Dr. Richard L. Hahn

BROOKHAVEN
NATIONAL LABORATORY

Chemistry Department 555
P.O. Box 5000
Upton, NY 11973-5000
Phone 631 344-4337
Fax 631 344-5815
Hahn1@bnl.gov

managed by Brookhaven Science Associates
for the U.S. Department of Energy

www.bnl.gov

July 25, 2000

Dear Colleague:

As promised, I am sending you a set of the transparency copies from the Radon Workshop that was held in Sudbury prior to the Neutrino-2000 Conference.

Sincerely,

Dick

Richard L. Hahn

Radon Background in Rare-Event Experiments

Satellite workshop of 'NEUTRINO 2000', SUDBURY, Canada

PROGRAM (quasi-final)

Date:	Wednesday June 14, 2000
Time:	8:00 a.m. – 6:30 p.m.
Place:	Alphonse Raymond Education Building, Laurentian University
Organizers:	G. Heusser, T. Kirsten and R.L. Hahn

The listed presentations should help to trigger intense discussions. Additional entries and ad-hoc contributions in addition to the program below are encouraged, as time permits (bring your transparencies etc., even if they are not formally organized). Scheduled times are approximate. Talks are nominally allocated 20 minutes, 15 for the presentation, and 5 for discussion. General aspects can also be discussed in the round-table discussion session at the end of the workshop.
No abstracts are required for the talks. We are considering posting copies of the transparencies on the Conference Web Site, so please bring a set of copies of your transparencies to give to the organizers.

8:00	REGISTRATION AND REFRESHMENTS.	
9:00	Introduction	
	Organizers	Welcome and definition of the problems to be discussed.
9:15	SNO	
	I. Levine	The SNO cover gas system
	H. Lee	Radon in the SNO Water System
	J. Farine	Two radon barriers (cavity liner, o-rings) and detection of 220Rn at SNO
10:15	HOMESTAKE	
	K. Lande	Radon induced background in Cl or I
10:35	COFFEE BREAK	
10:55	SAGE	
	I. Mirmov	Rn sources in chemical technology of SAGE and the methods of protection
	V. Gorbachev	The methods of suppression of Rn influence on measurements in SAGE
11:35	GALLEX/GNO	
	G. Heusser	Suppression of Rn in the GALLEX/GNO counting systems
	W. Hampel	Radon recognition and correction in the 71Ge-proportional counting
12:15	Super-Kamiokande	
	Y. Takeuchi	Present status and future prospects of radon reduction in Super-Kamiokande
12:35	LUNCH	

14:00	BOREXINO	
	G. Heusser	Radon off-line monitoring program in BOREXINO, some general remarks
	S. Schoenert	Radon measurements with the Counting Test Facility (CTF)
	W. Rau	Emanation measurements
	W. Rau	Radon in nitrogen and a new purification method for nitrogen
	J. Kiko	Electrostatic detector for radon monitoring in air
	H. Simgen	Measurements of Ra/Rn in water
	A. P. Pocar	The radon filtering VSA system
16:00	COFFEE BREAK	
16:20	Double Beta-Decay	
	A. Barabash	Experimental limit on 222Rn activity in liquid Ar
16:40	DARK MATTER SEARCH	
	V. Kudryavtsev	Alpha background from radon decay in the UK dark-matter search
17:00	ROUND TABLE DISCUSSION OF RADON TOPICS SUCH AS	
	(Discussion leaders to be announced)	
-	Generalization of experiences so far	
	-	Avoidance or reduction of Rn
	-	Analysis / monitoring
	-	Hard-core limits
	-	Radon limits in new projects
	-	Desirability of cross project exchanges
	-	Possibilities for practical collaboration
	-	Other items
18:30	End of Workshop	

The SNO Cover-Gas System

Ilan Levine, Carleton University
For the SNO Collaboration

Radon Workshop

The pages of the following ~~Poster~~ talk should be displayed as shown below. (This is page 0….)

1	2		8	9
13(portrait layout)	4		10	11
5	6	7		12

The drawings and graphs don't show up so well here.Duplicates are attached to this same log book entry for those who want to see higher quality images

Purpose:

1) Protect detector water from ^{222}Rn and O_2 in laboratory air:

- **H_2O O_2 <50ppb, Temp ~ 10°C (Biology)**
- **D_2O < $3.7*10^{-15}$ gTh/gD_2O and <$4.5*10^{-14}$ gU/gD_2O**
- **H_2O (fiducial) < $3.7*10^{-14}$ gTh/gH_2O and <$4.5*10^{-13}$ gU/gH_2O**
- **Laboratory air has radon levels ~10^5 times higher than target for U-chain.**

2) Protect structural integrity of water vessels:

- **ΔP (due to mine ventilation changes or water circulation.)**

Design Concept:

1) Gas above water normally stagnant → ^{222}Rn in gas space will decay, leaving radon free gas "blanket" over water.

2) System has both passive and active response to air ingress.

