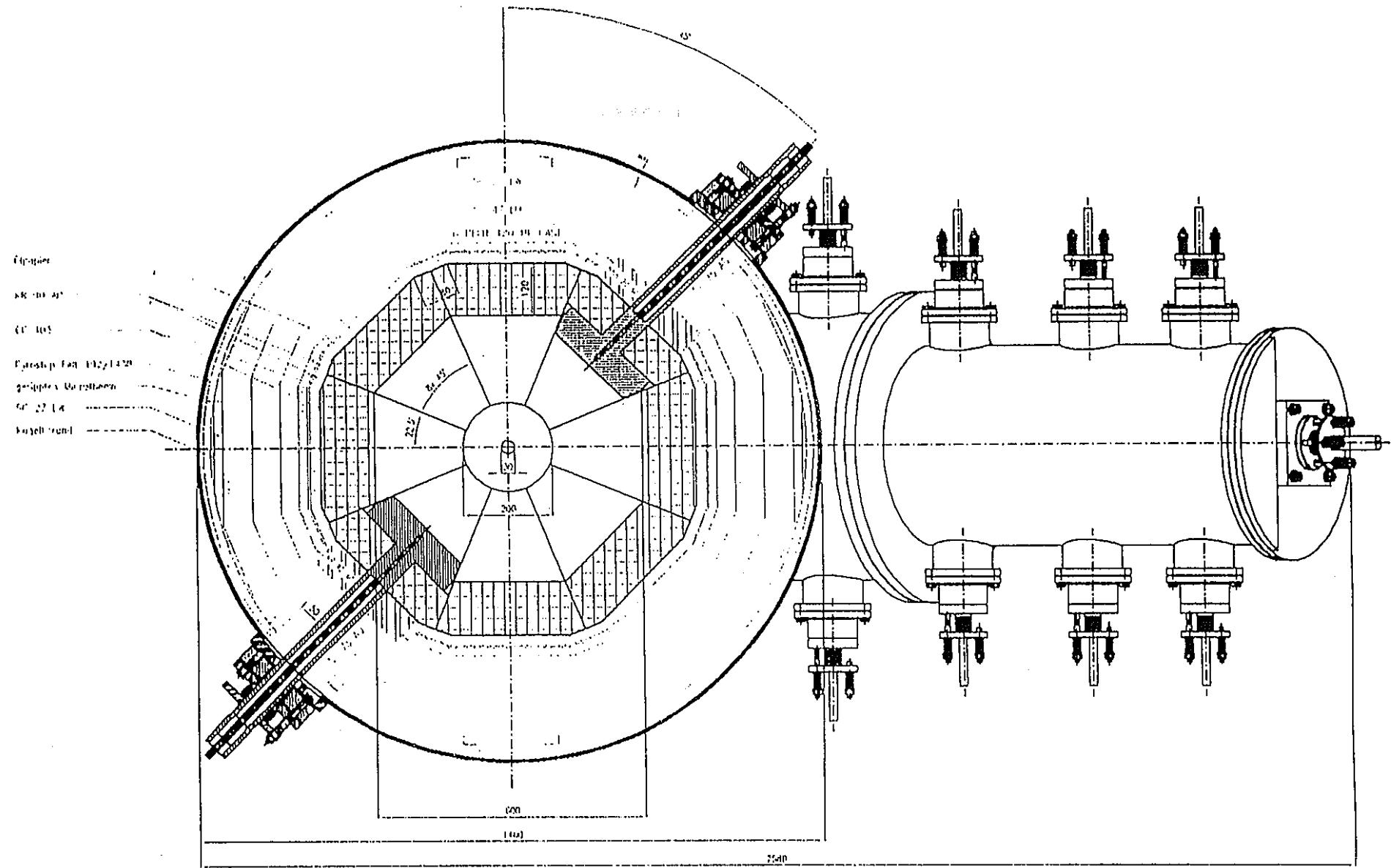


## Layout data ESM - Melter

	Full-scale melter	1/10-melter (ESM)
glass pool volume	2383 l	234 l
surface area	2.63 m <sup>2</sup>	0.298 m <sup>2</sup>
glass pool depth	0.86 m	0.78 m
feed rate	222 l/h (100 kg/h)	20 l/h (10 kg/h)
residence time	58.5 h	58.5 h



# ESM Schmelzofen Längsschnitt



ESM Schmelzofen, Horizontalschnitt

## Compilation of the main test run data

	Pretest	Noble metal test
Design feed rate	20 l/h	20 l/h
Corresponding glass production rate	10 kg/h	10 kg/h
Melting temperature	1150 °C	1150 °C
Temperature of heater pipes	900 °C	900 °C
Amount of feed	3 m³	14 m³
Oxide yield	139 g/l	140 g/l
Recycle	17 g/l	17 g/l
Glass frit	343 g/l	343 g/l
Total oxides	494 g/l	500 g/l
Waste glass loading	30.7 wt %	31 wt %
Amount of glass frit to be used	0.92 to	4.31 to
Glass production	1.3 to	6.3 to
Noble metals amount for this run		
Ruthenium	-	5.996 kg
Palladium	-	2.069 kg
Rhodium	-	1.646 kg
Number of glass canisters	4	17
Canister type	$\phi$ 430 x 1350 mm (400 kg maximum capacity)	
Number of pouring operations (per canister)	5 (75 kg each pouring)	
Glass sampling	25 samples are taken filling one canister	
Total Glass samples	90	420

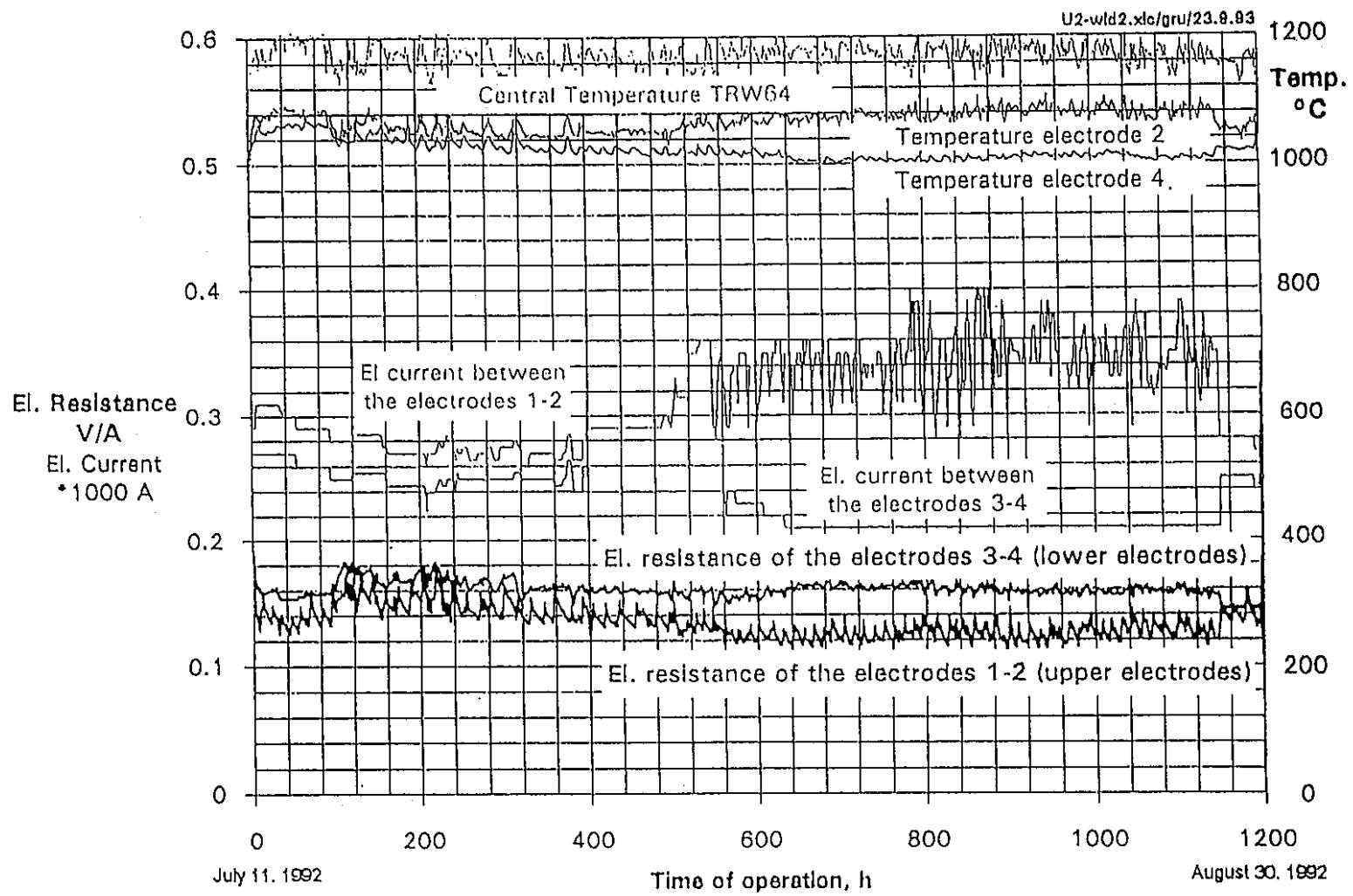


Fig. 5-17 Electric data of the ESM monitored by the PISA system during the U2 test run

3.4 Gravis process control and data acquisition system

# **GRAVIS**

## **Graphical Visualization and Information System**

Applied in the V-W1 vitrification mock-up test facility

Developed by KfK-INE

### Features

- Acquisition of process data
- Process visualization
- Determination of process-relevant parameters
- Calculation of mass balances
- Long-term storage of data
- Documentation

Experience available from two long-term test runs

## **INTOUCH Process control software**

Based on Windows/Windows NT

Modern man/machine interface

Easy to configure

Exceptional visualization tools

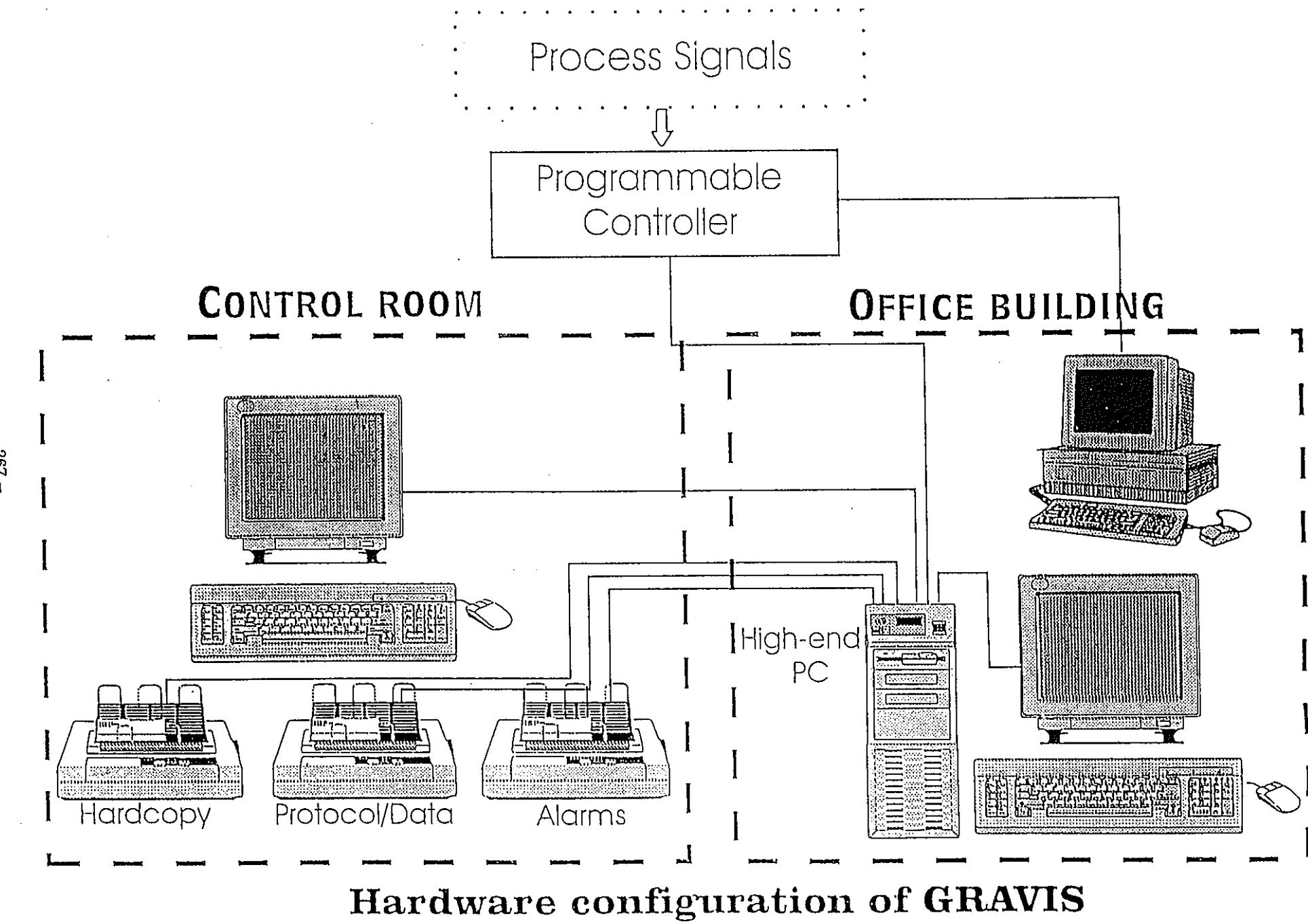
Historical data management

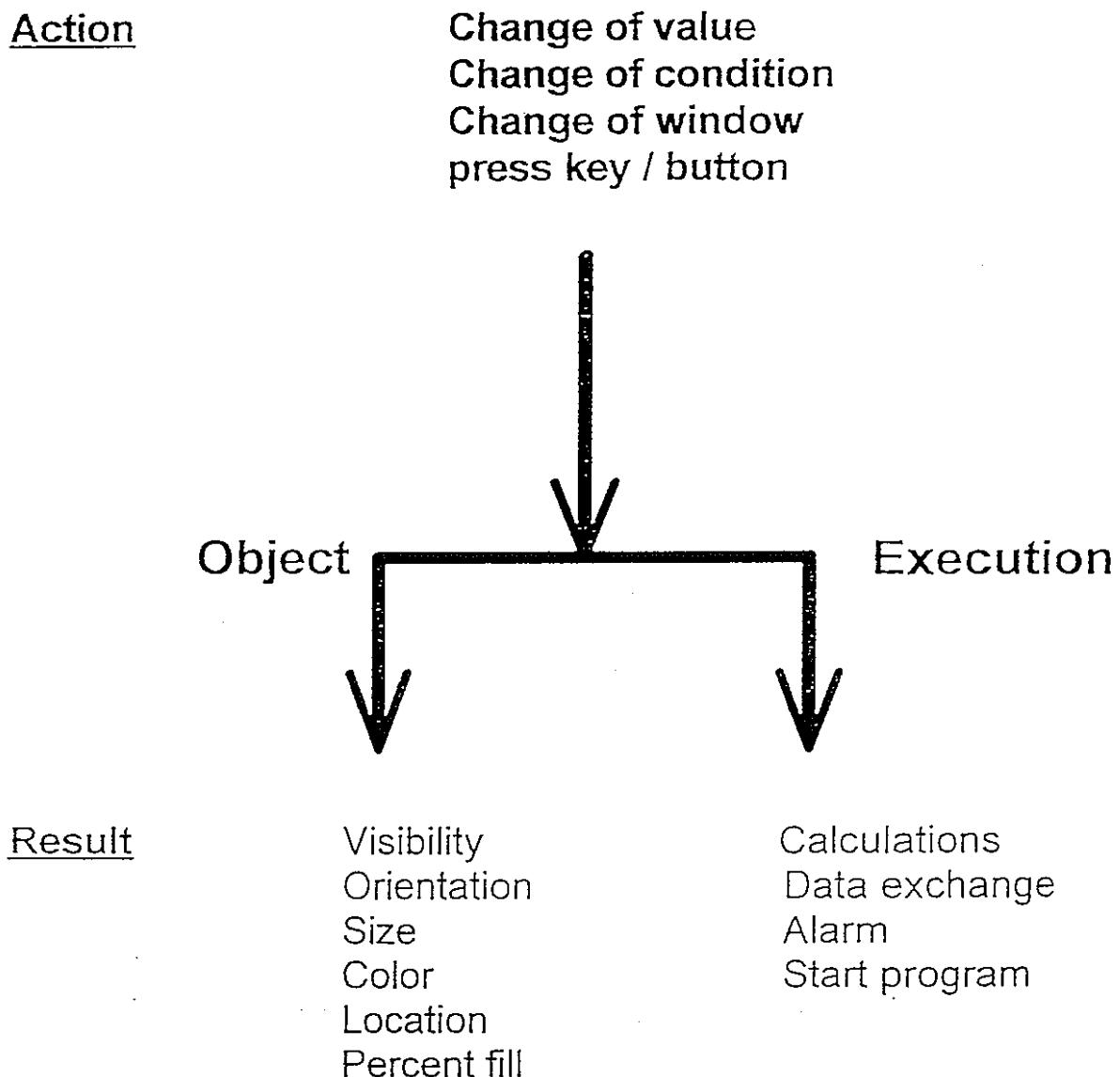
Real time/historical trending

Event-based background actions (calculations etc.)

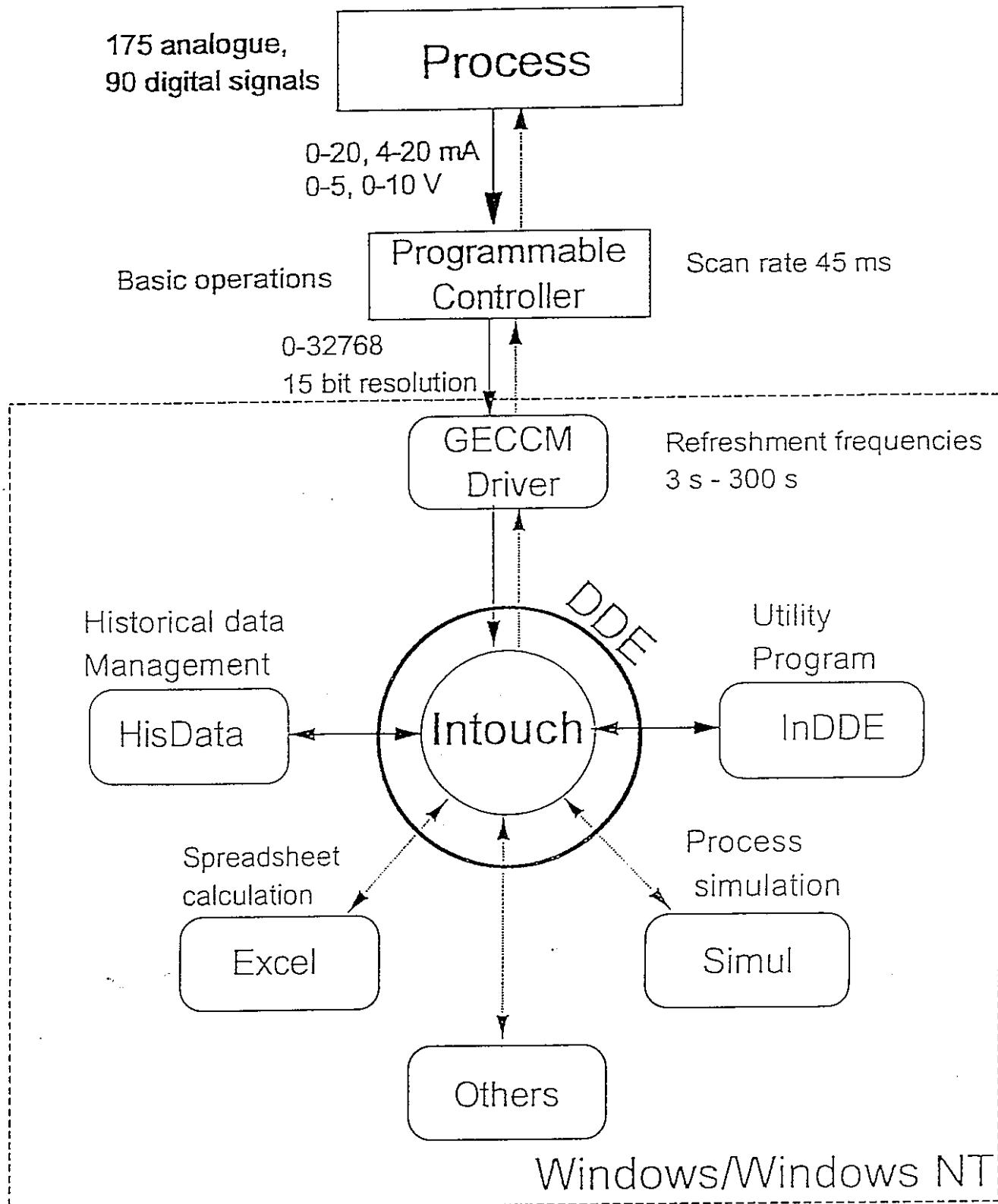
Parallel development/runtime execution

Easy data exchange with other programs/computers



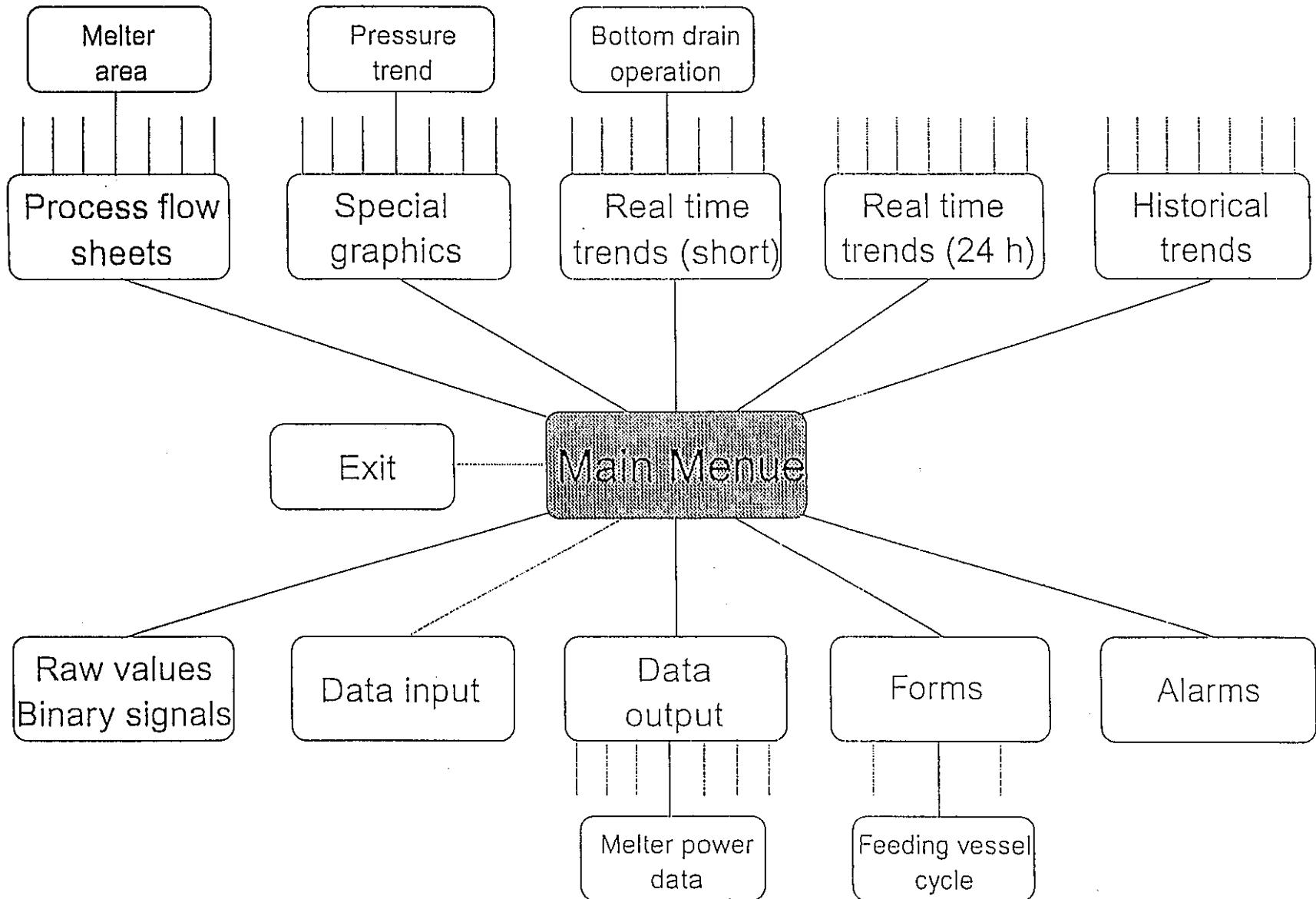


Examples of the dynamic features of InTouch



DDE="Dynamic Data Exchange"  
(Data exchange protocol)