- **System has vent open to laboratory air to allow pressure in vapor space to equalize.**
- **^{222}Rn-free N_2 gas in "buffer tanks" available when $P_{lab} > P_{vessel}$.**
- **Boil large quantities of nitrogen from liquid N_2 dewar when environment changes.**

Maximum Acceptable ^{222}Rn in D_2O

Criterion: no more than 10% of NC rate (~10 to 20 evt/day) due to ^{238}U chain

$$^{214}Bi \xrightarrow[2.8\%]{\beta\ 0.8\,MeV} {}^{214}Po^{*} \xrightarrow[55\%]{\gamma\ 2.45} {}^{214}Po$$

So ~1.5% of ^{214}Bi decays produce 2.45 MeV γ.

~ $\frac{1}{750}$ of these γ produces a free neutron in D_2O

$$\frac{dN_{214}}{dt} = 750 \cdot \frac{100}{1.5} = 50{,}000\ day^{-1}$$

$$= \lambda_{214} N_{214} = \frac{-\ln 1/2}{\frac{20\ min}{1440\ min/d}} N_{214}$$

$$= 49.9\ d^{-1}\ N_{214}$$

$$\rightarrow N_{214} = 1002\ ^{214}Bi$$

$$\lambda_{238} N_{238} = \lambda_{214} N_{214}$$

$$\Rightarrow N_{238} = \frac{49.9 \times 1002}{\ln 1/2 / 4.5 \times 10^9 y \cdot 365 \, d/y} \text{ atoms}$$

$$= 1.2 \times 10^{17} \text{ atoms}$$

$$= 1.2 \times 10^{17} \cdot \frac{238 g}{6 \times 10^{23}} = 4.7 \times 10^{-5} gm$$

Have 10^9 gm D_2O

$\rightarrow$ In ^{238}U max contamination

is $\sim 4.7 \times 10^{-14} g \, ^{238}U / g D_2O$

$$N_{222} = \lambda_{238} N_{238} / \lambda_{222}$$

$$= 1.2 \times 10^{17} \text{ atoms } 4.2 \times 10^{-13} d^{-1} / 0.18 d^{-1}$$

$$= 2.8 \times 10^{5} \text{ atoms in } D_2O$$

$$\rightarrow C_{222}^{D_2O} = \frac{2.8 \times 10^{5} \text{ atoms}}{\frac{10^6}{1.1} \ell} = 0.31 \frac{\text{atoms}}{\ell}$$

10°c Ostwald Coeff. = 0.385

20°c O = 0.277

$$O \equiv \frac{\text{Volume conc. Rn in } D_2O}{\text{Volume conc. Rn in Gas}} \Big]$$

$$C_{222}^{CG}(10°c) = \frac{0.31 \text{ atoms}/\ell}{0.388} = 0.8 \frac{\text{atoms}}{\ell}$$

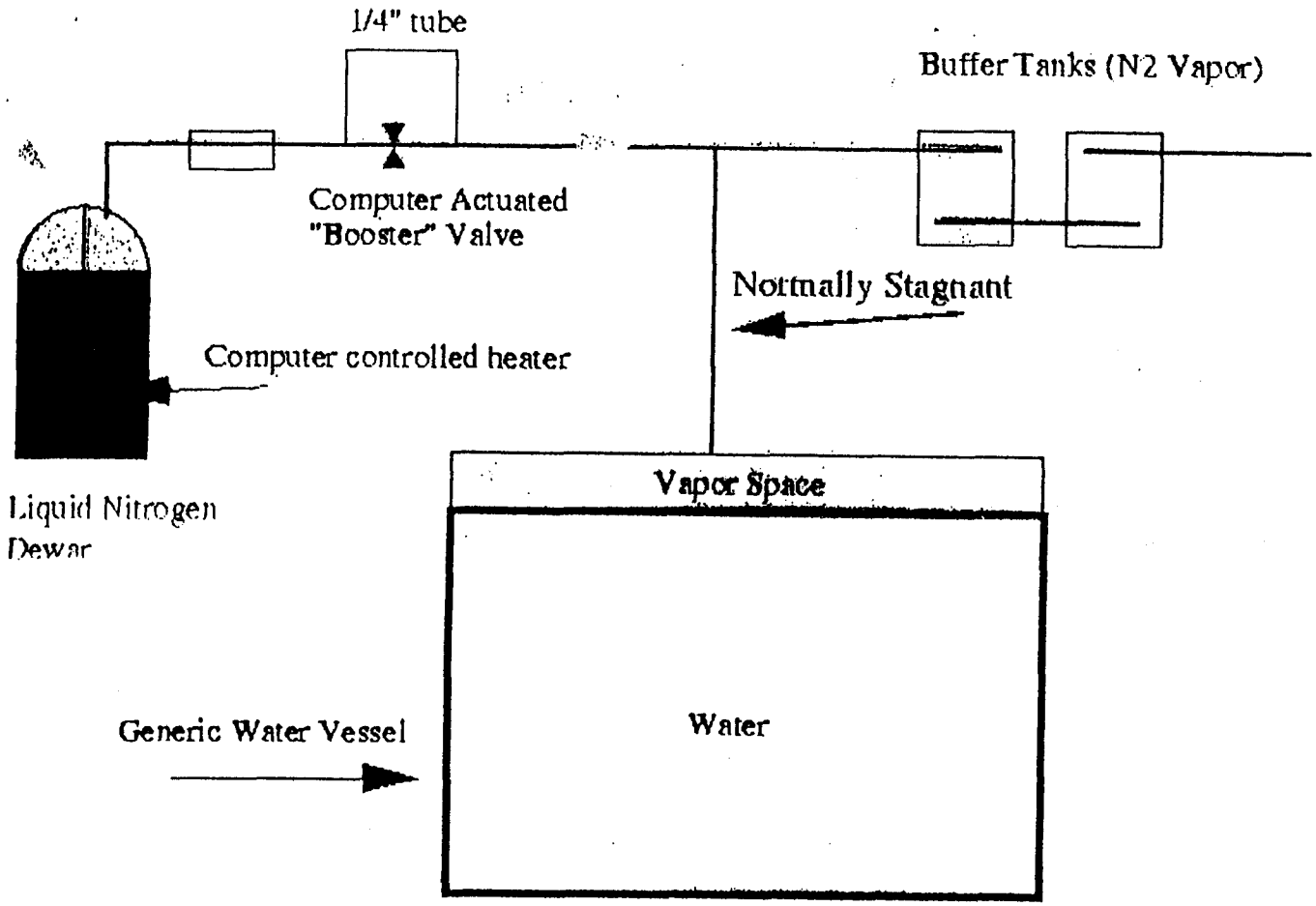
$$C_{222}^{CG}(20°c) = 1.1 \text{ atoms}/\ell$$

So Max acceptable ^{222}Rn in CG $\sim$ 1 atom/ℓ

Varies from $\sim 6 \times 10^4$ atoms/ℓ
to $\sim 18 \times 10^4$ atoms/ℓ

$\Rightarrow$ CG must be $\sim 10^5$
ower in ^{222}Rn than air.

Note: We have assumed that
ve get no benifit from
Stagnant Water in neck.
Very Pessimistic



Conceptual Design of the SNO Cover Gas System.

Normally, no flow in or out of vessel vapor space.
Flow of Rn–free N2 gas past the vapor space.
Buffer volume of relatively clean vapor in buffer tanks for quick pressure fluctuations.
Computer control of boil off rate (internal heater and "booster valve".)

Details of System:

1000 liter liquid N_2 dewar with computer controlled internal heater to boil liquid nitrogen. Typically supply ~30 liters gas/ minute (lpm).

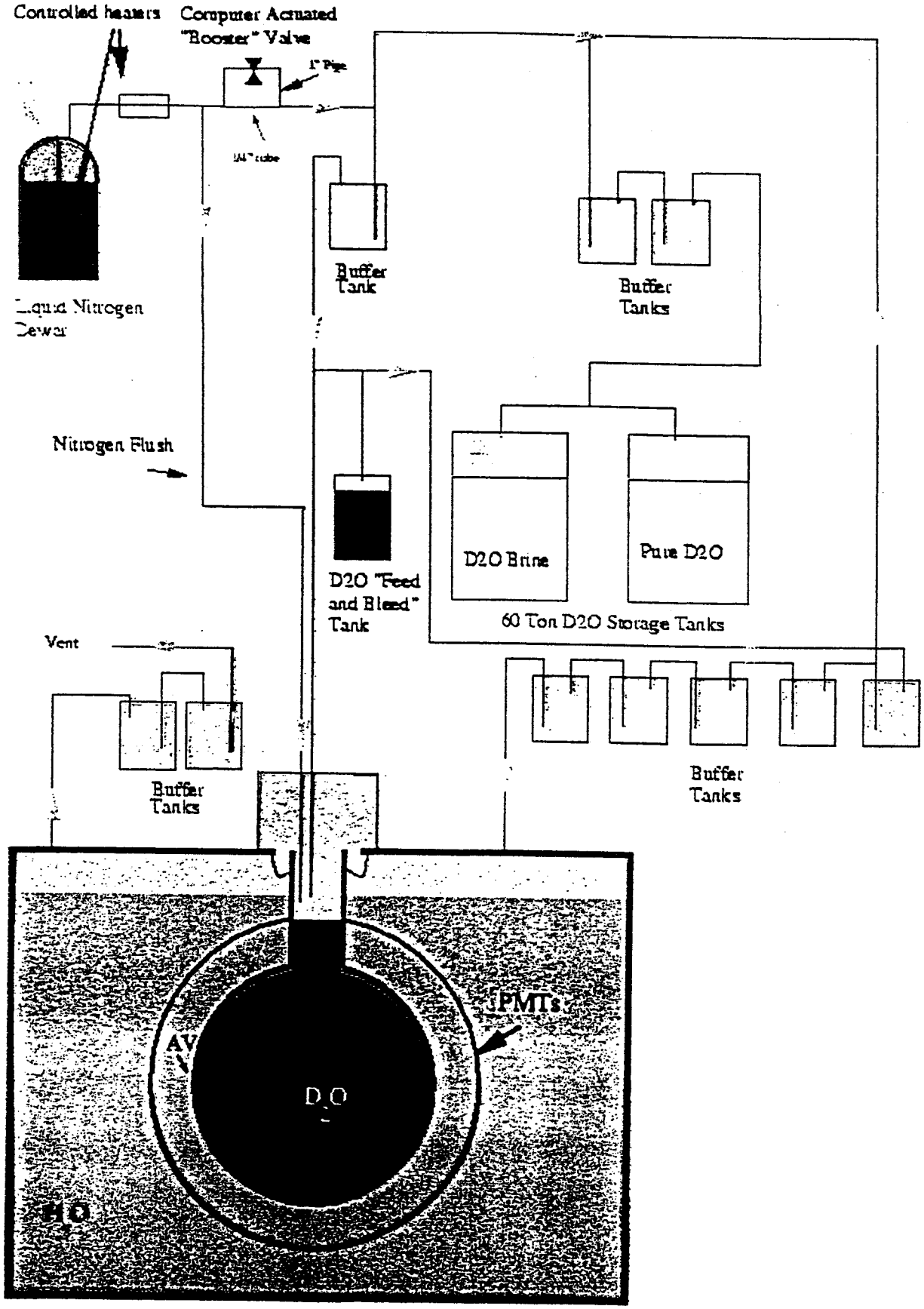
A direct line from the dewar to the D_2O vapor space provides ~10 liters/minute to "flush" radon enriched gas away from the D_2O and maintain radon concentration at ~ 10 atoms/liter gas.