## Software structure of GRAVIS



Graphical user interface of GRAVIS

# GRAVIS Calculations

## Balances

- HAWC balance via feeding vessel
- HAWC balance via intermediate tank
- Recycle balance via feeding vessel / dust scrubber
- Glass frit balance
- Melter hold-up balance
- Secondary liquid waste balance

## Others

- Determination of feeding rate
- Pressure drop along the off-gas line
- Melt level
- Electrical resistance
- Waste loading

3.5 Measurements of DF data for Sr, Cs and Ru for the melter  
and the off-gas line

**KfK-INE**

**PNC-KfK meeting**

# **Measurements od DF data for Sr, Cs and Ru for the melter K-6' and the off gas line**

S. Weisenburger

## Topics

- System description, HAWC composition
- Variation of processing conditions
- Test results, depending on processing conditions
- Conclusions

21.1033/M45/10/7-1/22

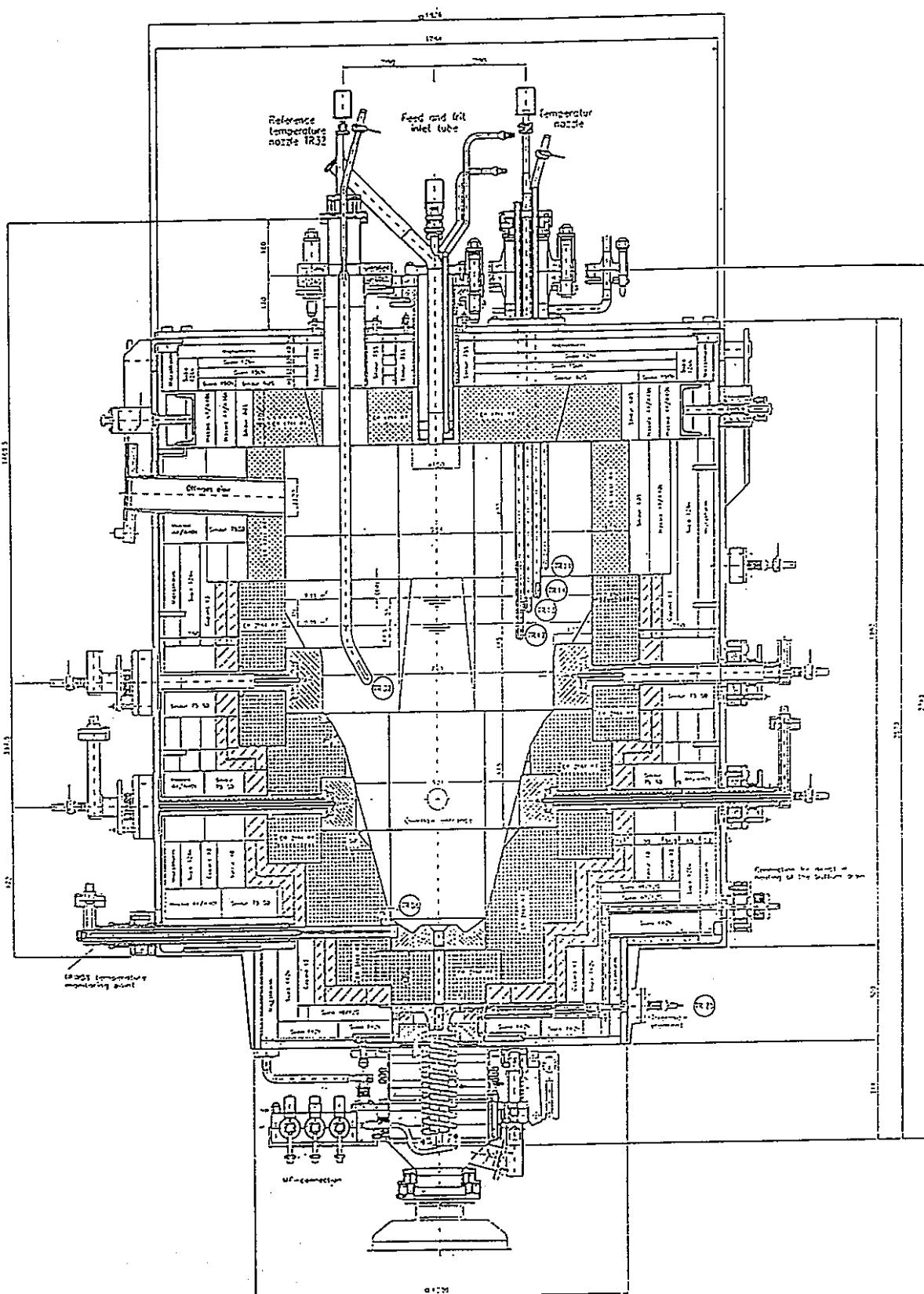


Fig.4-7: Transverse vertical cross section of the K-6' meller with the location of the most important temperature monitoring points TR08,TR11,TR12,TR13,TR14,TR25 etc. and TR32

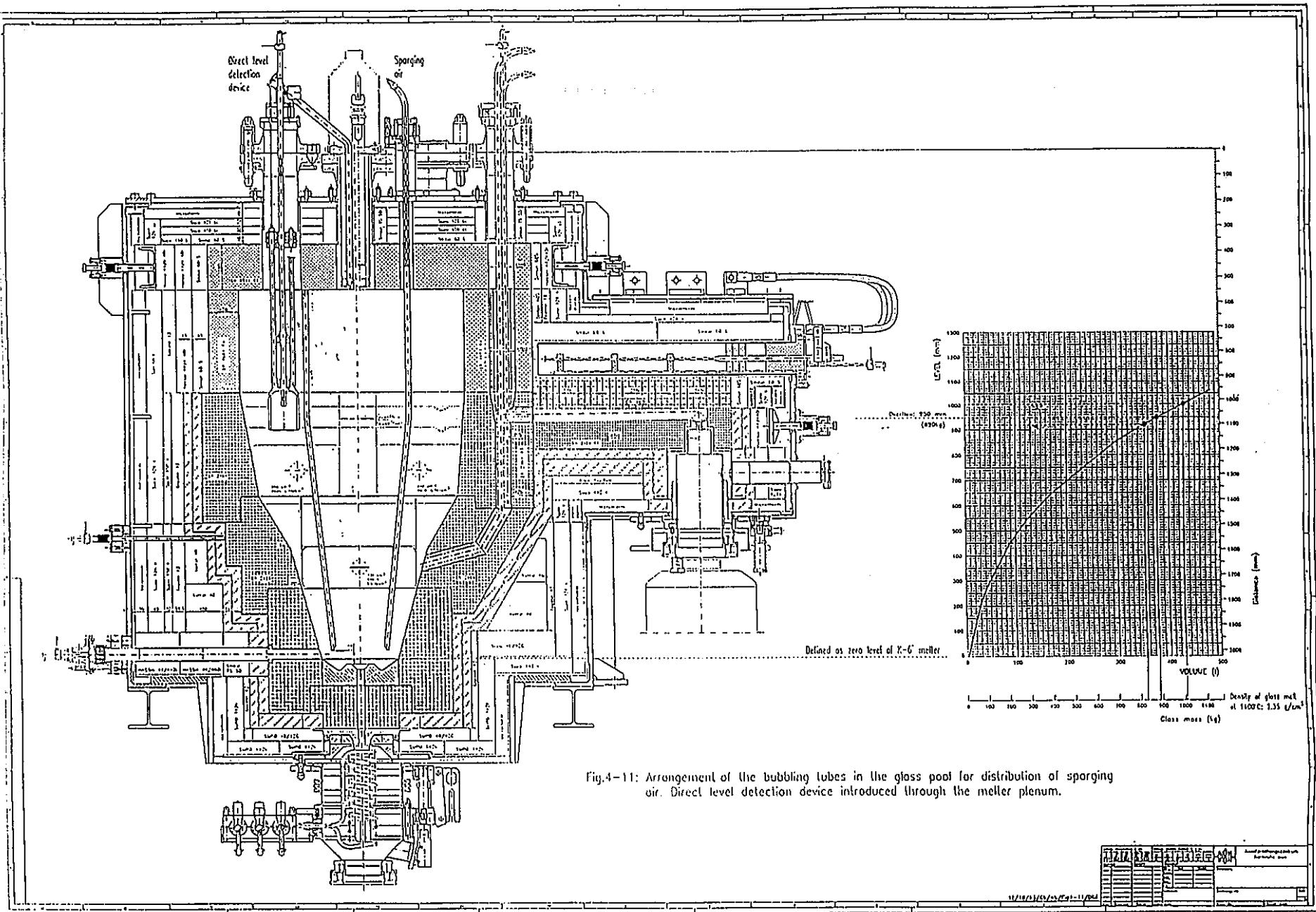


Fig.4-11: Arrangement of the bubbling tubes in the glass pool for distribution of sparging air. Direct level detection device introduced through the meller plenum.

Table 8-IX

Compilation of DF-data obtained from the test runs D2, D4, D5 and D6 of the HAWC-technology program

1278

Component	DF-data obtained in the several test runs																			
	Test run D2					Test run D4					Test run D5				Test run D6					
	Cs	Sr	Ru	Ce	Se	Cs	Sr	Ru	Ce	Se	Cs	Sr	Ru	Nd	Ce	Cs	Sr	Ru	Pd	Na
Melter	19.8	65.1	9.5	91	4.0	8.77	41.3	-	258	1.7	59	228	29	345	449	36.7	96	19.8	182.5	63.5
Dust scrubber	3.0	14.0	31.0	34	4.8	1.92	2.7	-	2.3	2.0	2.3	8.1	5.5	14.4	18.8	2.04	5.16	6.72	4.60	3.31
Condenser	1.40	1.35	2.10	1.2	1.42	4.39	7.2	-	2.2	2.4	1.6	1.5	3.2	1.6	1.5	1.61	1.65	2.01	1.58	1.54
Jet-scrubber	11.7	12.0	4.21	55	34.2	6.03	10.0	-	43	9.8	8.5	3.4	9.5	38.7	2.9	14.0	16.1	10.6	17.4	6.18
NOx-column	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
Total: Feed to NOx-absorber	973	14765	2604	204204	932	446	8029	-	51135	-	1730	9419	4849	-	-	1687	9458	2835	23144	1995

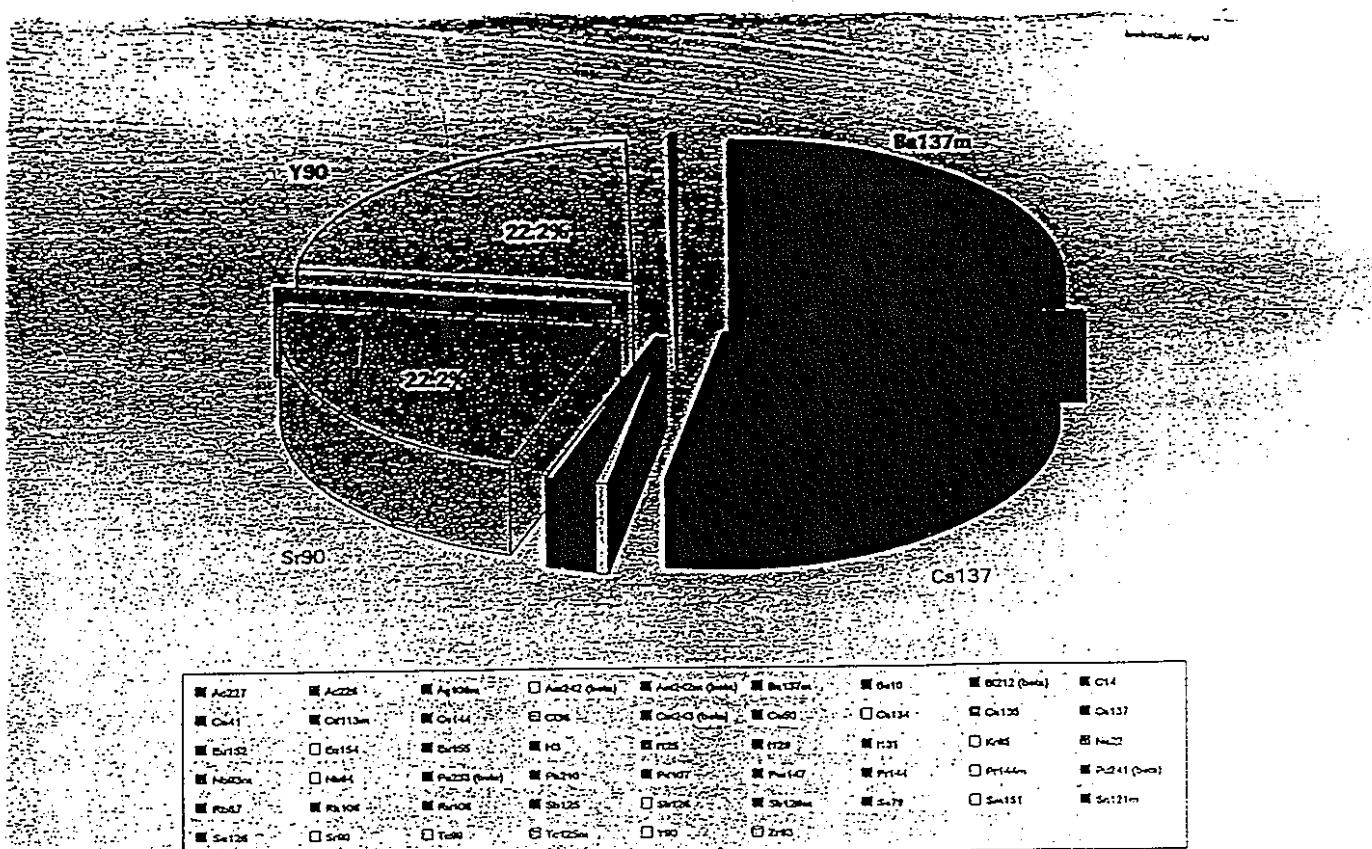


Fig. 8-1: Radioactivity (beta, beta/gamma, gamma) of individual isotopes of the WAK-HAWC referred to 1997

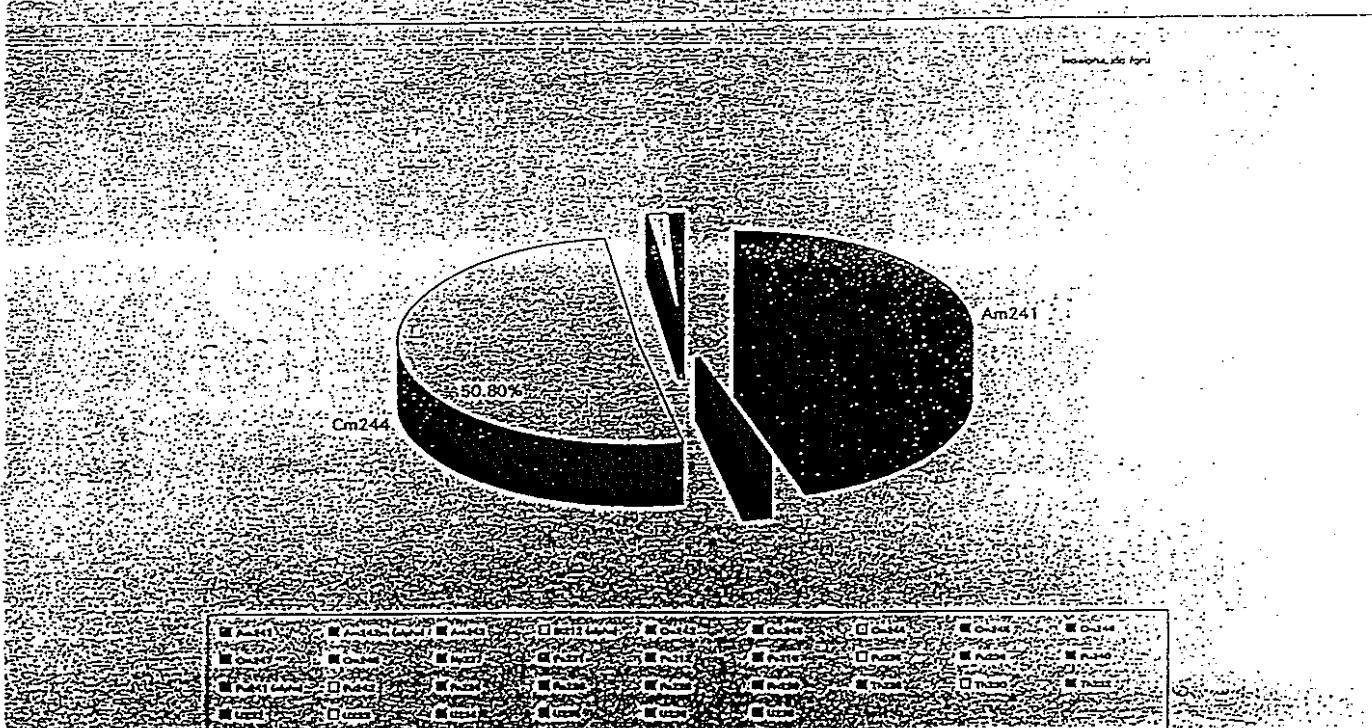


Fig. 8-2: Radioactivity (alpha) of individual isotopes of the WAK-HAWC referred to 1997

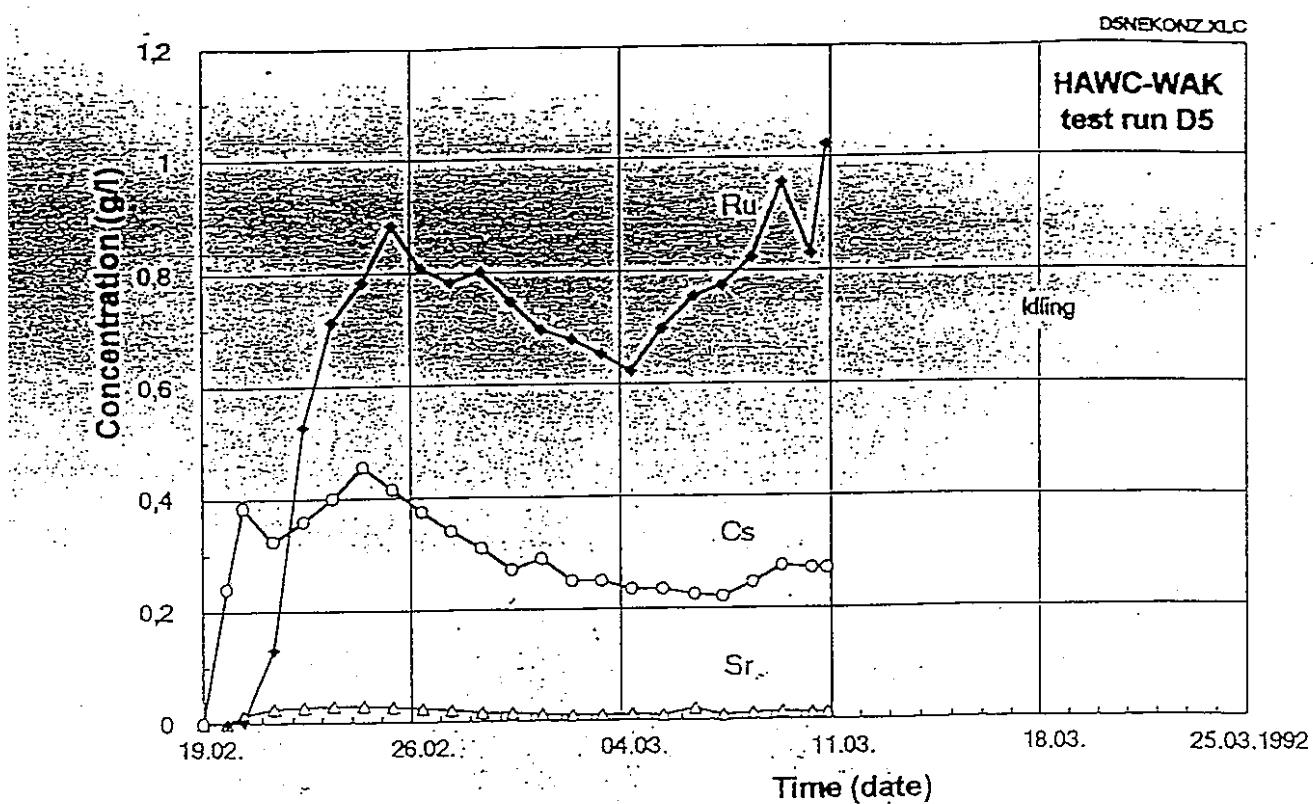


Fig. 8-5 : Concentrations of Ru, Cs and Sr in the dust scrubber wash solution.  
Test run D5