When lab pressure swing is detected, buffer tanks supply initial protection to vapor space. Simultaneously, internal heater turns to full power and high conductance path is opened ("booster valve"), generating ~ 200 lpm N_2.

Vapor space of entire system ~150 m^3.

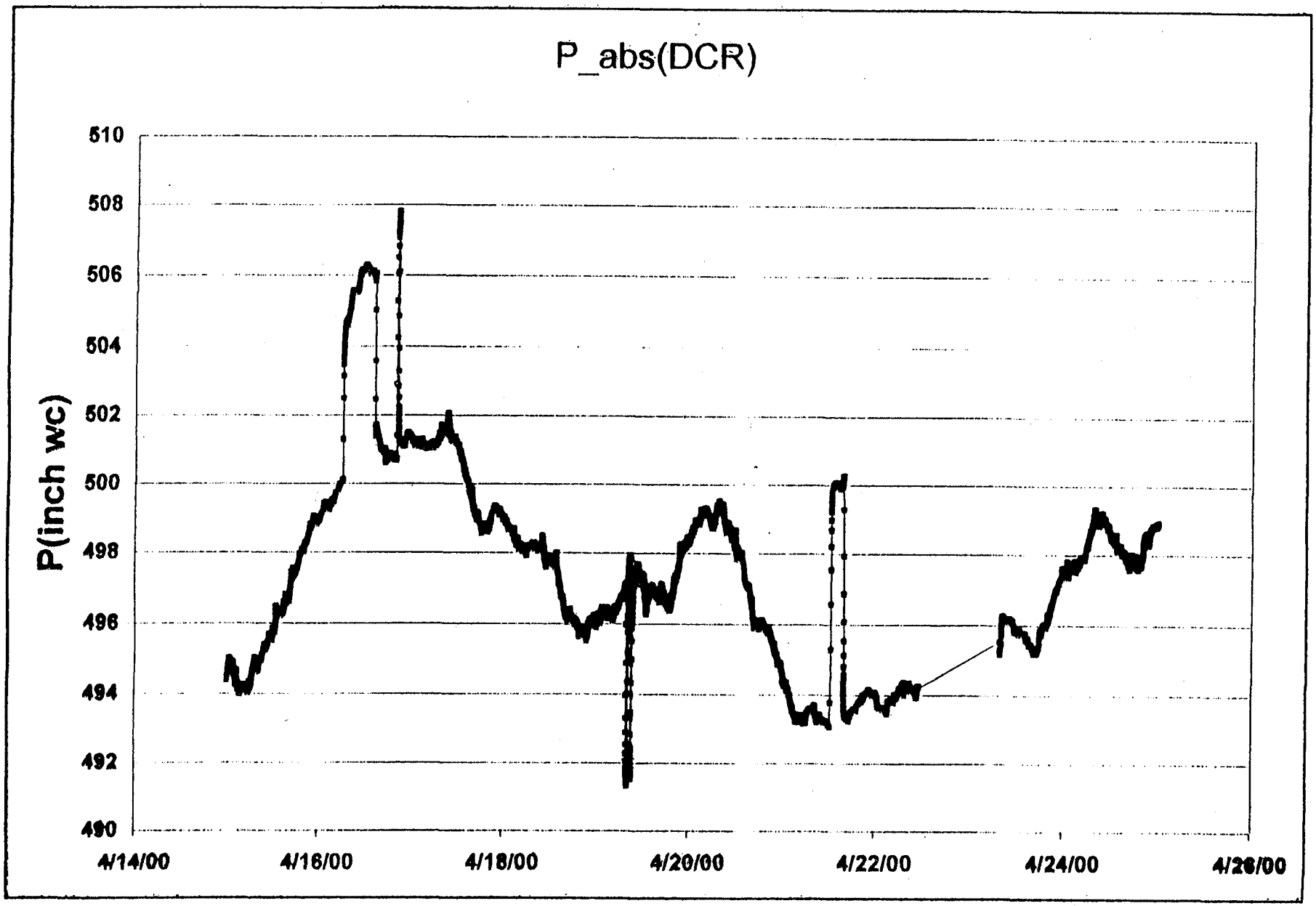
Not shown are dedicated cover gas assay lines.