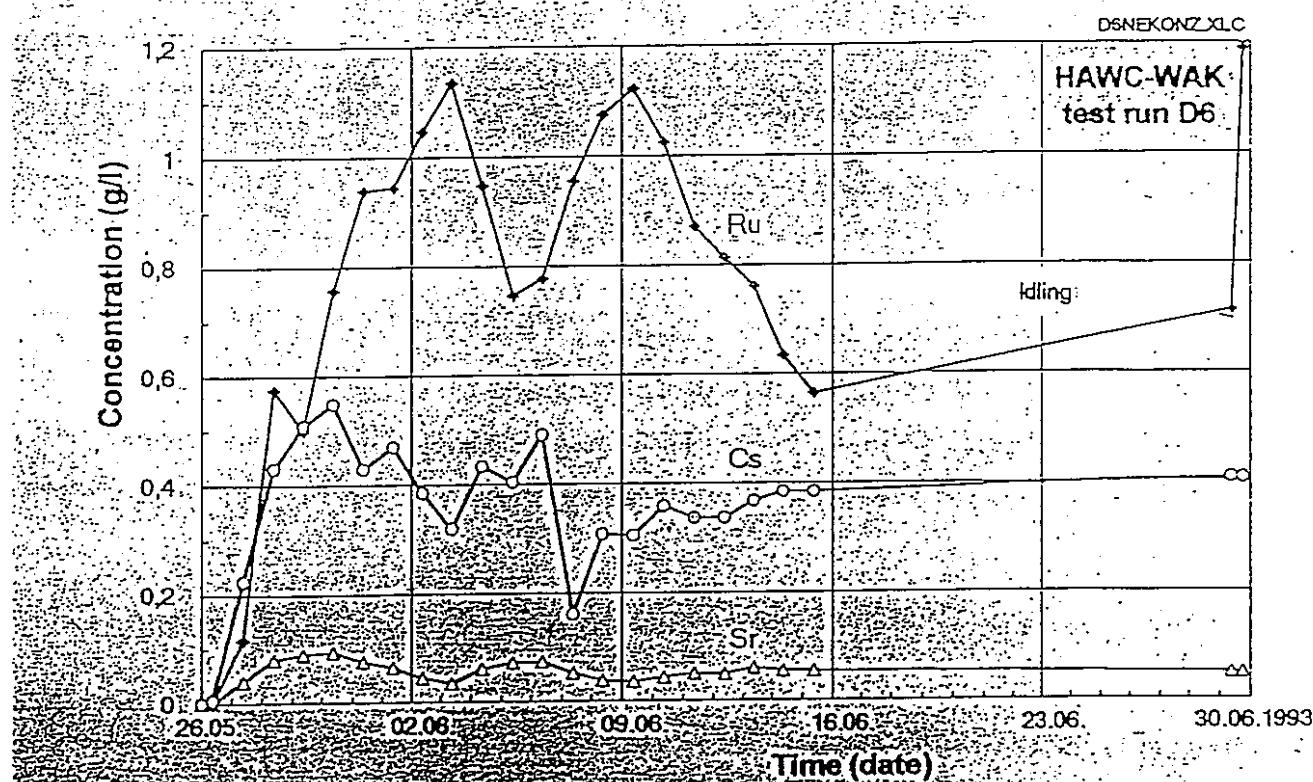


Fig. 8-6 : Concentrations of Ru, Cs and Sr in the dust scrubber wash solution.  
Test run D6

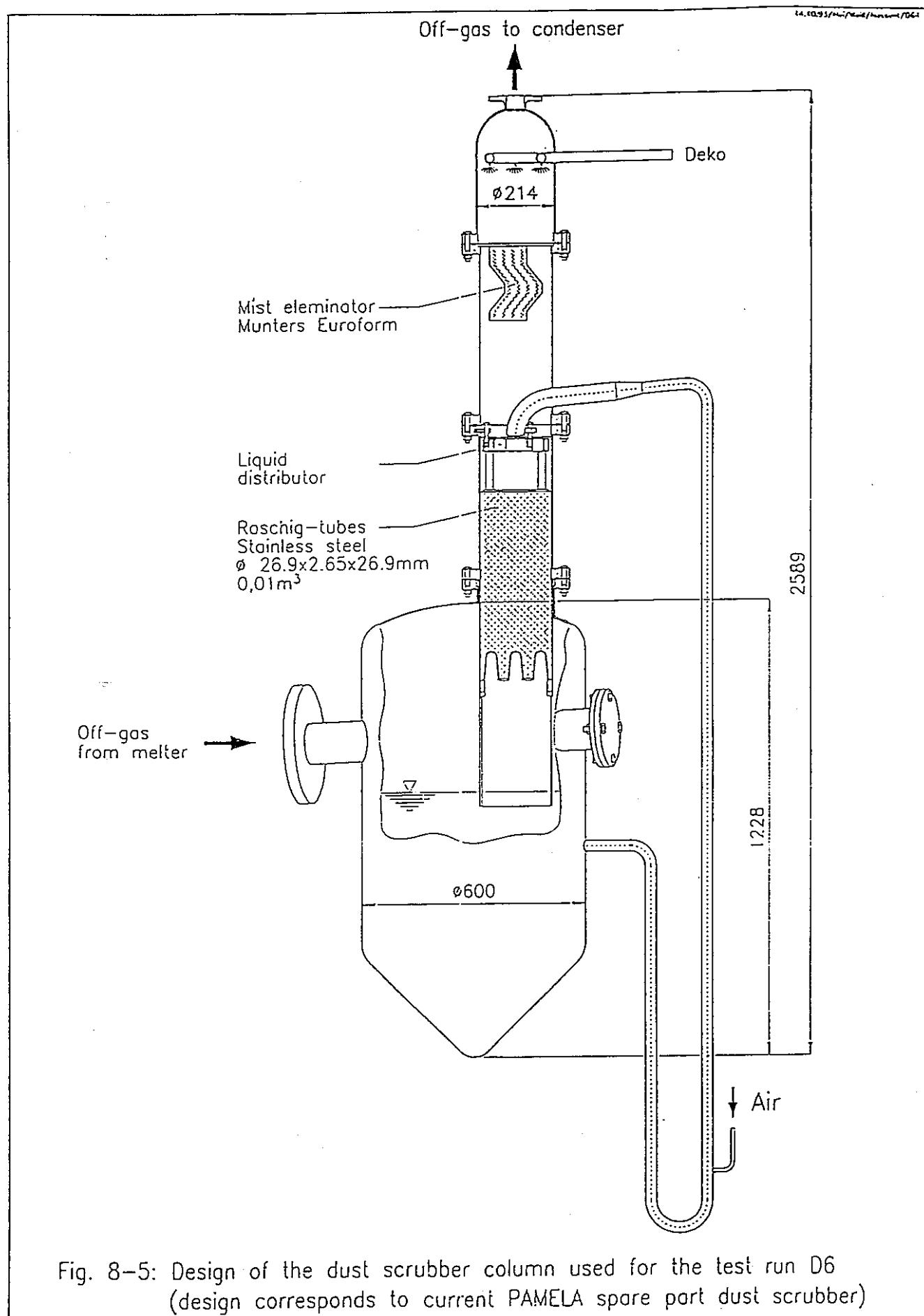


Fig. 8-5: Design of the dust scrubber column used for the test run D6  
(design corresponds to current PAMELA spare part dust scrubber)

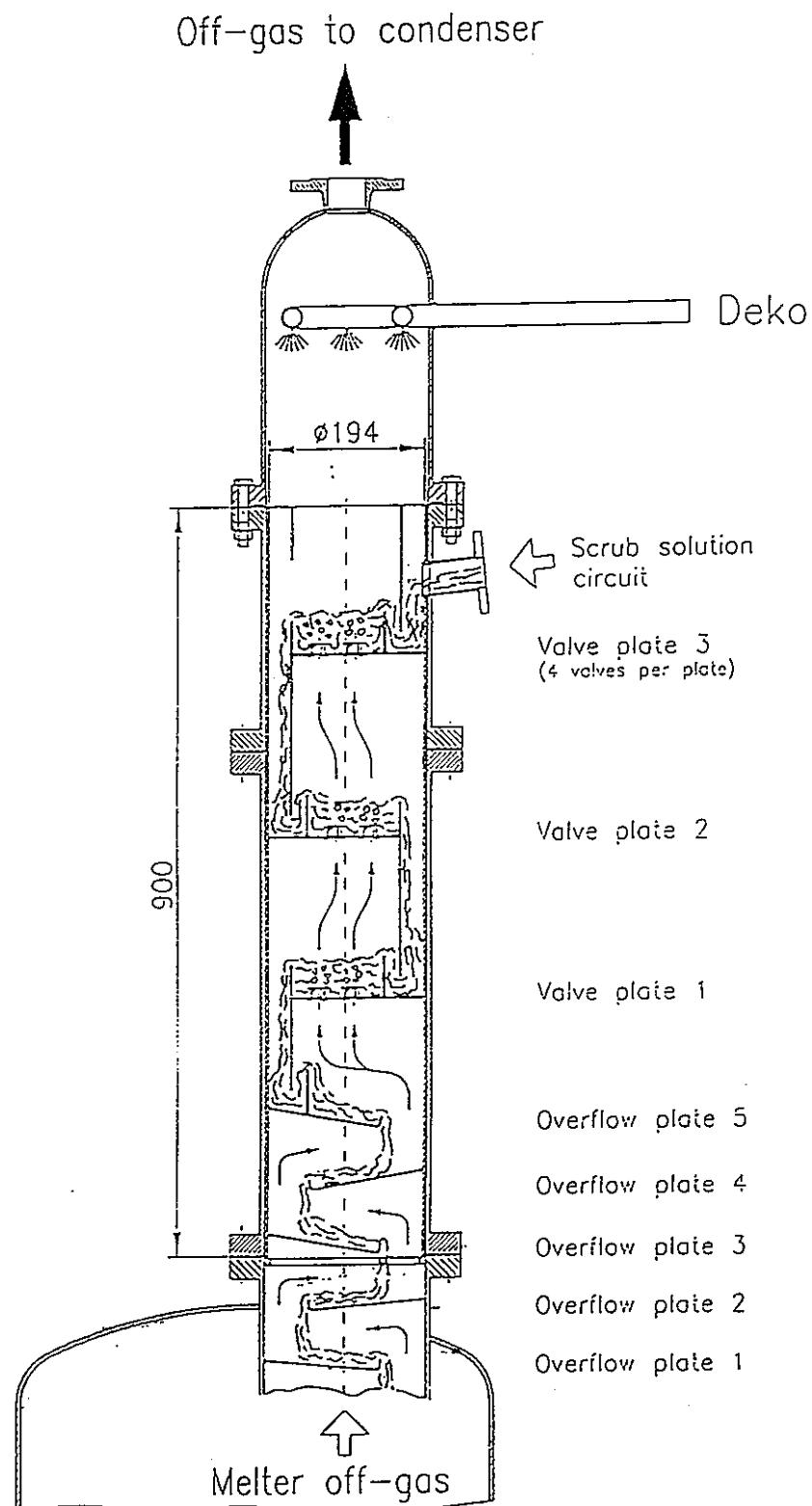


Fig. 8-3: Dust scrubber column design used in the test runs D2, D4 and D5

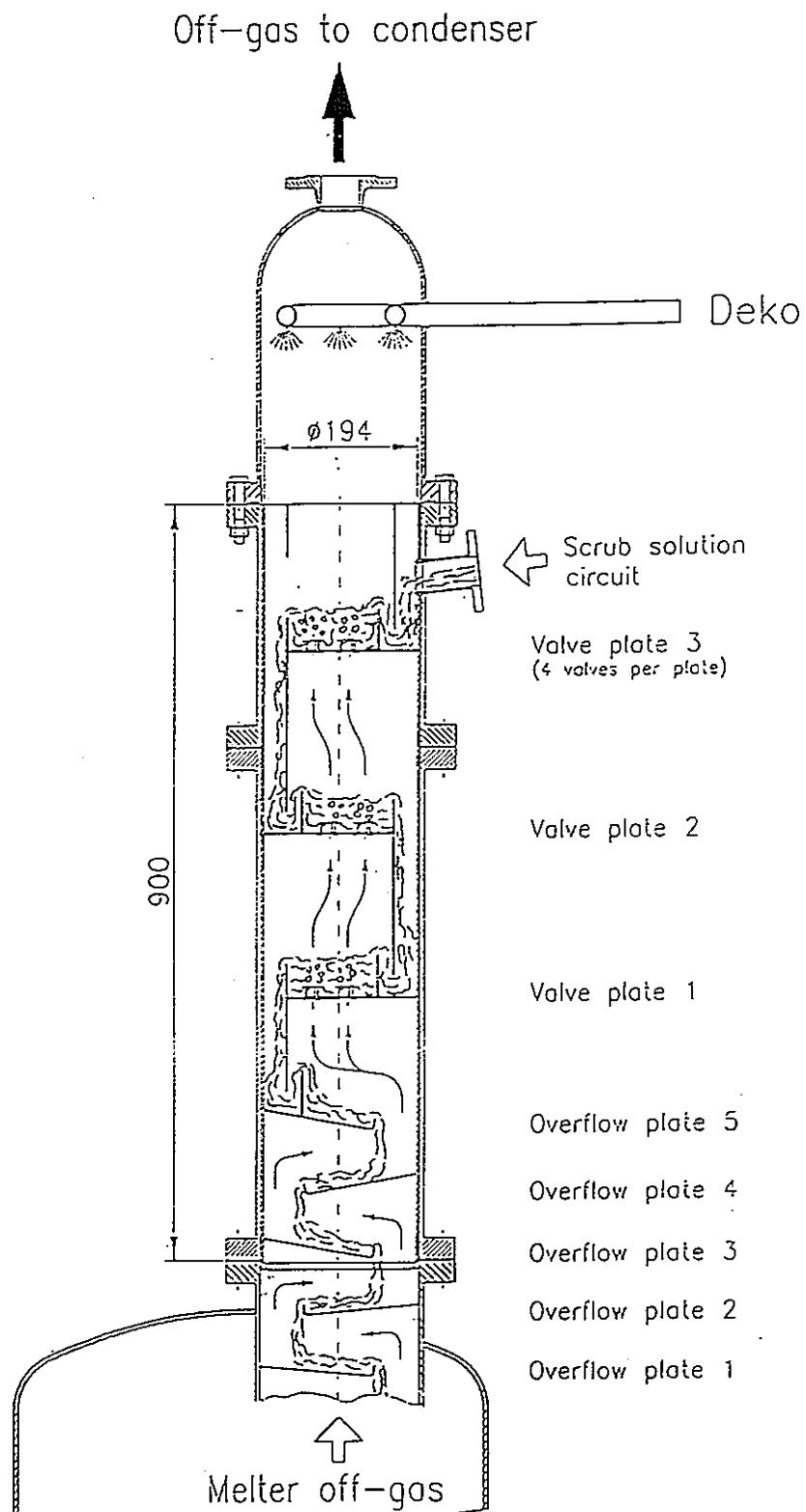


Fig. 8-3: Dust scrubber column design used in the test runs D2, D4 and D5

**Overview about the quantities of the elements Cs, Sr and Ru removed by the melter, dust scrubber, condenser, jet scrubber and NO<sub>x</sub>-column during the test run D6. Compilation of the resulting DF numbers.**

	Quantity of element (g)			DF		
	Cs	Sr	Ru	Cs	Sr	Ru
<b><u>Fed to melter</u></b>						
via HAWC-simulate	49 937	12 564	35 497	-	-	-
via HAWC-simulate and recycled dust scrubber solution	50 599	12 664	36 931	-	-	-
<b><u>Removed by</u></b>						
Melter	49 198	12 432	35 065	36.1	96	19.8
Dust scrubber	714	106.4	1588	2.04	5.16	6.72
Condenser	261	10	140	1.61	1.65	2.01
Jet scrubber	395	15	125	14.0	16.06	10.63
NO <sub>x</sub> -column	30	1	13	-	-	-

**Overview about the quantities of the elements Cs, Sr and Ru removed by the melter, dust scrubber, condenser, jet scrubber and NO<sub>x</sub>-column during the test run D6. Compilation of the resulting DF numbers.**

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via HAWC-simulate	49 937	12 564	35 497	-	-	-
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Jet scrubber	395	15	125	14.0	16.06	10.63
NO <sub>x</sub> -column	30	1	13	-	-	-

Thema: DF - data

Datum: Dec. 7, 1993

Seite

## Summary

- Melter DF's for Cs, Sr, Ru depend on "cold cap" conditions on top of the glass pool, and on bubbling
- DF-data variation found

	Melter	Dust scrubber
Cs	8.8 - 59	1.9 - 3
Sr	41.3 - 228	2.7 - 14

- DF data for condenser, jet scrubber may be partly influenced by the concentration of Cs, Sr, Ru in the off gas stream
- Different types of dust scrubber columns used showed similar DF's for Cs, Sr, Ru.
- IDF data : feed to entrance  $\text{NO}_x$ -absorber

Test run	D2	D4	D5	D6
Cs	970	446	1730	1687
Sr	14765	8029	9419	9458

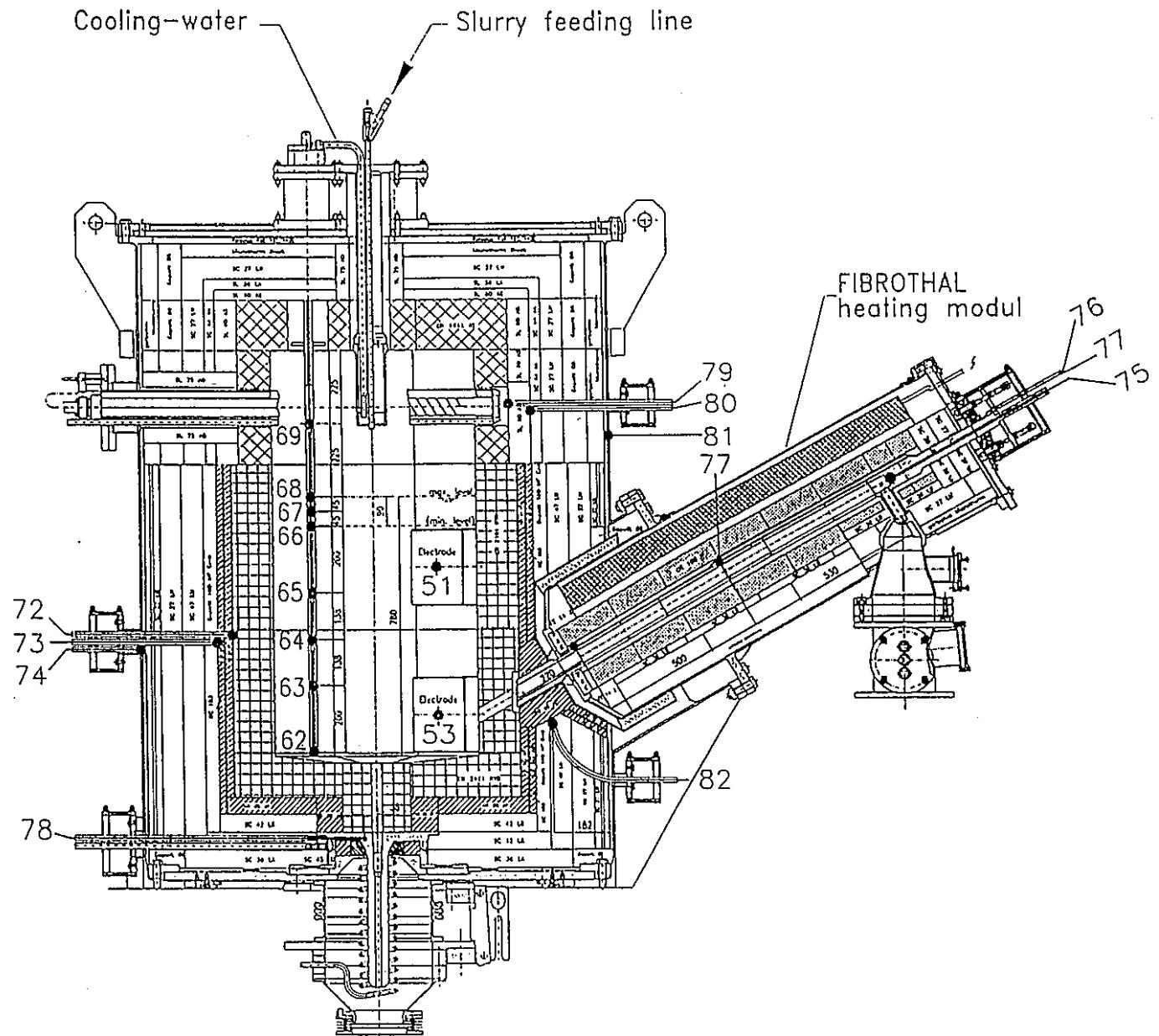
Acceptable ranges for the PAMELA approach to keep the overall feed to stack DF concept of 10<sup>8</sup> as a minimum