Not shown is the in-line humidifier for the flush line (necessary to prevent "neck events" - light generated due to flow of extra dry gas through plastic.)

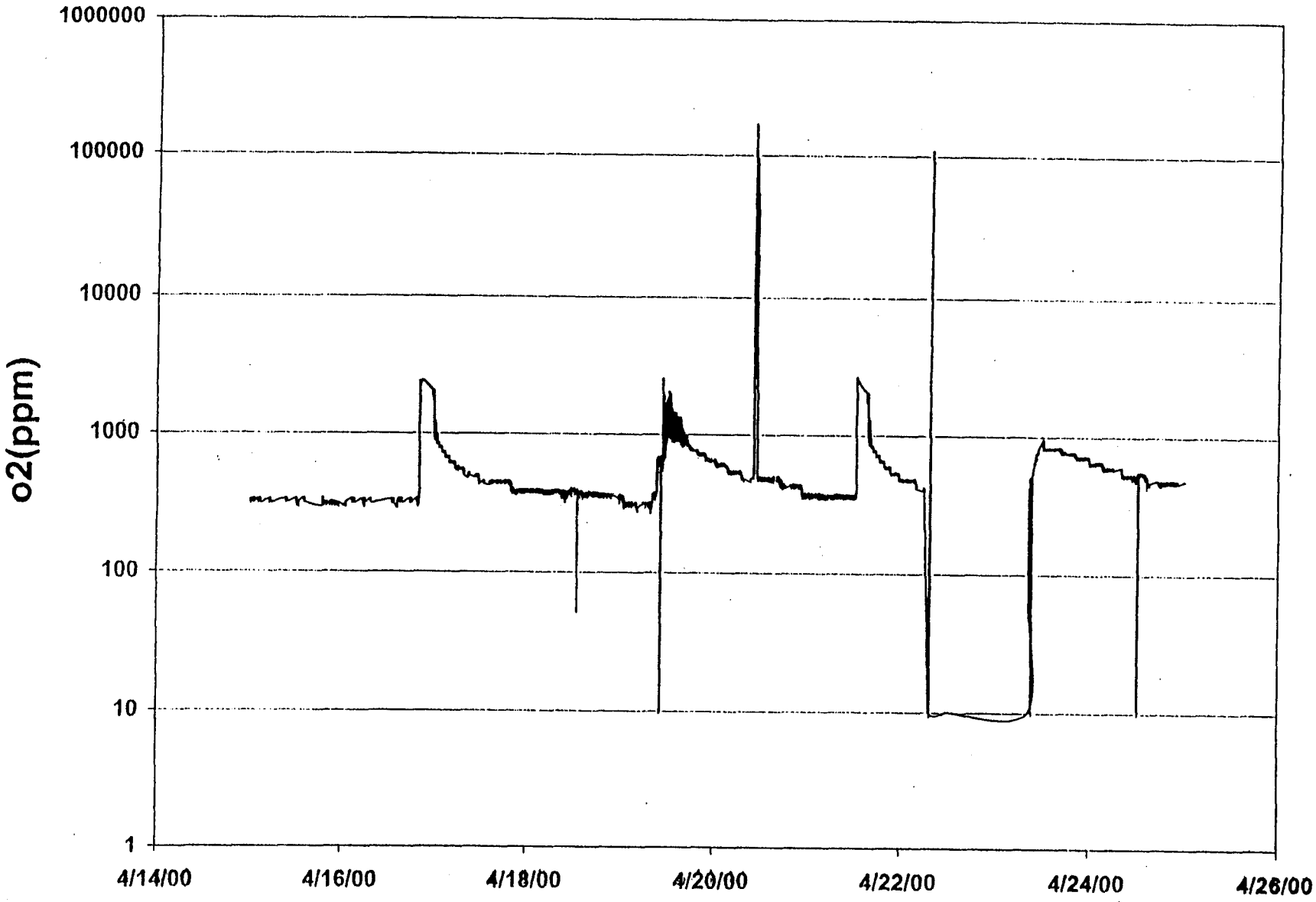


The SNO Cover Gas System.