- The second recycling circuit (recycling of evaporator concentrate from secondary liquid waste concentration) must be used to keep the amount of conditioned MLW acceptable

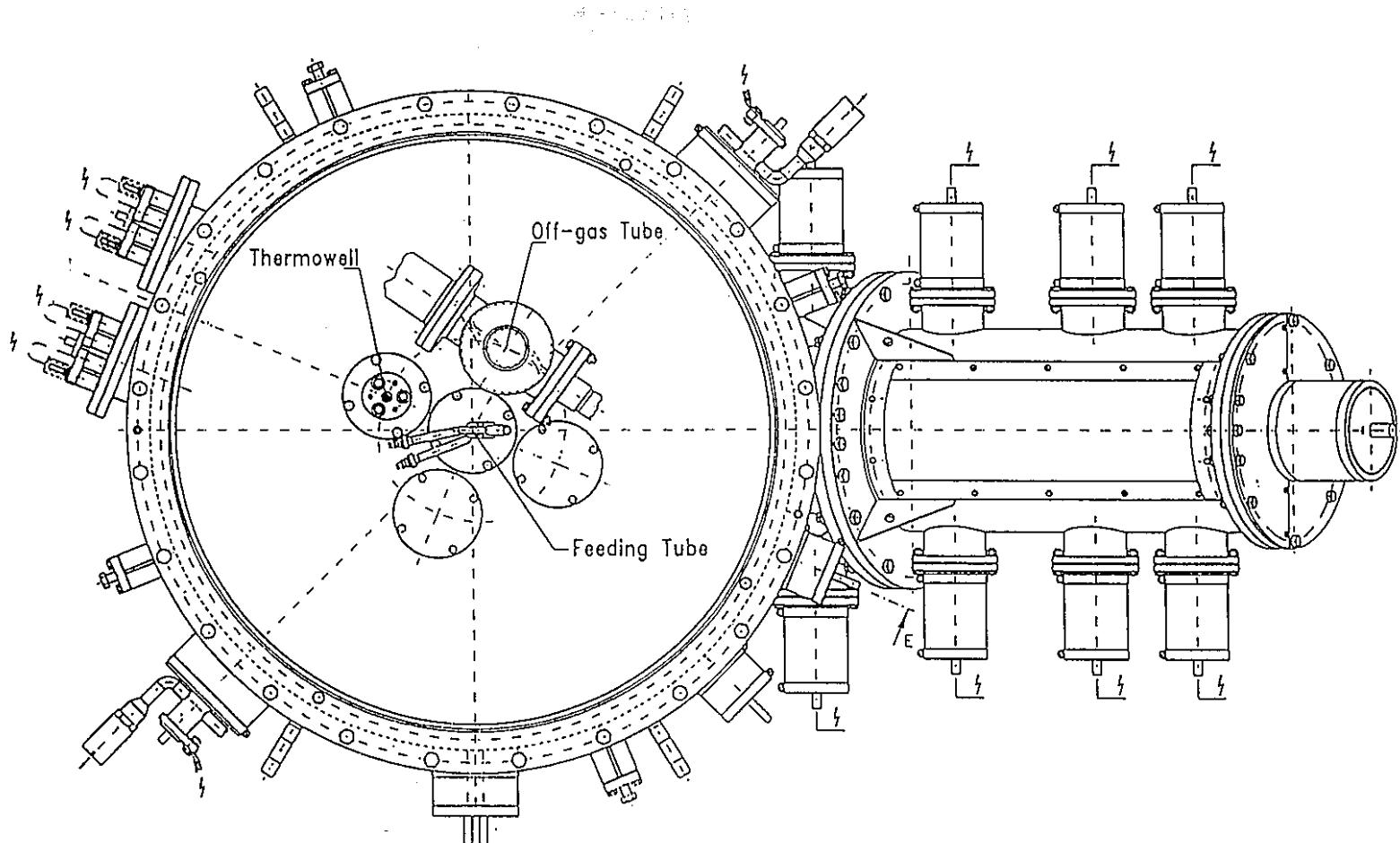
3.6 Melt level detection system recently tested in the K-6' and K-W3 melter and New overflow heating technique applied in the ESM melter

# Melter features

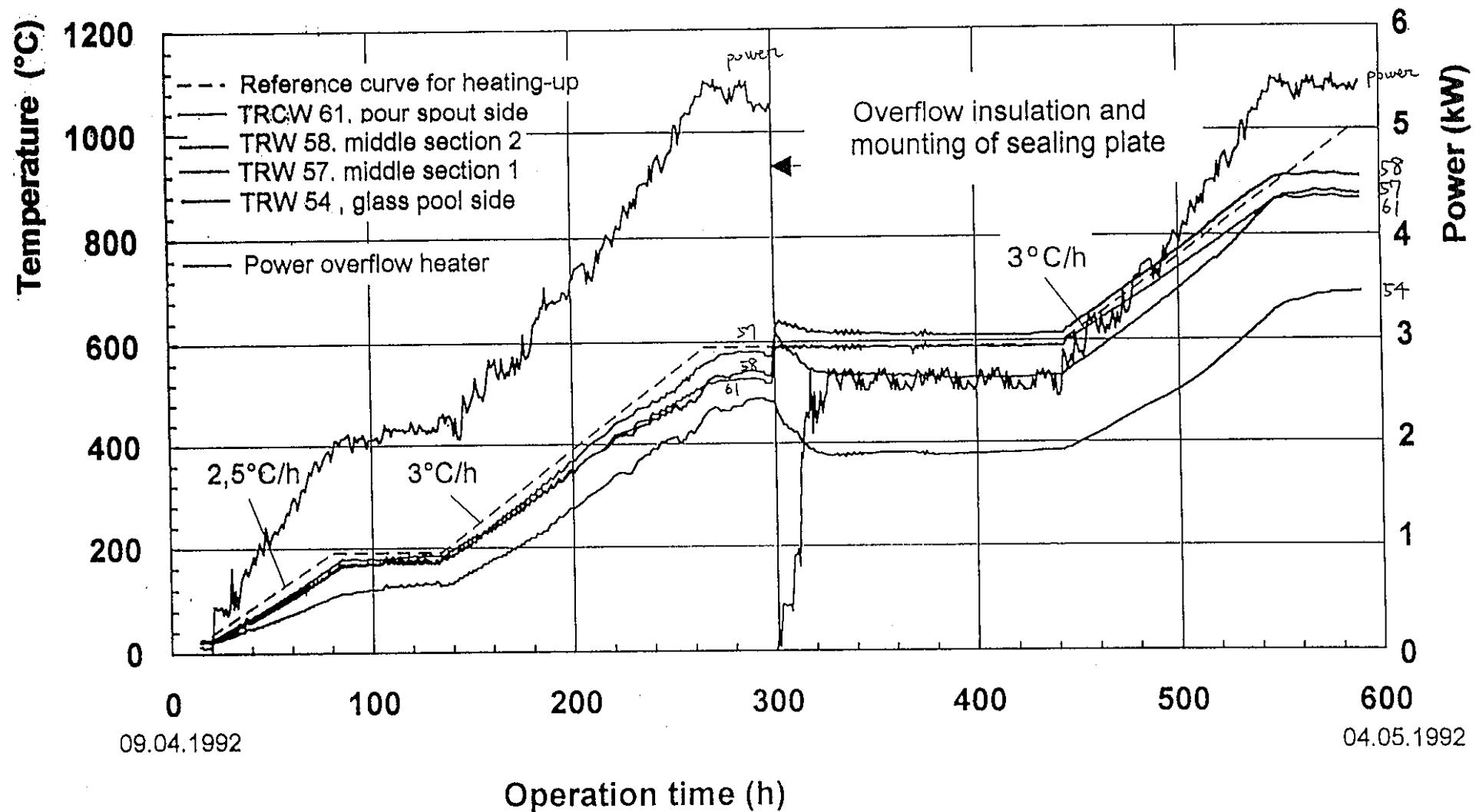
- ☞ 10 th scale of Hanford Waste Vitrification Plant melter
- ☞ flat bottomed melt tank, 550 kg glass capacity
- ☞ two pairs of uncooled Inconel electrodes
- ☞ two SiC melter plenum heaters
- ☞ designed throughput of 20 l/h (NCAW)
- ☞ routine glass pouring via overflow with underpressure support



Longitudinal section of ESM with temperature monitoring points



Top section view of the ESM



Start-up heating procedure for ESM melter overflow

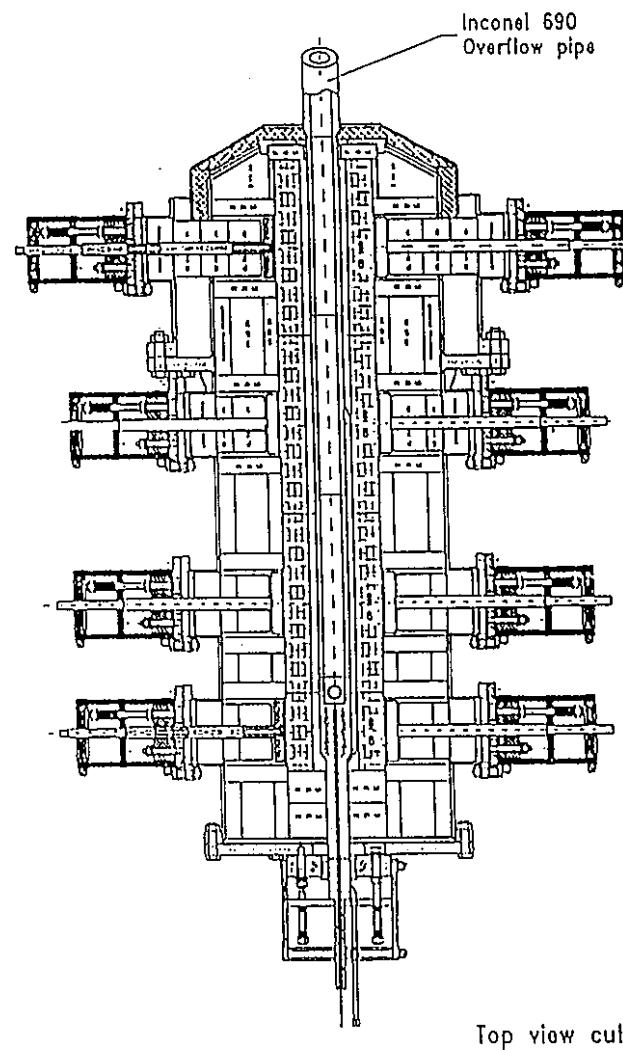
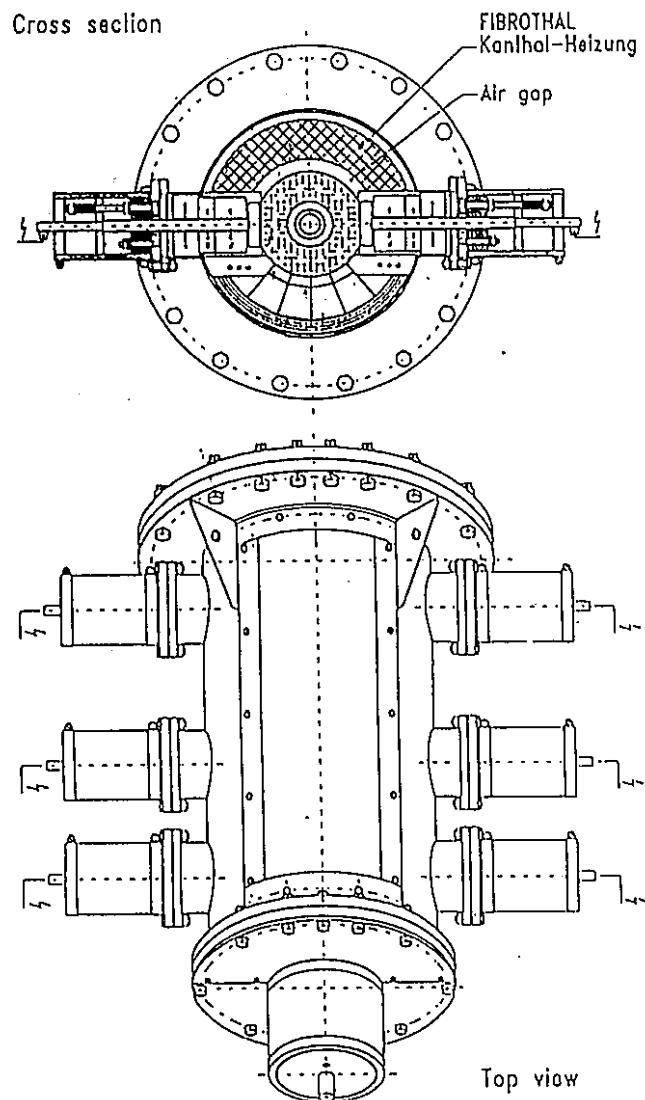
# New overflow heating technique applied in the ESM melter

**ESM**      Engineering Scale Melter

☞ designed, constructed, installed and operated by  
KfK/INE under contract and in cooperation with

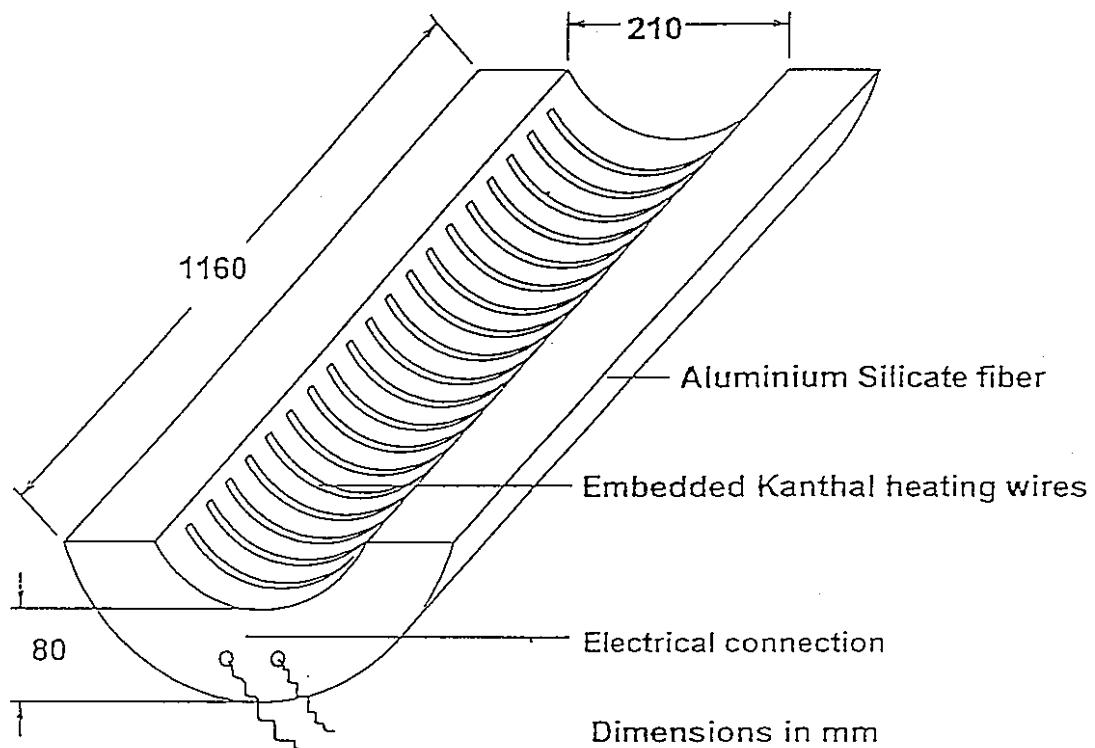
**PNL**      Pacific NL

☞ in 1991 - 1992



Glass overflow system of ESM and heating technique applied

## Used overflow *FIBROTHAL* heating modul



- heating surface       $0.3 \text{ m}^2$
- power loading         $6.0 \text{ kW / 220 VAC}$
- thermal conductivity  $0.242 \text{ W/mK (at } 1150^\circ\text{C)}$

### Advantages of *FIBROTHAL* heating modul

- even temperature distribution
- temperature flexibility
- easy replacement of modul
- low furnace weight (specific weight  $200\text{g/dm}^3$ )
- low price

3.7 Characterization and quality assurance for the HAWC-WAK  
waste glass if produced in the PAMELA and disposed in Germany

## 9. Results of the process control referred to glass product composition

### 9.0 Description of the problem

The composition of the waste glass poured from melter primarily results from a number of key parameters governing HAWC-processing. Those parameters include

- the composition of the waste solution
- the composition of the glass frit
- the waste glass loading
- the actual mass ratio of HAWC to frit under which the melter is fed and the variation of this ratio with time

The composition of the HAWC and that of the glass frit determines - together with the waste loading number - the target value for each individual constituent of the glass product. However, a number of processing parameters are involved. They can more or less influence the concentration in the final glass product. Such parameters include the

- Variation of the HAWC-composition
- Variation of the glass frit composition
- Inaccuracies caused by the HAWC- and glass frit feeding system, regarding especially the ratio between HAWC- and glass frit
- Inhomogenities of the glass melt due to poor homogenization of the melt pool or due to insufficient residence time
- Phase separation in the melt pool (noble metals for example)

These and a number of other influences, as for example excessive loss of waste elements into the off-gas line, can also cause more or less differences in the composition of the final glass product. A band width of scatter had therefore been established which must be tolerated. The actual glass product composition must be within this band. Usually concerns about glass product quality do not arise due to these allowed variations which can be within 1 to 2 wt% as to give an order to magnitude only. In the course of the HAWC-WAK technology program it had been tested what band width of scatter result with reasonable effort in comparison to the "target glass product composition".

#### 9.1 Reference composition of the inactive glass product

The target values of the glass product for each individual constituent of the glass are listed in Table 9-I. The first column shows the target numbers in case of vitrification of the active

HAWC. They are based on both the reference composition of the HAWC and the nominal waste glass loading of 16 wt%. The second column shows the corresponding numbers for the inactive HAWC-simulate and a waste glass loading of 14 wt %. The waste glass loading is lower compared to the active case due to the fact that the heavy actinides in the actual HAWC are not present in the simulate. The numbers of the second column are the targets by which the inactively produced glass product must be compared. The numbers refer to the nominal composition of the HAWC-simulate used for the D6 test run, and to the glass frit type GG WAK 1.

## 9.2 Actual composition of the produced glass compared to the target data

### 9.2.1 Selected guide elements

For comparison of the technically produced waste glass with the target composition, seven guide elements have been selected: Si, Cs, Sr, Nd, Na, Fe and Zr. The selection criteria included that

- the elements are relatively easy measurable
- they are homogeneously distributed in the glass matrix i.e. they do not show phase separations, or are insoluble or segregate in the melt (like the noble metals)
- they are not too much influenced by other elements regarding their uniform distribution
- they represent typical constituents of the frit and waste stream fed to the melter (Si and Na for the glass frit; Cs, Sr and Nd for the most active part of the HAWC; Zr for representing undissolved material, Na for process chemicals and Fe for corrosion product).

In the final glass product sodium represents both, a part of the frit and the dominant constituent of the HAWC referred to the mass. It is the strategy of the advanced process technique to achieve and assure the correct composition within a given band width of scatter, and this without the need for glass sampling and analysing. The assurance of the composition within the established tolerance band should be based on the process control itself - and not on a direct product control. The results achieved in this respect in the inactive test run D6 are presented as an example that this is well possible.

### 9.2.2 XRF-results achieved for the glass product of the D6 run

The XRF-results initially gained for the glass product of the D6 run are plotted in the Figs. 9-1 to 9-3. Figure 9-1 shows the concentration of SiO<sub>2</sub> in the glass versus the mass of poured

Table 9-IV

Analytical data of SiO<sub>2</sub>, ZrO<sub>2</sub>, SrO and Cs<sub>2</sub>O measured by the analytical glass lab of Company Schott/Mainz. Date of analysis Nov. 4, 1993 under code FEA-2/Mec. Glass samples taken from glass pouring stream into canister No. 13 of the D6 test run

D6 test run

Pouring of canister No. 13, D6 run	Concentration (wt %)			
	SiO <sub>2</sub>	ZrO <sub>2</sub>	SrO	Cs <sub>2</sub> O
Target 1)	51.60	0.508	0.16	0.569
Pouring batch A				
Sample No. 1	51.84 ± 0.136*)	0.50	0.14	0.46
2	51.77 ± 0.097	0.49	0.14	0.46
3	51.91 ± 0.144	0.49	0.14	0.47
4	51.66 ± 0.093	0.49	0.14	0.46
5	51.58 ± 0.098	0.49	0.14	0.46
6	51.81 ± 0.137	0.51	0.14	0.46
7	51.65 ± 0.212	0.49	0.14	0.47
8	51.68 ± 0.115	0.47	0.14	0.48
9	51.75 ± 0.232	0.48	0.15	0.47
10	51.81 ± 0.281	0.49	0.14	0.47
11	51.76 ± 0.241	0.49	0.15	0.48
12	51.90 ± 0.137	0.50	0.14	0.47
13	51.90 ± 0.047	0.49	0.14	0.47
14	51.73 ± 0.064	0.51	0.15	0.47
15	51.75 ± 0.165	0.50	0.14	0.48
16	51.81 ± 0.127	0.50	0.15	0.47
Pouring batch B				
Sample No. 1	51.85 ± 0.099	0.49	0.14	0.47
2	51.85 ± 0.098	0.49	0.14	0.46
3	51.89 ± 0.193	0.51	0.14	0.47
4	51.57 ± 0.076	0.50	0.14	0.46
5	51.72 ± 0.095	0.51	0.14	0.46
6	51.68 ± 0.095	0.51	0.14	0.47
7	51.64 ± 0.199	0.49	0.14	0.46
8	51.66 ± 0.158	0.48	0.15	0.48
9	51.54 ± 0.135	0.49	0.15	0.47
10	51.73 ± 0.240	0.49	0.14	0.47
11	51.84 ± 0.075	0.49	0.15	0.47
12	51.76 ± 0.089	0.50	0.14	0.47
13	51.75 ± 0.039	0.52	0.14	0.47
14	51.73 ± 0.110	0.52	0.15	0.48
15	51.83 ± 0.220	0.50	0.15	0.47
16	51.84 ± 0.203	0.50	0.15	0.48

1) Referred to the technically produced simulated HAWC-glass

\*) standard deviation ± s

Table 3-IVa: Composition of the radioactive HAWC reference solution, compared with the simulate used for the D6 campaign

Element	Filtrate (dissolved)			Solids (not dissolved)			Element concentration g/l	Oxide residue g/l	Oxide residue Wt %	Total			Simulate, D6 (total)		
	Element concentration g/l	Oxide Oxide residue, g/l	Oxide residue, Wt %	Element concentration g/l	Oxide residue g/l	Oxide residue Wt %				Element concentration g/l	Oxide residue g/l	Oxide residue Wt %	Element concentration g/l	Oxide residue g/l	Oxide residue Wt %
34 Se	0,046	SeO <sub>2</sub>	0,065	0,080	0,016	0,022	0,237	0,062	0,087	0,096	0,062	0,087	0,106		
37 Rb	0,268	Rb <sub>2</sub> O	0,293	0,361	0,019	0,020	0,218	0,286	0,313	0,347	1)	1)	1)		
38 Sr	0,788	SrO	0,932	1,150	0,003	0,003	0,033	0,791	0,935	1,035	0,791	0,935	1,140		
39 Y	0,570	Y <sub>2</sub> O <sub>3</sub>	0,724	0,093	0,005	0,007	0,073	0,575	0,731	0,808	0,575	0,731	0,894		
40 Zr	0,054	ZrO <sub>2</sub>	0,073	0,090	2,141	2,093	31,011	2,196	2,966	3,281	2,196	2,966	3,628		
42 Mo	1,535	MoO <sub>3</sub>	2,303	2,041	1,840	2,761	29,602	3,375	5,064	5,602	3,375	5,064	6,194		
43 Tc	0,901	TcO <sub>3</sub>	1,339	1,651	0,011	0,017	0,180	0,913	1,355	1,499	2)	2)	2)		
44 Ru	2,120	RuO <sub>2</sub>	2,791	3,443	0,112	0,147	1,579	2,232	2,939	3,251	2,232	2,939	3,595		
45 Rh	0,560	Rh <sub>2</sub> O <sub>3</sub>	0,690	0,851	0,019	0,024	0,256	0,579	0,714	0,790	-	-	-		
46 Pd	0,916	PdO	1,054	1,300	0,356	0,409	4,309	1,272	1,461	1,619	1,272	1,464	1,791		
47 Ag	0,002	Ag <sub>2</sub> O	0,002	0,003	0,111	0,119	1,279	0,113	0,121	0,134	-	-	-		
48 Cd	0,078	CdO	0,090	0,111	0,004	0,005	0,055	0,083	0,095	0,105	-	-	-		
50 Sn	0,046	SnO <sub>2</sub>	0,059	0,073	0,012	0,015	0,157	0,058	0,074	0,082	0,058	0,074	0,091		
51 Sb	0,003	Sb <sub>2</sub> O <sub>3</sub>	0,004	0,004	0,008	0,009	0,100	0,011	0,013	0,014	-	-	-		
52 Te	0,400	TeO <sub>2</sub>	0,500	0,616	0,123	0,154	1,647	0,523	0,653	0,723	0,523	0,653	0,799		
55 Cs	2,762	Cs <sub>2</sub> O	2,928	3,612	0,375	0,390	4,265	3,137	3,326	3,679	3,137	3,326	4,067		
56 Ba	2,231	BaO	2,491	3,073	0,096	0,107	1,150	2,327	2,598	2,875	2,327	2,598	3,178		
57 La	2,027	La <sub>2</sub> O <sub>3</sub>	2,378	2,934	0,023	0,027	0,286	2,050	2,405	2,660	2,050	2,405	2,942		
58 Ce	2,923	CeO <sub>2</sub>	3,591	4,429	0,008	0,108	1,156	3,011	3,699	4,092	3,011	3,699	4,524		
59 Pr	1,417	Pr <sub>2</sub> O <sub>3</sub>	1,658	2,045	-	-	-	1,417	1,658	1,834	1,417	1,658	2,028		
60 Nd	5,040	Nd <sub>2</sub> O <sub>3</sub>	5,878	7,252	0,019	0,022	0,236	5,059	5,900	6,528	6,313	7,362	9,005		
61 Pm	0,004	Pm <sub>2</sub> O <sub>3</sub>	0,004	0,005	-	-	-	0,004	0,004	0,005	3)	3)	3)		
62 Sm	1,117	Sm <sub>2</sub> O <sub>3</sub>	1,295	1,597	0,004	0,005	0,054	1,121	1,300	1,438	3)	3)	3)		
63 Eu	0,129	Eu <sub>2</sub> O <sub>3</sub>	0,149	0,183	-	-	-	0,129	0,149	0,164	3)	3)	3)		
64 Gd	0,383	Gd <sub>2</sub> O <sub>3</sub>	0,442	0,545	0,002	0,002	0,027	0,385	0,444	0,491	0,385	0,444	0,543		
92 U	5,705	U <sub>3</sub> O <sub>8</sub>	6,728	8,299	0,026	0,030	0,321	5,731	6,758	7,476	-	-	-		
93 Np	0,455	Np <sub>2</sub> O <sub>3</sub>	0,501	0,618	0,004	0,005	0,052	0,459	0,506	0,559	-	-	-		
94 Pu	0,146	PuO <sub>2</sub>	0,165	0,204	0,038	0,043	0,459	0,183	0,208	0,230	-	-	-		
95 Am	0,424	Am <sub>2</sub> O <sub>3</sub>	0,467	0,576	0,001	0,009	0,125	0,468	0,518	-	-	-	-		
96 Cm	0,017	Cm <sub>2</sub> O <sub>3</sub>	0,019	0,023	-	-	0,017	0,019	0,021	-	-	-	-		
24 Cr	1,728	Cr <sub>2</sub> O <sub>3</sub>	2,525	3,115	0,132	0,192	2,063	1,860	2,718	3,007	1,860	2,718	3,324		
25 Mn	0,211	MnO <sub>2</sub>	0,334	0,412	0,004	0,007	0,075	0,215	0,341	0,377	1,120	1,789	2,100		
26 Fe	6,869	Fe <sub>2</sub> O <sub>3</sub>	9,821	12,115	0,189	0,270	2,890	7,058	10,091	11,163	7,058	10,091	12,343		
28 Ni	1,244	NiO	1,584	1,953	0,040	0,051	0,547	1,204	1,635	1,808	1,284	1,635	2,000		
29 Cu	-	CuO	-	-	0,015	0,018	0,197	0,015	0,018	0,020	0,015	0,018	0,022		
30 Zn	-	ZnO	-	-	0,009	0,011	0,117	0,009	0,011	0,012	0,009	0,011	0,013		
82 Pb	-	PbO	-	-	0,003	0,003	0,032	0,003	0,003	0,003	0,003	0,003	0,004		
11 Na	18,310	Na <sub>2</sub> O	24,602	30,447	-	-	-	18,310	24,602	27,305	18,310	24,602	30,109		
12 Mg	0,410	MgO	0,680	0,839	-	-	-	0,410	0,680	0,752	0,410	0,680	0,831		
13 Al	0,057	Al <sub>2</sub> O <sub>3</sub>	0,108	0,133	-	-	-	0,057	0,108	0,119	0,057	0,108	0,132		
19 K	0,217	K <sub>2</sub> O	0,261	0,323	-	-	-	0,217	0,261	0,289	0,503	0,605	0,740		
20 Ca	0,353	CaO	0,494	0,609	-	-	-	0,353	0,494	0,546	0,353	0,494	0,604		
F <sup>-</sup>	0,019	F <sup>-</sup>	0,019	0,024	-	-	-	0,019	0,019	0,021	0,019	0,019	0,023		
Cl <sup>-</sup>	0,014	Cl <sup>-</sup>	0,014	0,017	-	-	-	0,014	0,014	0,015	0,014	0,014	0,017		
P	0,406	P <sub>2</sub> O <sub>5</sub>	0,931	1,149	0,621	1,122	15,243	1,027	2,353	2,603	1,027	2,353	3,090		
Fiss.Prod.	Σ 26,322		31,731	39,144	5,300	7,275	77,992	31,710	39,006	43,153	-	-	-		
Aclnides	Σ 6,747		7,880	9,721	0,069	0,079	0,844	6,815	7,958	8,804	-	-	-		
Cor.Prod.	Σ 10,052		14,264	17,596	0,391	0,552	5,291	10,443	14,816	16,391	-	-	-		
Chemicals	Σ 19,786		27,188	33,540	0,621	1,422	15,243	20,406	28,610	31,652	-	-	-		
Total	Σ 62,907		81,063	100,00	6,468	9,328	100,00	69,376	90,391	100,00	-	81,757	100,017		
Free HNO <sub>3</sub>	4 M		-	-	-	-	-	-	-	-	4 M	-	-		

1) Rb portion replaced by K 2) Te portion replaced by Mn 3) Pm, Sm and Eu portions replaced by Nd

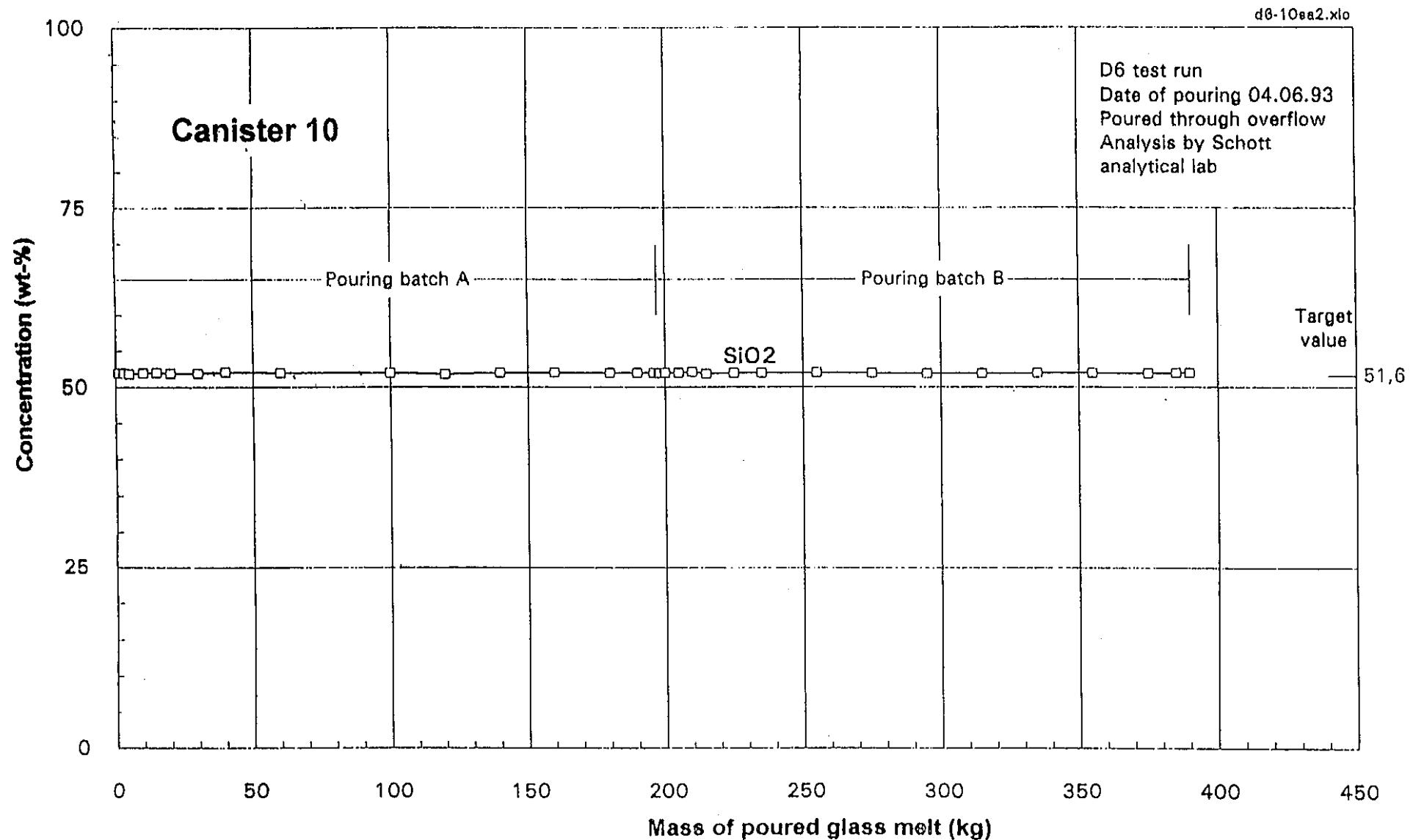


Fig. 9-4a: Concentration of SiO<sub>2</sub> in glass samples taken from overflow glass pouring stream while filling canister 10 of the D6 test run. Analysis data obtained from analytical lab of company Schott/Mainz.

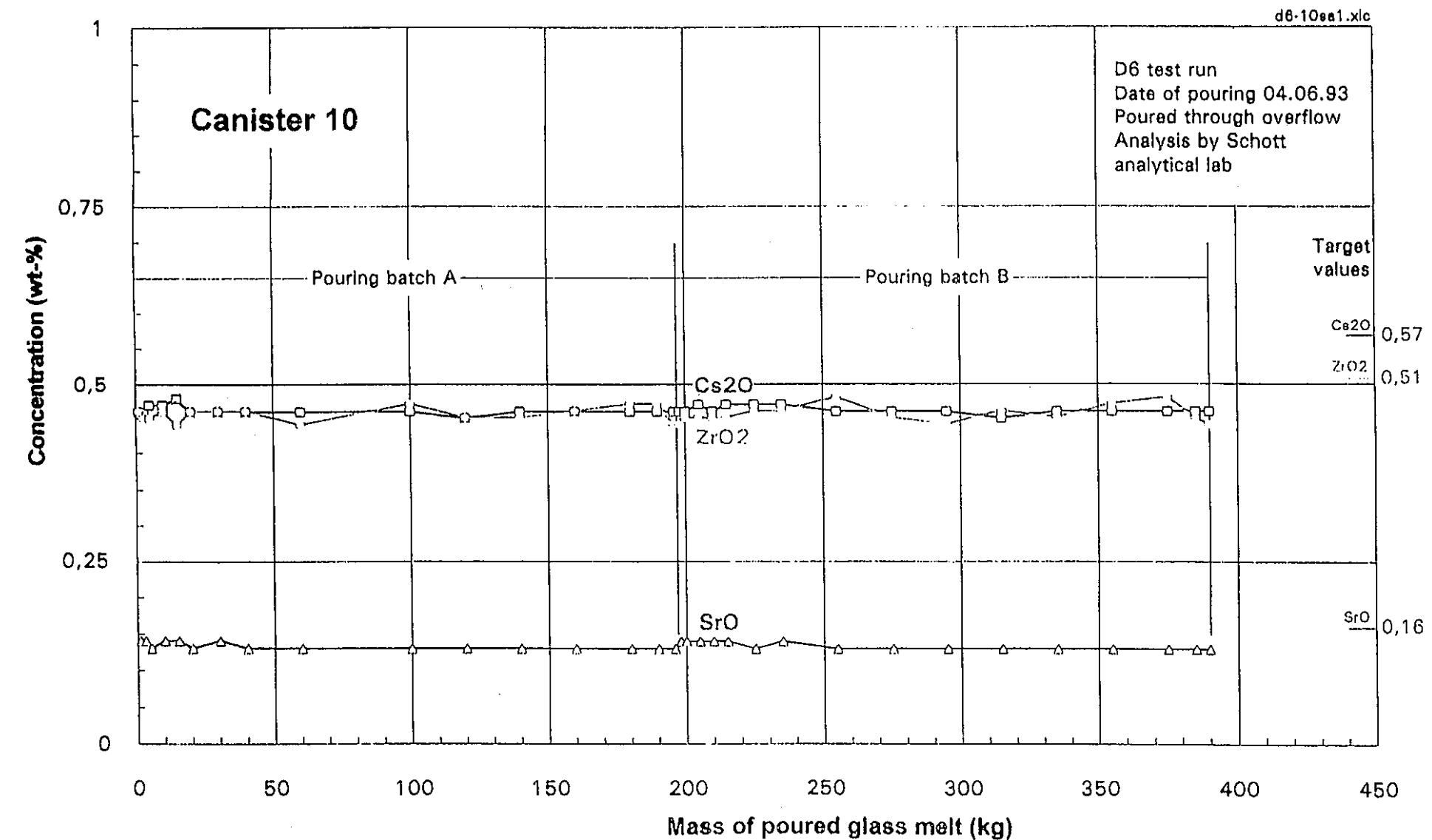


Fig. 9-4b: Concentration of ZrO<sub>2</sub>, Cs<sub>2</sub>O and SrO in glass samples taken from overflow glass pouring stream while filling canister 10 of the D6 test run. Analysis data obtained from analytical lab of company Schott/Mainz.

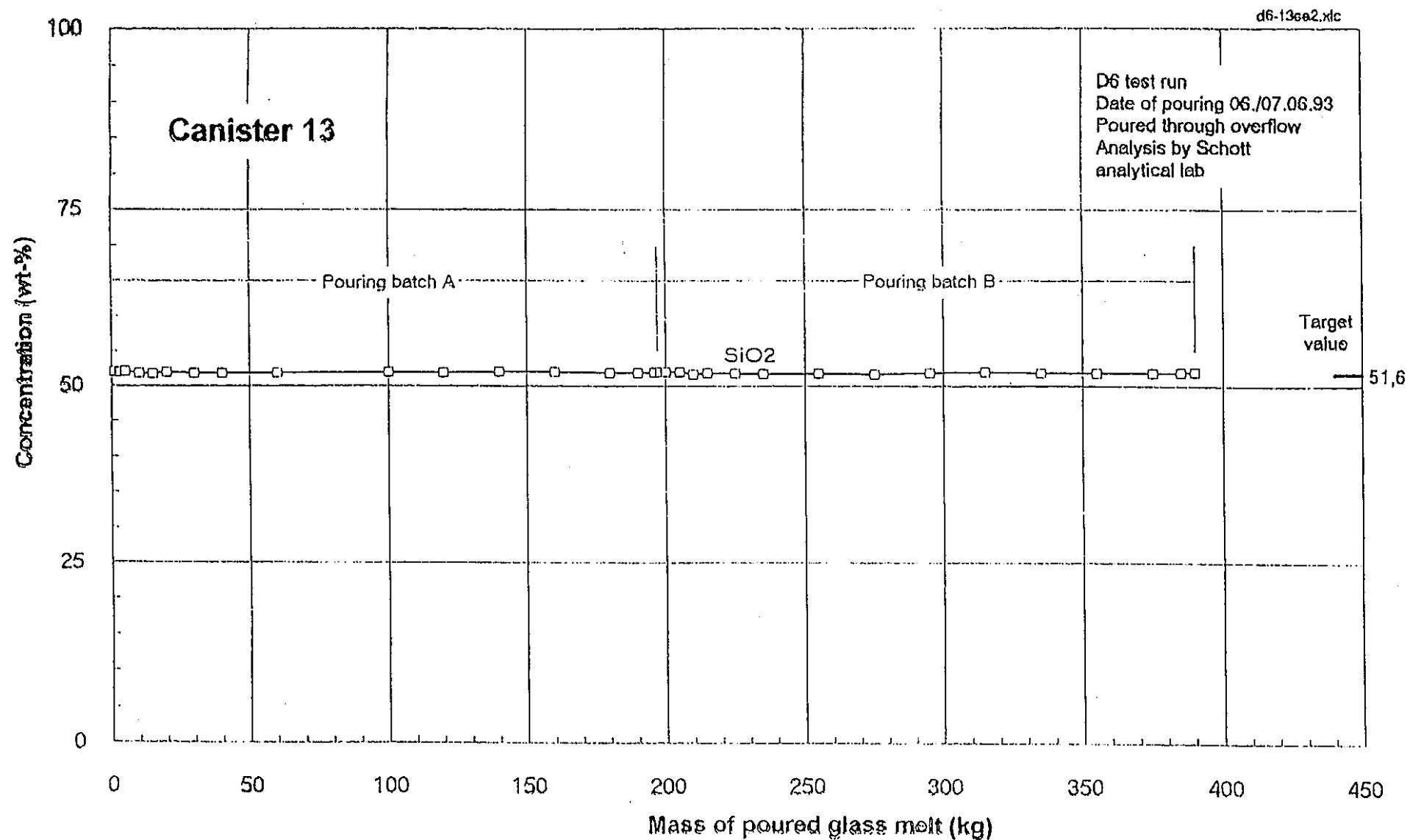


Fig. 9-5a: Concentration of SiO<sub>2</sub> in glass samples taken from overflow glass pouring stream while filling canister 13 of the D6 test run. Analysis data obtained from analytical lab of company Schott/Mainz.