Lab air Pressure



O_2 content of D_2O cover Gas



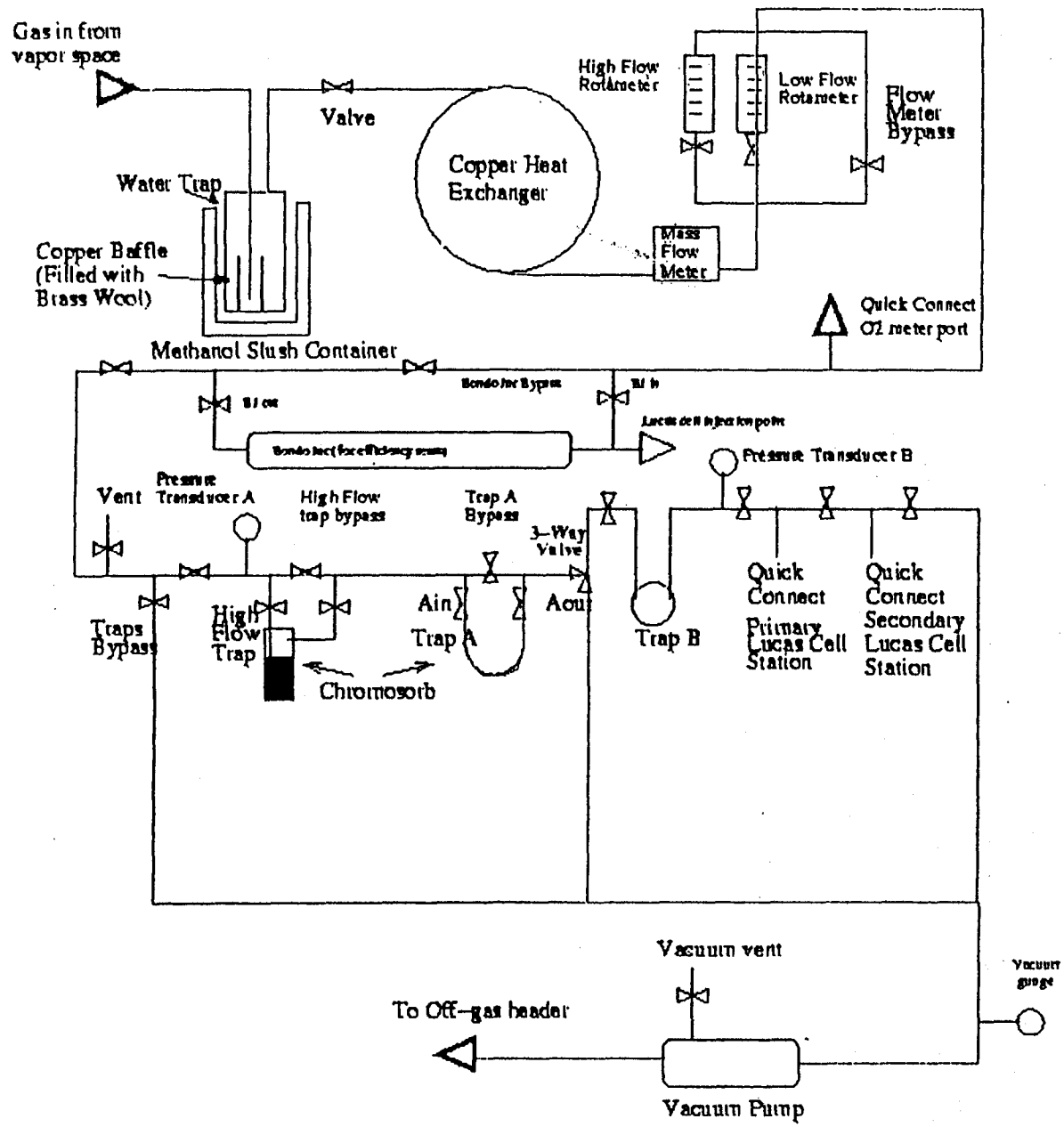
How Radon contamination is measured for the Cover Gas System.

Assay when vapor space is expected to have low Rn concentration*:

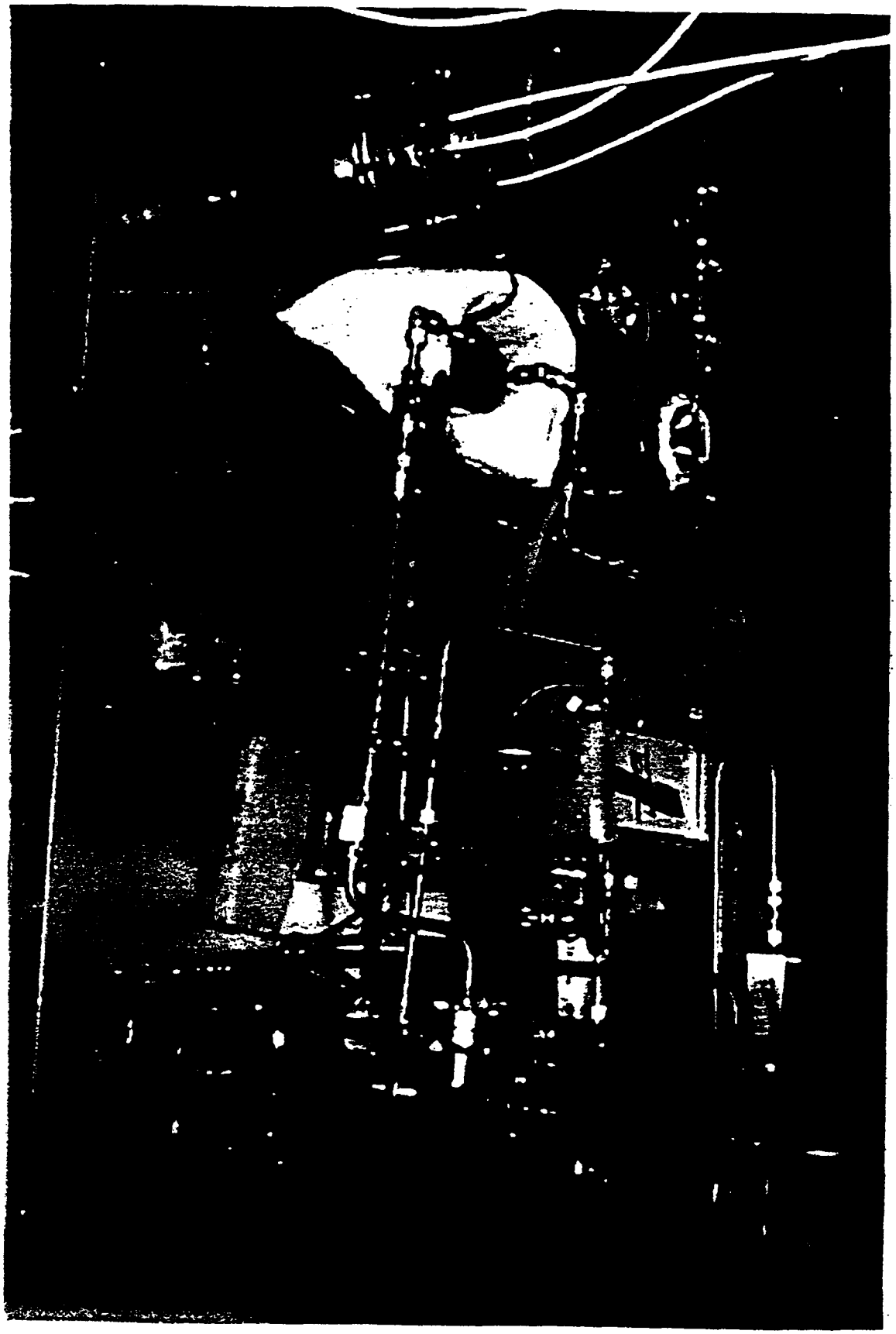
Gas is drawn from the vapor space of interest, using a vacuum pump. Vapor first passes through a methanol-slush cooled (-98°C) water trap to remove water vapor. The gas then goes through a heat exchanger, mass flow meter, and rotameter. The gas then goes through a Chromosorb * filled U-tube (trap A), cooled with liquid nitrogen (-196°C), trapping Rn and CO_2 while most of the O_2 and N_2 are pumped out of the lab. After a sufficient sampling time, the liquid N_2 bath is replaced by a methanol slush bath to remove residual O_2 and N_2. The trap is then connected to a condensing trap (trap B, volume ~2cc) Trap B is cooled with liquid nitrogen, while trap A is heated (+200°C). After 1 hour of cryopumping, trap B is connected to an evacuated Lucas cell (volume ~15.5cc) and radon is transferred to the cell by volume sharing. The radon board has a measured efficiency of about (69±2)%. The 3 quick succession of α decays of ^{222}Rn, ^{218}Po, and ^{214}Po are observed with photomultiplier tubes and, using the volume of gas assayed, are used to infer the amount of radon in the sample gas (^{210}Po which builds up in the cell forms a constant background.) The counting efficiency of this kind of cell has been measured to be about (63±3)%.

*To reduce contamination to the Lucas cell [see poster on Radon assays of Water] with 22-year ^{210}Pb, the cells are filled with uncondensed gas, when radon concentration in a sample is expected to be very high. When intermediate levels of radon are expected, we capture the sample gas in the water trap (1.7 liters) and condense the radon in that gas volume.