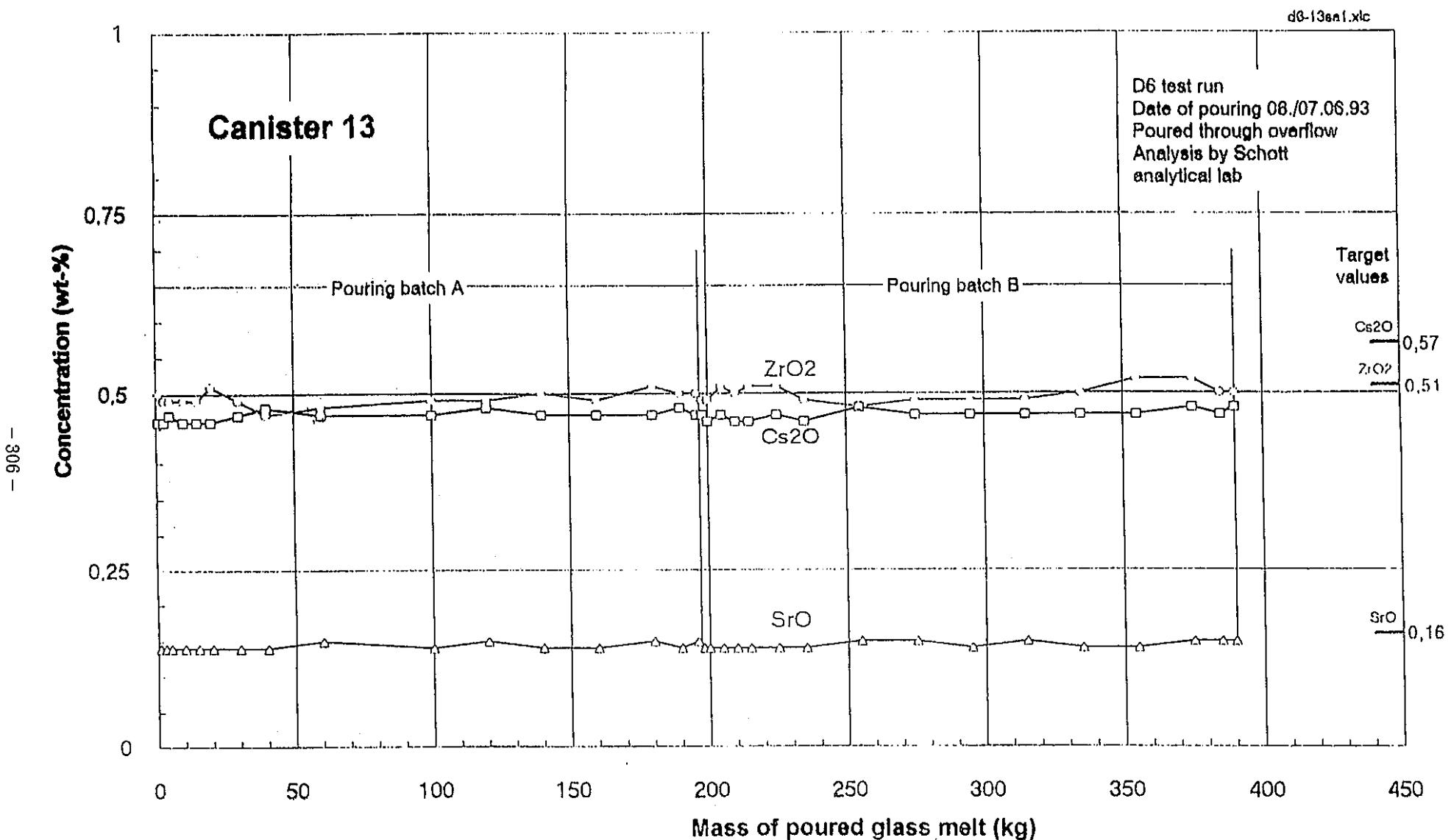


Fig. 9-5b: Concentration of ZrO<sub>2</sub>, Cs<sub>2</sub>O and SrO in glass samples taken from overflow glass pouring stream while filling canister 13 of the D6 test run. Analysis data obtained from analytical lab of company Schott/Mainz.

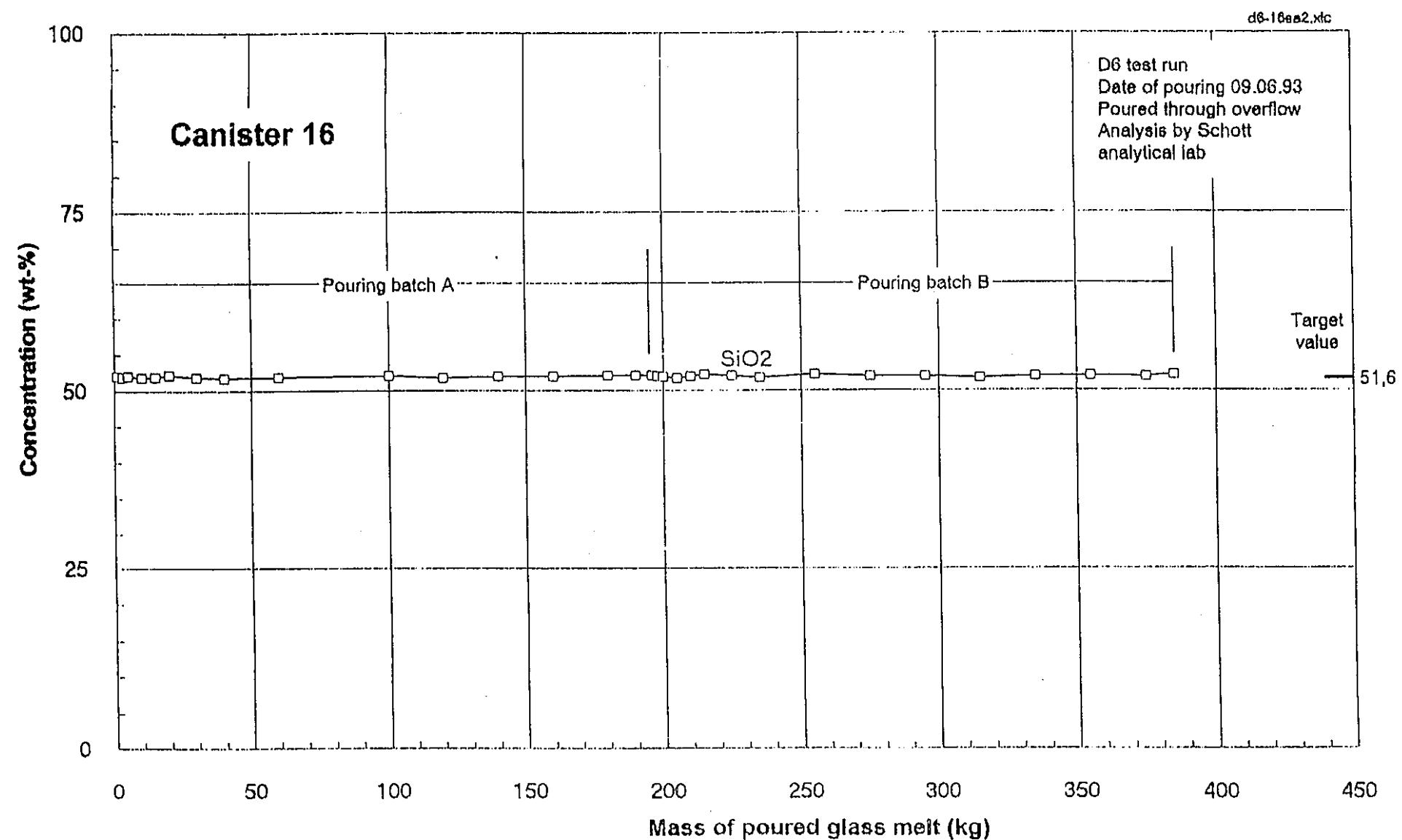


Fig. 9-6a: Concentration of SiO<sub>2</sub> in glass samples taken from overflow glass pouring stream while filling canister 16 of the D6 test run. Analysis data obtained from analytical lab of company Schott/Mainz.

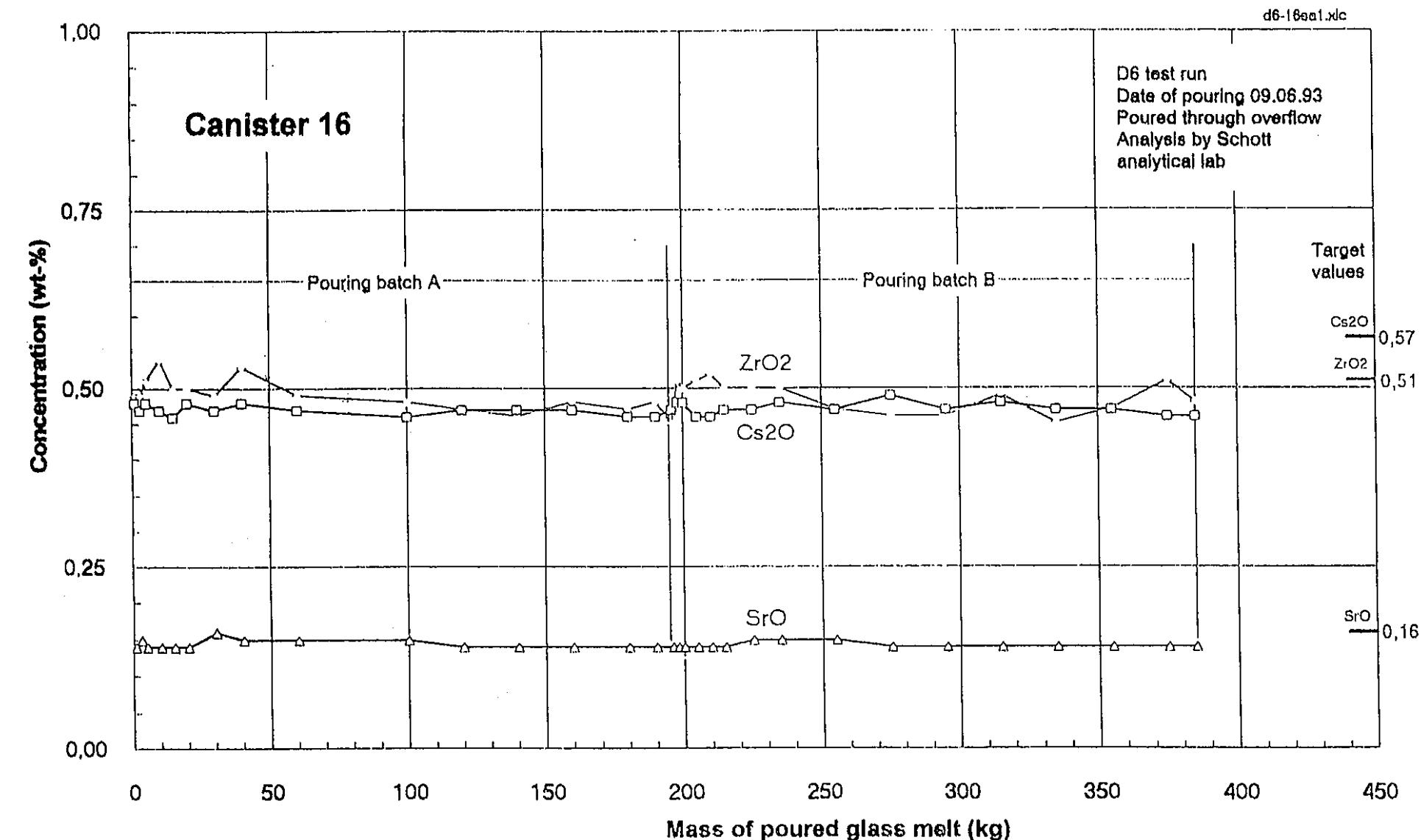


Fig. 9-6b: Concentration of ZrO<sub>2</sub>, Cs<sub>2</sub>O and SrO in glass samples taken from overflow glass pouring stream while filling canister 16 of the D6 test run. Analysis data obtained from analytical lab of company Schott/Mainz.

ENTWURF

TEAWK/Vo/933400

24.07.1993

~~HFA/1250/7A~~

HFA/1250/7A

W0788359



## *Specification*

*of the Vitrified Residue*

*of WAK High Level Waste*

GNS B TE 002/93

Revision: "00"

Date: 24.07.1993

Name

Signature

Author: Dr. W. Kunz / TEA

Verification: E. Ewest / TQ1

Release GNS: R. Weh / TE

This report has been written on behalf of  
Wiederaufarbeitungsanlage Karlsruhe BGmbH (WAK),  
Eggenstein-Leopoldshafen, Germany

## *Introduction*

Within a period of nearly 20 years the Karlsruhe Reprocessing Plant (Wiederaufarbeitungsanlage Karlsruhe - WAK), which was designed as a pilot plant for the development of the future commercial reprocessing, has reprocessed spent fuel by the PUREX process of several German nuclear reactor types.

Up to its shut down in late 1990 the plant has processed a total of 212 t<sub>E</sub> of oxide fuel reloaded from the following reactor types:

- PHWR fuel with natural uranium: MZFR, FR2
- BWR fuel with low enriched uranium ( $\leq 3.5\%$ ): VAK, HDR and KRB
- PWR fuel with low enriched uranium ( $\leq 3.5\%$ ): KWO, GKN, KKS and NS Otto Hahn.
- PWR mixed oxide fuel (MOX, equivalent to 2.8 % enrichment): KWO and GKN (very small amount).

### *Liquid waste concentrate (HAWC)*

The remaining high active liquor (the so-called HAWC) of about 60 m<sup>3</sup> shall be converted into a solid waste form with a high chemical stability being suitable for final disposal.

In order to reach that goal the production of an borosilicate glass product is intended by using the recommended PAMELA vitrification process. By a highly sophisticated process developed by KfK/Germany and successfully demonstrated during the hot operation of PAMELA plant at Dessel/Belgium the high quality standard of the glass product will be ensured.

In this specification the resulting product will be characterized by both nominal and guaranteed values, taking the 16 relevant properties for final disposal into account as identified by the recommendation of the 'BMFT Arbeitskreis HAW-Produkte' /1/.

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## 1. Description of the PAMELA Vitrification Process

The PAMELA vitrification process as shown in fig. 1 is based on a liquid fed Joule heated ceramical <sup>-cined glass</sup> melter. By this one-step process (throughput about 30l/h) a liquid acid HLLW-solution is fed together with solid borosilicate glass<sup>(frit)</sup> particles (the so-called frit) into <sup>the mitter</sup> an oven. There, at a maximum temperature of about 1,150 °C, chemical and physical reactions take place, as following:

- *On the top of the glass pool surface*  
Drying of the aqueous fission product solution by evaporation,
- Calcination (i.e. converting nitrates into oxides) by heat impact,
- Vitrification by forming a homogeneous glass melt of frit and fission-product oxides.

After a mean residual time of about 80 hours the molten glass product is poured into a 180 l stainless steel canister. The canister is, however, filled by up to four ~~separately~~  
~~different~~ <sup>65</sup> pouring steps. batches.

Having passed a controlled cooling-down procedure the canister's lid is welded and the surface is decontaminated, hereafter. Up-to-its-transportation the canister is stored <sup>internally</sup> for further cooling down in a plant own pit storage.

The vitrification process will be carried out in the Belgian Belgoprocess-owned PAMELA Plant, taking quality measures as defined in DIN ISO 9000 into consideration. Furthermore the process control will ensure that the a.m. relevant properties will be taken <sup>accomplished</sup> into consideration. Although operating under Belgian responsibility, quality control checks and audits by German authorities or their representatives are possible to be carried out as agreed in the contract between WAK/KfK on the German side and ONDRAF/NIRAS and Belgoprocess on the Belgian side.

Details concerning the product quality are described in the Handbook of Process Qualification /2/ which is a document related to this specification.

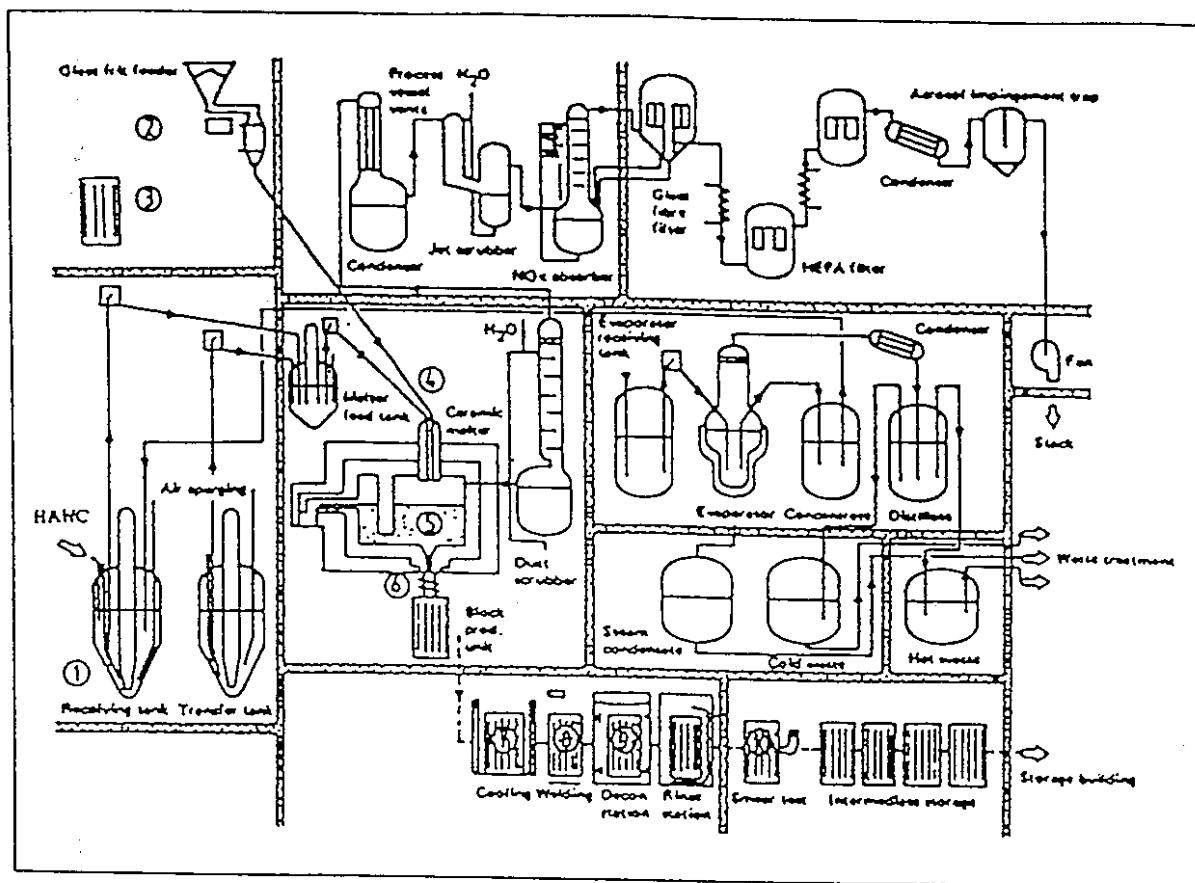


Figure 1: Simplified Flowsheet of PAMELA-Process

## 2. Characteristics of the Raw Waste

### 2.1 General Aspects

The aqueous liquid waste is presently stored in two tanks:

- about 43 m<sup>3</sup> liquor resulting from reprocessing in the time between 1971 up to 1987,
- about 16 m<sup>3</sup> liquor resulting from reprocessing up to 1990 including rinsing solutions,

~~the tank contents~~ <sup>have</sup> being of a slightly different chemical composition. Vitrification will be carried out by using a blend produced by a sophisticated mixing procedure /3/ to guarantee a uniform product quality.

The maximum specific total β-activity of these liquor is: ..... 1.85 E13 Bq/l,

~~and~~ <sup>7%</sup> minimum concentration of nitric acid is: ..... 3.5 mol/l.

Due to WAK's function of being a pilot plant the burn up of the reprocessed fuel varies in a wide range between 2,000 and 40,000 MWd/t, whereas a mean of about 25,000 MWd/t has to be taken into consideration.

As reprocessing in WAK was terminated in late 1990 and the fuel elements processed in WAK had a minimum discharge time of 3 years, and as vitrification will not take place earlier than 1997, every compound of the HAWC will have been cooled down at least for 10 years.

### 2.2 Chemical Composition of the Liquid Waste - HAWC

<sup>(WAK)</sup>  
The High Level Liquid Concentrate (called 'HAWC') is a composition of four main types of constituents:

- Fission products, especially cesium and strontium as main sources of β/γ-activity, reduced by the amount which was removed by feed clarification;

- *Uranium, plutonium and higher actinoids*, as main sources of  $\alpha$ -activity;
- *Corrosion elements*, especially iron and nickel, as inactive parts;
- *Chemicals* of the reprocessing process, especially sodium, as inactive parts, too.

While most of these elements or nuclides are ionogenically solved in the highly acid liquor, there is a certain amount of them left either undissolved or reprecipitated, forming a suspended solid phase. The mean concentration of this phase is about 14 g/l (as in store for the time being).

In table 1 the (calculated) nominal element concentrations as well as calculated maximum and minimum values, evaluated by the uncertainty margins either of the analytical methods or of the expected dilution by steam jets and air lifts, are listed in table 1. Beside the element concentration a calculated value is given for a virtual oxide concentration ( $\equiv$  calcinaté) as those numbers are relevant for the vitrification process itself.

Table 1: Nominal element concentrations and fractions of virtual oxide composition  
 (mean, minimum and maximum values) of the blend of both tanks storing  
 WAK-HAWC.

Element	Minimum values		Nominal values		Maximum values	
	Oxide residue	[wt.-%]	Element	[g/l]	Oxide residue	[wt.-%]
34 Se		0.08		0.06	0.10	0.11
37 Rb		0.39		0.29	0.35	0.32
38 Sr		0.75		0.79	1.04	1.22
39 Y		0.77		0.58	0.81	0.83
30 Zr		2.64		2.20	3.28	3.78
42 Mo		5.92		3.38	5.60	5.40
43 Tc		1.55		0.91	1.50	1.46
44 Ru		3.63		2.23	3.25	2.99
45 Rh		0.88		0.58	0.79	0.73
46 Pd		1.16		1.27	1.62	1.92
47 Ag		0.13		0.11	0.13	0.14
48 Cd		0.10		0.08	0.11	0.11
50 Sn		0.07		0.06	0.08	0.10
51 Sb		0.01		0.01	0.01	0.02
52 Te		0.72		0.52	0.72	0.72
55 Cs		4.17		3.14	3.68	3.35
56 Ba		3.26		2.33	2.88	2.61
57 La		3.00		2.05	2.66	2.43
58 Ce		2.12		3.01	4.09	4.73
59 Pr		1.75		1.42	1.83	1.89
60 Nd		4.30		5.06	6.53	8.00
62 Sm		0.44		1.12	1.44	2.10
63 Eu		0.13		0.13	0.16	0.19
64 Gd		0.52		0.39	0.49	0.47
92 U		9.15		5.73	7.48	6.35
93 Np		0.64		0.46	0.56	0.50
94 Pu		0.26		0.18	0.23	0.21
95 Am		0.60		0.43	0.52	0.47
96 Cm		0.03		0.02	0.02	0.02
24 Cr		3.05		1.86	3.01	2.97
25 Mn		0.33		0.22	0.38	0.41
26 Fe		11.84		7.06	11.16	10.69
28 Ni		1.76		1.28	1.81	1.83
11 Na		30.12		18.31	27.31	25.38
12 Mg		0.00		0.41	0.75	1.25
13 Al		0.00		0.06	0.12	0.20
19 K		0.10		0.22	0.29	0.42
20 Ca		0.62		0.35	0.55	0.41
F		2.02		0.02	0.02	0.02
Cl		0.02		0.01	0.02	0.02
P		1.97		1.03	2.60	3.08
$\Sigma$ Total		100.00		69.38	100.00	100.00

There is to point out that more than 25% of the mentioned material is inactive ~~so~~ <sup>WT.</sup> ~~sodium~~ <sup>sodium</sup> ~~oxide residue~~ listed waste oxide residue. ~~Na<sub>2</sub>O~~ sodium nitrate (or as calcinate  $Na_2O$ ) which was added as nitrite to the reprocessing process in order to stabilize a certain plutonium oxidation stage needed for obtaining a high factor of separation.

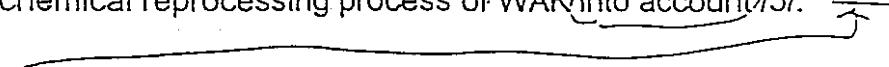
As concentration values are subject to changes due to dilution effects the total amount of the a.m. types of constituents is given in table 2.

Table 2: Total amount of HAW/constituents

<del>oxide residue</del>	
Total amount, as oxides (calcinate):	7,940 kg
Total amount, as elements:	6,095 kg
• Fission products: <del>elements</del>	2,785 kg
• Actinoids: <del>elements</del>	560 kg
• Corrosion elements:	918 kg
• Process chemicals: <del>elements</del>	1,792 kg
Part of elements as suspended particles:	820 kg

These total values are deemed to remain unchanged. The decay of the radionuclides will cause a certain conversion into stable elements of equivalent mass. However, there is no significant change in the chemical composition within the near future.

### 2.3 Activity Inventory of the HAWC Blend

While the chemical composition - as characterized above - is based on a chemical analysis, the activity-related data have been evaluated by burn up calculations performed with the KORIGEN code /4/, as they have to be predicted for the probable time of vitrification in 1997. Nevertheless, a detailed verification programme was carried out in order to confirm the reliability of the code and to introduce correction factors, taking all effects on the waste composition of the chemical reprocessing process of WAK into account /5/. 

Hence, the totals of the radiological data will be as following.

**Table 3:** Totals of activity related data (valid for 01.01.1997)

Total α-activity: .....	8.0 E15 Bq
Total β-activity: .....	8.9 E17 Bq
Total photon emission: .....	4.2 E17 s <sup>-1</sup>
Total neutron emission: .....	1.6 E10 n/s
Total heat generation: .....	76.4 kW

In addition the activity of main α- and β-nuclides are listed in Table 4.

**Table 4:** Nuclide activity of the WAK-HAWC blend, total values, valid for 01.01.1997 /4,5/.

Nuclide inventory***		Values
H-3**	[Bq]	7.5 E 12
Sr-90/Y-90 *	[Bq]	4.0 E 17
Tc-99	[Bq]	5.6 E 13
Ru-106/Rh-106 *	[Bq]	1.1 E 14
Sb-125	[Bq]	6.8 E 14
Cs-134	[Bq]	2.4 E 15
Cs-135	[Bq]	1.8 E 12
Cs-137/Ba-137m*	[Bq]	4.7 E 17
Ce-144/Pr-144 *	[Bq]	4.2 E 13
Eu-154	[Bq]	4.1 E 15
Eu-155	[Bq]	1.0 E 15
U-235	[Bq]	2.7 E 08
U-238	[Bq]	6.2 E 09
Np-237	[Bq]	1.0 E 12
Pu-238	[Bq]	1.1 E 14
Pu-239	[Bq]	2.2 E 13
Pu-240	[Bq]	3.8 E 13
Pu-241 (β)	[Bq]	2.7 E 15
Am-241	[Bq]	3.6 E 15
Am-242	[Bq]	2.4 E 13
Cm-242	[Bq]	2.0 E 13
Cm-244	[Bq]	4.1 E 15

\* = Sum of activity of mother and daughter nuclide

\*\* = 1 % of activity of KORIGEN burn up calculation

\*\*\* = Based on KORIGEN calculation, taking analytical values for U, Pu and actinoids into regard.

### 3. Characteristic Data of the Glass Frit

Borosilicate glass called GG WAK 1 has been chosen as a matrix material, having a composition as given in table 5. This granulated material is delivered by the manufacturer in the shape of small cylindric bodies, having a mean length and a diameter of 2 mm each.

Table 5: Composition of the glass frit GG WAK 1 including the tolerable bandwidth  $\pm$  scatter  $t_{tol}$

Oxide	Fraction [wt.-%]	Tolerances <small><math>t_{tol}</math> bandwidth of scatter</small>
SiO <sub>2</sub>	60.0	$\pm 0.5$ *
B <sub>2</sub> O <sub>3</sub>	17.6	$\pm 0.6$ *
Al <sub>2</sub> O <sub>3</sub>	3.1	$\pm 0.2$
Li <sub>2</sub> O	3.5	$\pm 0.2$
Na <sub>2</sub> O	7.1	$\pm 0.4$
CaO	5.3	$\pm 0.4$
MgO	2.2	$\pm 0.4$
TiO <sub>2</sub>	1.2	$\pm 0.2$

\* = In case of SiO<sub>2</sub> and B<sub>2</sub>O<sub>3</sub> a bandwidth of scatter of 1.5 wt.-% should be tolerable by way of exception due to reasons concerning the highly sophisticated manufacturing process. However the product should never be outside guaranteed parameters which will be ensured by suitable QA procedures.

#### 4. *Characteristic Data of the Empty Canister*

The canister is made of heat-resistant stainless steel, being a cylindrical package with an outer diameter of 430 mm. The purpose of the canister is

- to be the mould for the drained molten glass product,
- to provide attachment means for handling,
- to guarantee the integrity of the package during interim storage and handling.

The main dimensions are listed up in table 6, while a workshop drawing of the canister is given in figure 2. It is possible to stack a number of 10 filled canisters on each other. Top and bottom of the canister are formed in such a way that stacking can be easily done.

Table 6: Dimensions of the stainless steel canister

Main data of the canister	Value
Total volume	180 l
Net volume	150 l
Wall thickness	5 mm
Weight of empty canister	ca. 80 kg
Height including lid at room temperature	1.338 mm
Stacking height at storage temperature	≤ 1.275 mm
Diameter at room temperature	430 ± 2 mm
Maximum diameter at storage temperature	≤ 435 mm

In addition some characteristical properties of the canister's material are listed up in Table 7 which are relevant for the product quality, the heat flux, on the integrity during cooling down procedure of the glass and the interim storage period.

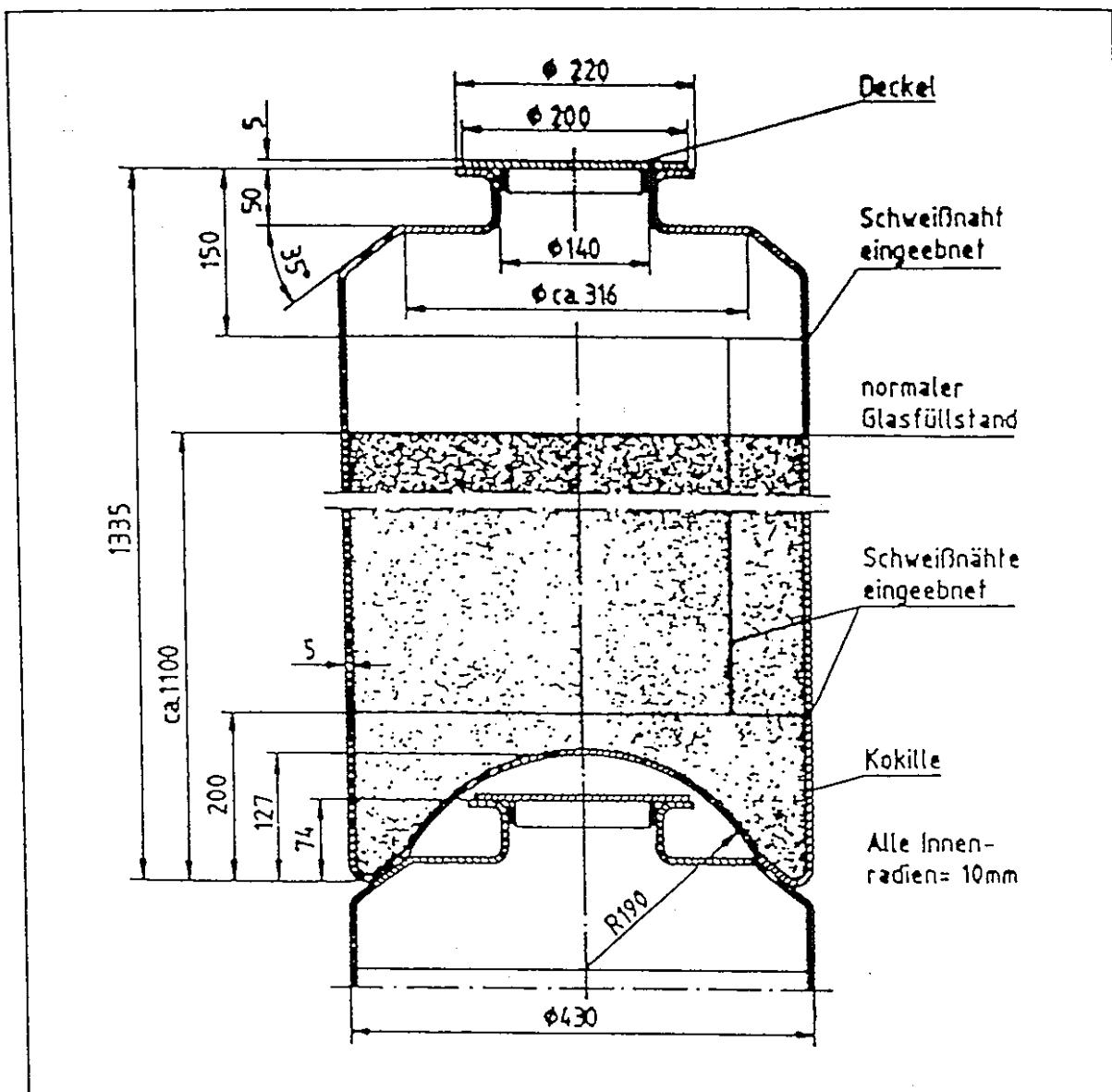


Figure 2: Workshop drawing of the stainless steel canister, dimensions valid for room temperature conditions, only.

Table 7: Main characteristic data of the canister's material

Property	Data						
Material	X15CrNi24/3 (1.4833 at DIN)						
Composition [wt.-%], * = max. value	C*      Si      Mn*      P*      S*      Cr      Ni 0.08    1.0    2.0    0.045    0.03    21-23    12-15						
Density	7.9 g/cm <sup>3</sup> (20 °C)						
Hardness	HB 192						
0.2-% Elasticity limit	210 N/mm <sup>2</sup>						
Tensile strength	500 - 750 N/mm <sup>2</sup>						
Expansion coefficient [1E-06·1/K]	200 °C 16	400 °C 17.5	600 °C 18	1000 °C 19.5			
Thermal conductivity [W/mK]	20 °C 15	500 °C 19					
Spec. heat capacity	0.5 J/gK (20 °C)						
Max. temperature of scaling resistance	1000 °C in air						
Emissivity	rolled, new	annealed	decontaminated				
- 200 °C	0.23	0.61	0.62				
- 300 °C	0.23	0.51	0.45				

Table 8: Mean composition of the WAK HAW glass product, calcinate incorporation 16 wt.-%

Compound	wt.-%
SiO <sub>2</sub>	50.4
B <sub>2</sub> O <sub>3</sub>	14.8
Al <sub>2</sub> O <sub>3</sub>	2.6
Na <sub>2</sub> O (frit)	6.0
Li <sub>2</sub> O	2.9
CaO	4.5
MgO	1.8
TiO <sub>2</sub>	1.0
Total, frit	84.0
ZrO <sub>2</sub>	0.5
MoO <sub>3</sub>	0.9
(Ru, Rh, Pd)O <sub>x</sub>	0.9
Cs <sub>2</sub> O	0.6
other fission products	4.0
U <sub>3</sub> O <sub>8</sub>	1.2
other actinoides	0.2
Fe <sub>2</sub> O <sub>3</sub>	1.8
CrO <sub>3</sub>	0.5
other corrosion elements	0.3
Na <sub>2</sub> O	4.4
PO <sub>4</sub> <sup>3-</sup>	0.4
other chemicals	0.5
Total, calcinate	16.0

### 5.2 Mean Radionuclide Inventory

The main radiological data of a waste glass canister are listed up in table 9 assuming that all the liquid waste will be conditioned in no more than 124 packages.

## 5. Characteristic Data of the Vitrified Product

The target value of waste calcinate incorporation into the glass product will be 16 wt.-%. The product yielded is a borosilicate glass having certain properties as discussed <sup>in the</sup> following taking requirements of the melting process, interim storage, and final disposal into regard.

### 5.1 Chemical Composition

By use of a target composition <sup>fix</sup> ~~84~~ <sup>(~~16~~)</sup> wt. % of the glass frit as specified in chapter 3, and <sup>wt.</sup> 16% of the average liquid waste composition as described in chapter 3, <sup>fix</sup> a borosilicate glass product will result as given in table 8.

Basing on an average filling level of 150 l for each canister the weight of the glass product will be about 400 kg ~~and a load of waste calcinate of 64 kg~~.

~~Canister capacity  
containing~~  
~~a portion of 64 kg in waste calcinate~~

**Table 9:** Mean total radiological data of a 150 l waste glass package (data valid for 01.01.1997)

total activity: .....	7.2 E15 Bq
$\alpha$ -activity: .....	6.5 E13 Bq
Photon emission: .....	3.4 E15 s <sup>-1</sup>
Neutron emission: .....	2.6 E08 n/s
Heat generation: .....	0.62 kW
Contact dose rate: .....	< 550 Gy/h
Dose rate in 1 m distance: .....	< 52 Gy/h

In table 10 the main radionuclides are listed up in addition. The nuclides have been selected by criteria as following:

- actual activity and surface dose rate,
- fissile material content,
- long term safety of a repository.

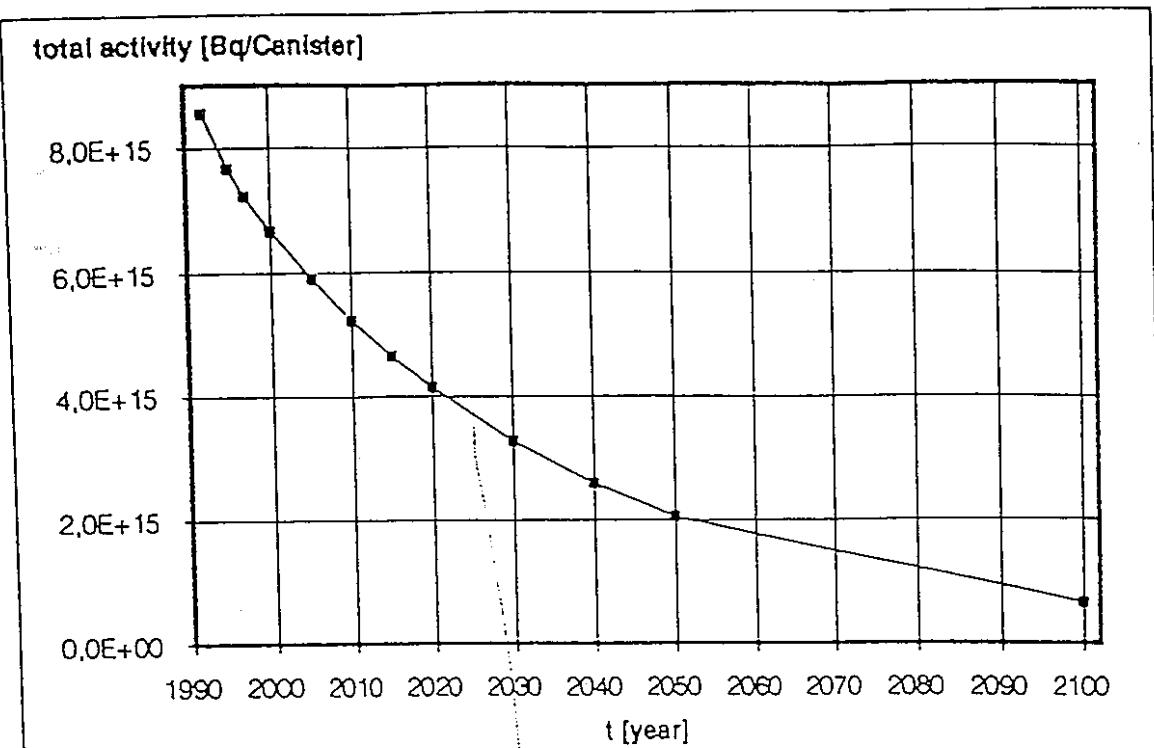
The data are the result of burn-up calculations /4/, which were verified by several radiochemical investigations /3, 5/ under the provision that volatile nuclides (especially the rare gases, carbon and iodine) will not be incorporated in a glass matrix.

Table 10: Mean nuclide inventory of a waste glass canister (150 l glass, 16 wt-% calcinate incorporation, nominal raw waste composition, data valid for 01.01.1997)

Nuclide inventory	Activity [Bq/Canister]
Sr-90/Y-90 *	3.2 E 15
Tc-99	4.5 E 11
Ru-106/Rh-106 *	8.9 E 11
Pd-107	3.4 E 09
Sb-125	5.5 E 12
Cs-134	1.9 E 13
Cs-135	1.5 E 10
Cs-137/Ba-137m *	3.8 E 15
Ce-144/Pr-144 *	3.4 E 11
Pm-147	9.2 E 13
U-235	2.2 E 06
U-238	5.0 E 07
Np-237	8.1 E 09
Pu-238	8.9 E 11
Pu-239	1.8 E 11
Pu-240	3.1 E 11
Pu-241 ( $\beta$ )/Am-241 *	5.7 E 13
Am-242	2.0 E 11
Cm-242	1.6 E 11
Cm-244	3.3 E 13

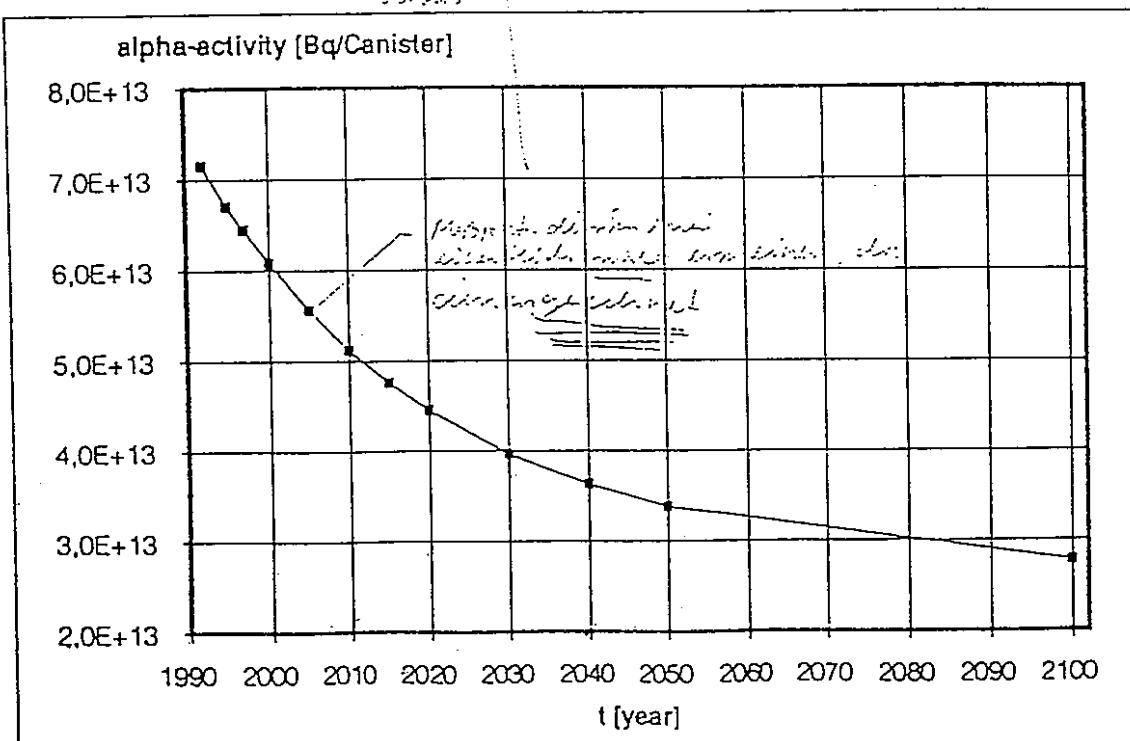
\* = Sum of activity of mother and daughter nuclides

For long-term studies the decrease of the total activity,  $\alpha$ -activity, the  $\gamma$  photon emission, the neutron emmission rate, and/ the heat generation are of certain importance. The curves are shown in figures 3 a, b, c, d, e /4/.



*Calculated decrease of total activity over a glass canister versus time*

Figure 3 a: Total activity vs. time diagramme



*Calculated decrease of alpha-activity over a glass canister versus time*

Figure 3 b: alpha-activity vs. time diagramme

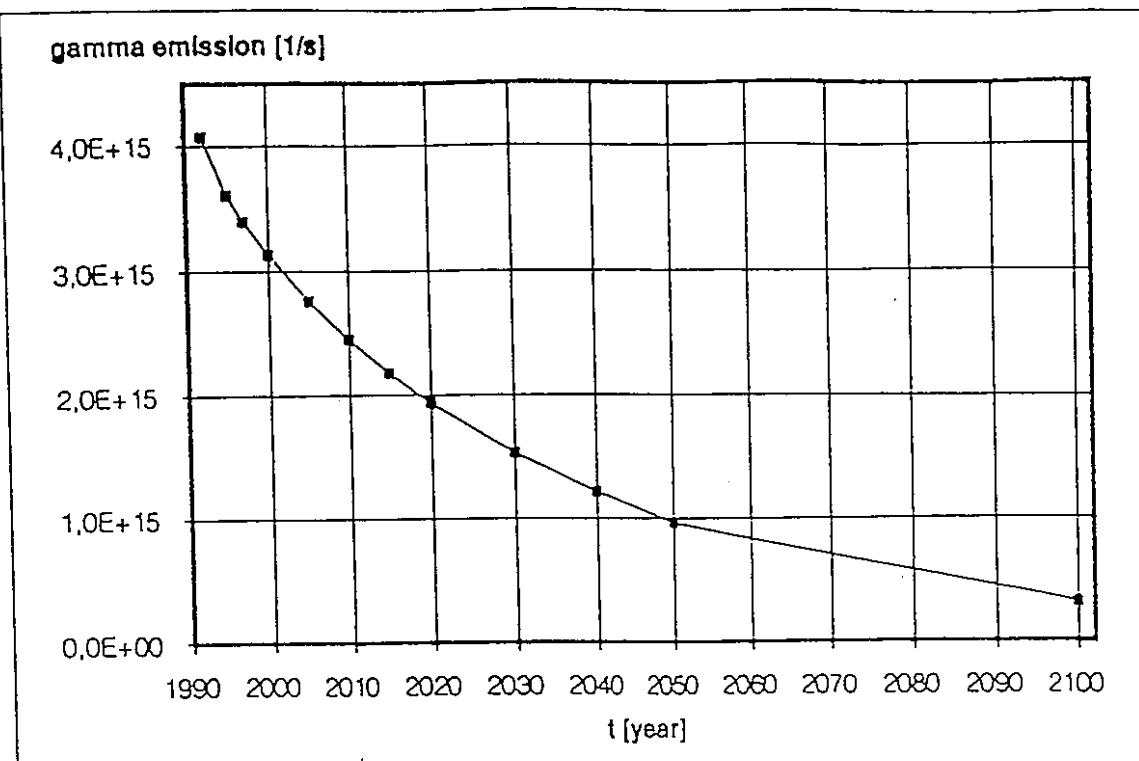


Figure 3 c: Photon emission vs. time-diagramme  
*measured values*  
*calculated values*

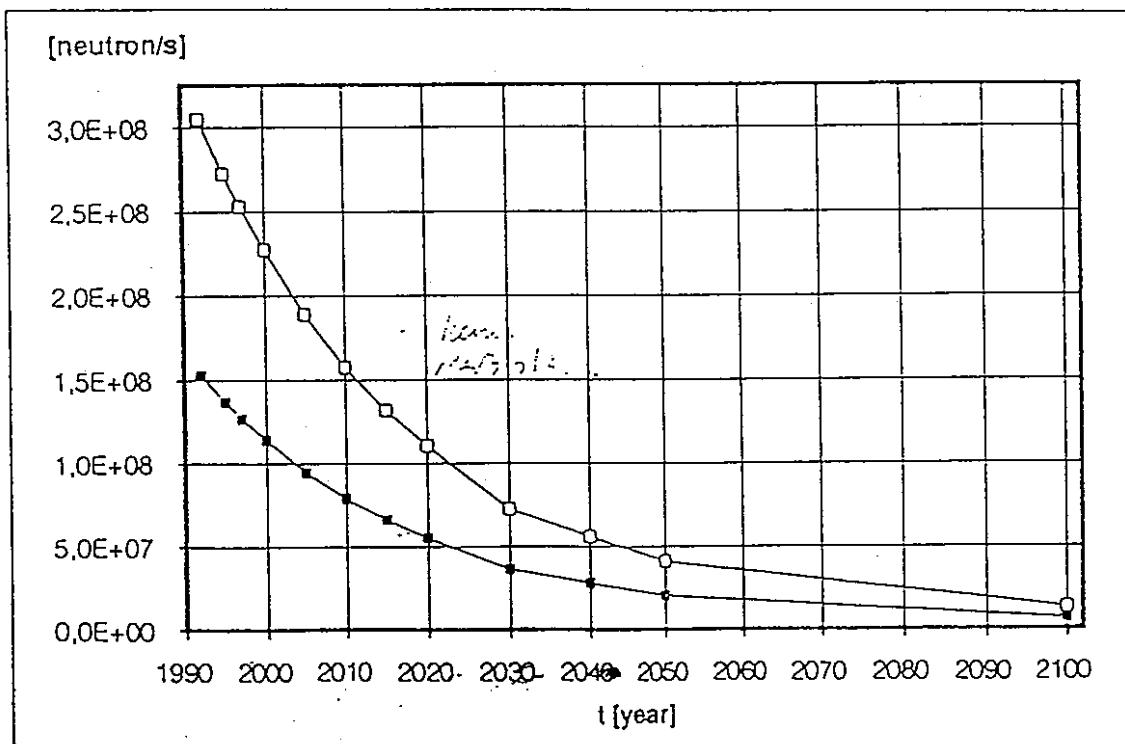
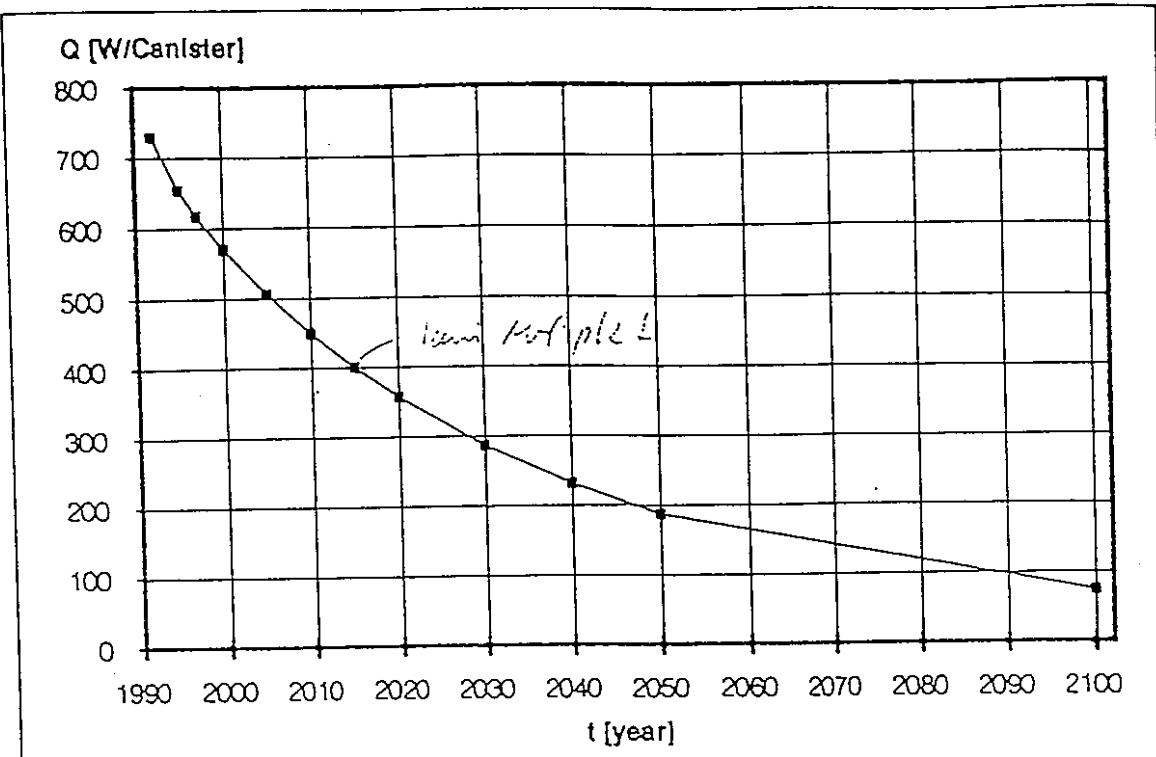


Figure 3 d: Neutron emmission rate vs. time-diagramme  
*calculated*  
*values*

*(of one glauconite)*



*calculated*  
Figure 3 e: Heat decay rate vs. time diagramme-

### 5.3 Characteristical Physical Data /6/

For operation of the melter, for interim storage, and final disposal a series of various physical properties has to be taken into consideration, which are listed up as following.

*phys. prop.*  
**Dynamic Viscosity:** A value which characterises the fluidity behaviour of the melt during the vitrification process and while pouring the product into the canister. The viscosity is specified as following:

Viscosity of glass product at 1,150 °C: .....	45 dPa·s
Viscosity of glass product at 950 °C: .....	513 dPa·s
Viscosity of pure frit at 1,150 °C: .....	101 dPa·s
Viscosity of pure frit at 950 °C: .....	1390 dPa·s

**Specific electrical resistance:** This property is a key value for the operation of a Joule heated ceramical melter.

*ceram. elem.*  
Spec. electrical resistance of the glass product at 1,150 °C: ..... 7 Ω·cm

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Spec. electrical resistance of the glass product at 950 °C:	19 Ω·cm
Spec. electrical resistance of pure frit at 1,150 °C:	9.4 Ω·cm
Spec. electrical resistance of pure frit at 950 °C:	27 Ω·cm

*Density:* By pycnometrical measurements the following data were determined:

Density of the glass product at 20°C:	2.70 g/cm <sup>3</sup>
Density of the glass frit at 20°C:	2.48 g/cm <sup>3</sup>
Density of the glass melt at 1,150°C:	2.38 g/cm <sup>3</sup>

*Characteristical Temperatures and Thermal Expansion:* These figures describe the influence of heat on a glass product. The transformation temperature ( $T_G$ ) indicates a viscosity of 1 E 13 dPa·s. Up to this temperature the thermal expansion behavior can be described by a linear coefficient ( $\alpha$ ). In excess of  $T_G$  the softening temperature ( $T_E$ ) is reached when a glass sample loses its shape rapidly by melting. Another important value is the temperature of maximum cristal growth ( $T_K$ ). It characterises a range of temperature where crystallisation leads to thermodynamical stable products of a cristalline phase supported by kinetic factors at elevated temperatures.  $T_G$ ,  $T_E$  and  $\alpha$  are determined by dilatometric measurements.

Transformation temperature, $T_G$ :	513 °C
Softening temperature, $T_E$ :	569 °C
Temperature of maximum crystallisation rate, $T_K$ :	750 °C
Thermal expansion coefficient (20 - 400 °C), $\alpha$ :	9.3E-06/K

*Heat conductivity and heat capacity:* These data are relevant for the heat dissipation especially during the initial interim storage phase:

Heat conductivity at 400 °C:	1.42 W/m·K
Heat conductivity at 130 °C	1.20 W/m·K
Specific heat capacity at 400 °C:	(?) 1.22 xx) 1.xx J/g·K
Specific heat capacity at 300 °C:	(?) 1.17 xx) 1.xx J/g·K

*Volatilisation:* An overall value for volatilisation has been measured for the glass melt:

*gratig*  
Rate of volatilisation at 1,150 °C: 5.0 E-03 g/cm<sup>3</sup>/h

*mit aust. melt surface bevoegen!*

xx) W/K - Ermittlung

*For*

In case of interim storage conditions ( $< 450^{\circ}\text{C}$ ) the release of volatile compounds is negligible. Slightly volatile oxides of Cs and Ru will become solid at least at the canisters inner surface which is cooler than the glass product's center. The production of gaseous nuclides by spontaneous fission (like Kr-85 and I-129) and the decay chains of U- and Pu-Isotopes (Rn-220) will not cause a significant contribution to any activity release of the package *[7]*.

*must done  
because in  
zeni :: :: ::  
sehr  
angeneh  
Brenn wär  
bei  $T < 450^{\circ}\text{C}$   
verhindern  
in aktiver  
umgebung!*

*Cristallisation and Phase Separation:* During the cooling down procedure ( $600 - 800^{\circ}\text{C}$ ) some crystalline phases may grow, however, the fraction of crystalline phases is expected to be lower than 1 % by volume. In addition the formed phases like Diopside do not have less favorable properties as the glass itself. Furthermore the comprehensive R&D-work did not give any hints for the occurrence of highly soluble 'yellow phase'.

Noble metals like Ru, Rh and Pd may also form separate phases like  $\text{RuO}_2$  needles like crystals or spherical alloys separation ( $\text{RhPd}_x$ ), both at micron scale. However, these phases do not have any relevance concerning long term safety, as there is no significant activity contribution of highly radiotoxic nuclides with long half lives. By the layout of the melter geometry a homogeneous distribution of these phases will be ensured. In addition they will be completely imbedded by the glass.

*regarding to  
particle size.*

#### 5.4 Corrosion in Aqueous Solutions and Brines

By an Soxhlet-Leach-Test in distilled water at temperatures of  $100^{\circ}\text{C}$  an indicative rates ( $R_L$ ) for short term corrosion were yielded

$R_{L,1}$ (mean value after 1d):	(?) 9,5 $E^{-04} \text{ *} \text{ }$	x,x E-xx g/cm <sup>2</sup> /d
$R_{L,3}$ (mean value after 3d):	(?) 8 $E^{-04} \text{ *} \text{ }$	x,x E-xx g/cm <sup>2</sup> /d
$R_{L,10}$ (mean value after 10d):	(?) 6 $E^{-04} \text{ *} \text{ }$	x,x E-xx g/cm <sup>2</sup> /d
$R_{L,30}$ (mean value after 30d):	(?) 4 $E^{-04} \text{ *} \text{ }$	9,0 E-04 g/cm <sup>2</sup> /d

using inactive samples with a waste load of 16 wt.-%.

With respect to long term safety static corrosion experiments which were carried out following the MCC-rules at  $110^{\circ}\text{C}$  and  $190^{\circ}\text{C}$  in brines as:

- solution 1:  $\text{NaCl}-\text{KCl}-\text{MgCl}_2-\text{Na}_2\text{SO}_4-\text{H}_2\text{O}$

*\* WAK-Ermittlung*

- solution 2: NaCl-CaSO<sub>4</sub>-(KCl-MgCl<sub>2</sub>)-H<sub>2</sub>O

changing the (sample)-surface to (brine)-volume ratio S/V from 1.000 to 10.000 m<sup>-2</sup> using inactive samples with a 13, 16 and 19 wt.-% waste incorporation. The results are given in fig. 4.

[Hinweis von INE über die Art der Ergebnisdarstellung benötigt] /8/.

Umhän  
Formu  
lung

## 6. Guaranteed Parameters

The data provided in chapter 5 are nominal or indicative values describing an standard glass product on basis of an average acitivity level and an average chemical composition of wase solution and of glass frit. By an accepted bandwidth *of scatter* of the waste load of 13 to 19 wt.-%, several data are subject to vary, however they will always be covered by the guaranteed parameters.

Table 11: Guaranteed parameters (maximum values) of the vitrified product of WAK-HAWC

Guaranteed parameters	Guaranteed value
Maximum load of calcinate	19 wt.-%
Bandwidth of glass composition	see table 12
Mass of package	≤ 550 kg/package
Dimensions at room temperature	see fig. 2
Max. stacking height	1,275 mm
Transferable surface contamination: α-Emitters	≤ 0.4 Bq/cm <sup>2</sup>
β-Emitters	≤ 4.0 Bq/cm <sup>2</sup>
Maximum activity Sr-90/Y-90 *	4.4 E15 Bq/package
Maximum activity Cs-137/Ba-137m *	5.3 E15 Bq/package
Maximum total β/γ-activity	1.0 E16 Bq/package
Neutron emission	≤ 4.0 E08 n/s/package
Uranium, total weight	≤ 6,880 g/package
Plutonium, total weight	≤ 206 g/package
Curium, total weight	≤ 20.2 g/package
Dose rate: β/γ at surface	≤ 762 E2 Gy/h
β/γ at 1 m distance	≤ 72 E1 Gy/h
Neutrons at surface	≤ 5 E-3 Gy/h
Neutrons at 1 m distance	≤ 8 E-4 Gy/h
Heat generation	≤ 850 W/package

\* = Sum of activity of mother and daughter nuclide.

Taking the mentioned bandwidth *of scatter* of composition into account as well as the most unfavorable activity distribution in the HAWC, and accepting a filling level of 175 l of glass product in the canister, the main maximum values of a package are as listed up in table 11.

The guaranteed and accepted bandwidth of the chemical composition is provided in table 12.

*(f scatt)*  
Table 12: Guaranteed bandwidth of glass product composition

Compound	Minimum load* [wt.-%]	Maximum load* [wt.-%]
Fraction of frit	87.0	81.0
• SiO <sub>2</sub>	≤ 52.7	≥ 48.1
• B <sub>2</sub> O <sub>3</sub>	≤ 15.9	≥ 13.7
• Al <sub>2</sub> O <sub>3</sub>	≤ 2.9	≥ 2.3
• Na <sub>2</sub> O	≤ 6.6	≥ 5.4
• Li <sub>2</sub> O	≤ 3.3	≥ 2.6
• CaO	≤ 4.9	≥ 4.0
• MgO	≤ 2.2	≥ 1.5
• TiO <sub>2</sub>	≤ 1.2	≥ 0.8
Fraction of calcinate	13.0	19.0
• Fission products + Zr	≥ 4.6	≤ 9.4
- ZrO <sub>2</sub> (hulls)	≥ 0.3	≤ 0.8
- MoO <sub>3</sub>	≥ 0.7	≤ 1.2
- (Ru, Rh, Pd)O <sub>2</sub>	≥ 0.6	≤ 1.3
- Cs <sub>2</sub> O	≥ 0.4	≤ 0.8
- others	≥ 2.6	≤ 5.3
• Actinoids	≥ 0.9	≤ 2.1
- U <sub>3</sub> O <sub>8</sub>	≥ 0.8	≤ 1.8
- others	≥ 0.1	≤ 0.3
• Corrosion elements	≥ 1.9	≤ 3.3
- Fe <sub>2</sub> O <sub>3</sub>	≥ 1.3	≤ 2.3
- CrO <sub>3</sub>	≥ 0.3	≤ 0.6
- others	≥ 0.3	≤ 0.4
• Process chemicals	≥ 3.6	≤ 6.8
- Na <sub>2</sub> O	≥ 3.2	≤ 5.8
- P <sub>2</sub> O <sub>5</sub>	≥ 0.4	≤ 0.6
- others	≥ 0.0	≤ 0.4

\*) = Due to the maximum/minimum calculation mode the sum of certain wt.-% data is ≠ 100 g.

## 7. Container Data Sheet

Following the list of relevant properties and characteristical values of HAW glass products in a final storage facility, and the requirements for non-heat generating residues /9/ - as already practiced /1/ the data sheet will provide information-data-as listed hereafter:

- Type of waste product, name of specification referred
- Package identification label
- Type and dimensions of canister [mm]
- Name and address of conditioner
- Type of conditioning process
- Date of conditioning
- Process qualification certificate
- Total  $\beta/\gamma$ -activity [Bq/canister]
- Total  $\alpha$ -activity [Bq/canister]
- Activity of relevant nuclides [Bq/canister]
- Code used for calculating the actual nuclide spectrum
- U-233 and U-235 content [g/canister]
- Pu-239 and Pu-241 content [g/canister]
- Content of neutron absorber elements [g/canister]
- Activities of other fissile actinoids [Bq/canister]
- Thermal power [W/canister]
- $\gamma$  contact dose rate [Gy/h]
- $\gamma$  dose rate in 1 m distance [Gy/h]
- Neutron contact dose rate [Gy/h]
- Neutron dose rate in 1 m distance [Gy/h]
- Transferable  $\alpha$ -surface contamination
- Transferable  $\beta/\gamma$ -surface contamination
- Fraction of incorporated waste calcinate [wt.-%]
- Total weight of package [kg]

Data which are defined as guaranteed parameters were listed up separately and were always compared with the limits agreed upon in chapter 6.

## 8. Quality Assurance Policy

The main goal of the operation of the PAMELA vitrification <sup>plant</sup> process is to yield a glass product whose properties are at least within the limits of the a.m. *Guaranteed Parameters* while, however, it is worth striving for to be as close as possible to the *Nominal Values*.

In order to achieve that goal certain measures have been taken which were described in the following:

- The company policy of the operator of the vitrification plant is laid down in the company's Quality Assurance Handbook, which will clearly point out that the safe operation and a high quality of the product are the main goals.
- Relevant quality standards like the ISO 9000 series are guiding the operational and administrative work, especially in dealings with suppliers of goods, materials and services.
- To ensure independence on matters which relate to the product quality the QA-Manager will have direct access to the company's directors, being independent from the operational manager.
- To make sure that requirements of the final disposal in Germany can be fulfilled a handbook for the process qualification has been written /2/ which is a related document to this specification. The handbook describes In the measure to be taken in order to ensure the reliability and tolerable bandwidth of the data provided in the container data sheet (as described in chapter 7), which are in concordance with the a.m. mentioned relevant properties for final disposal /2,9/.

*Documents related/Literature*

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- /2/ E. Ewest, W. Kunz: Handbuch zur Verfahrensqualifikation: Verglasung des WAK-HAWC in der PAMELA, GNS-B 130/91 (Rev. 4), Feb. 1992.
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- /4/ WAK-Dokument W 0961806 von 2/93, HAWC-Datensatz (KORIGEN-Rechnung für WAK-HAWC).
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- /9/ P. Brennecke, E. Warnecke (Hrsg.): Anforderungen an endlagernde radioaktive Abfälle (vorläufige Endlagerungsbedingungen, Stand April 1990 in der Fassung vom Juli 1991) - Schachtanlage Konrad -, BfS-Bericht ET-3/90, Rev. -1, Salzgitter (1991).