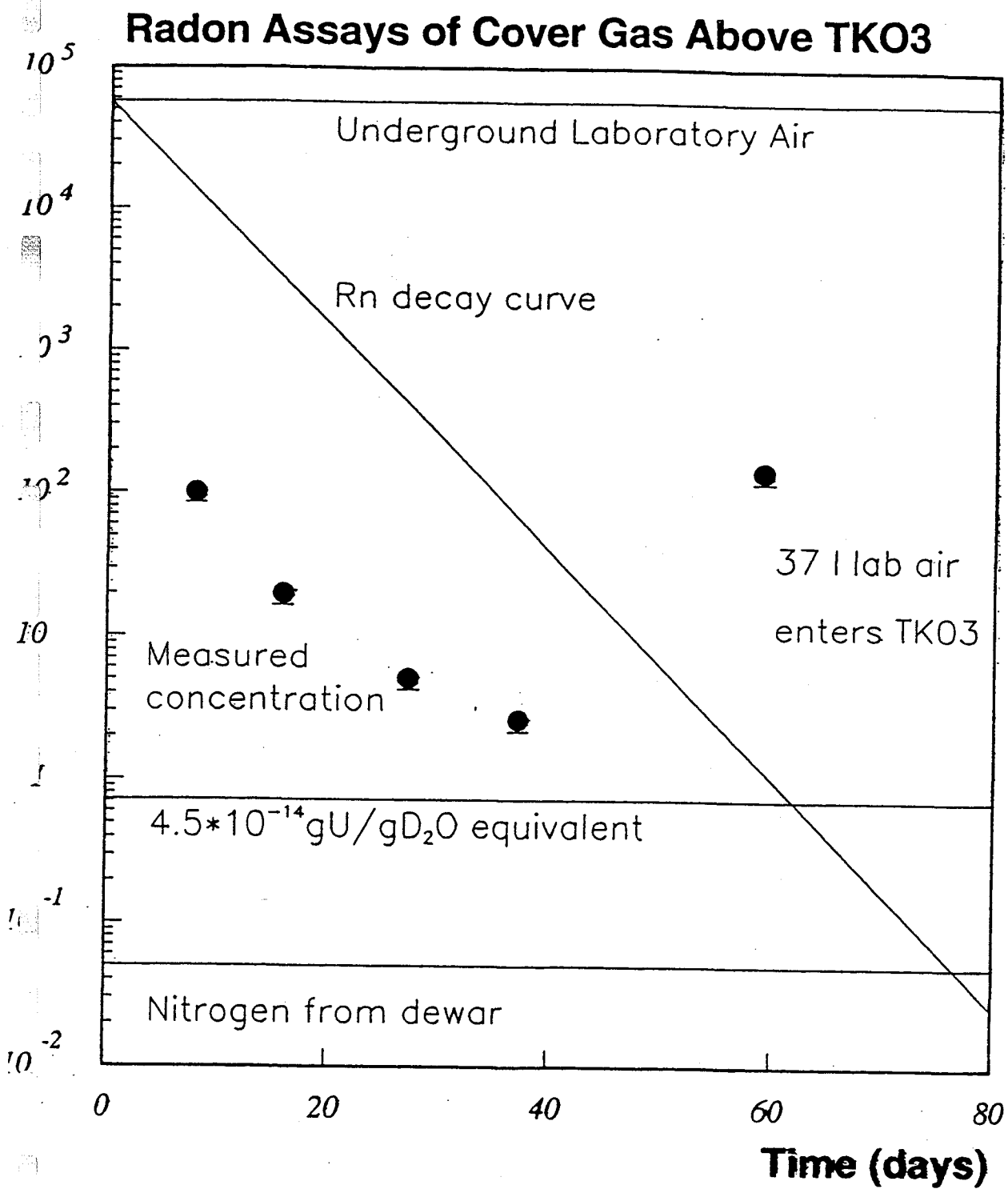
- Chromosorb 102, mesh size 100/120, manufactured by Johns-Manville is a styrene-divinyl benzene polymer with an effective surface area of 300-400m^2/g. This sorbent is the same as that used by the SAGE and GALLEX solar neutrino experiments.



Radon Board for the Cover–Gas System.



SNO Preliminary Report



How Radon is measured for the Cover Gas System II.

The largest contributions to systematic uncertainty in the assays are from measurement of the gas volume sampled and from the Lucas cell counting efficiencies. Work is almost complete on calibrating the cells to the ~5% level. The volumetric calibration will be done next.

Source	Estimated Contribution
Gas Volume (filling cells technique)	3.2
Gas Volume (fixed volume [water trap])	1.7
Gas Volume (flow technique)	15.4
Trap B to Lucas cell	1.7
Lucas cell efficiency	15.0
Cell Background	2.3
Radon Board Background	3.8
Combined (filling cells technique)	**16.1**
Combined (fixed volume [water trap])	**15.8**
Combined (flow technique)	**22.0**

H_2O Cover Gas Radon Assay.

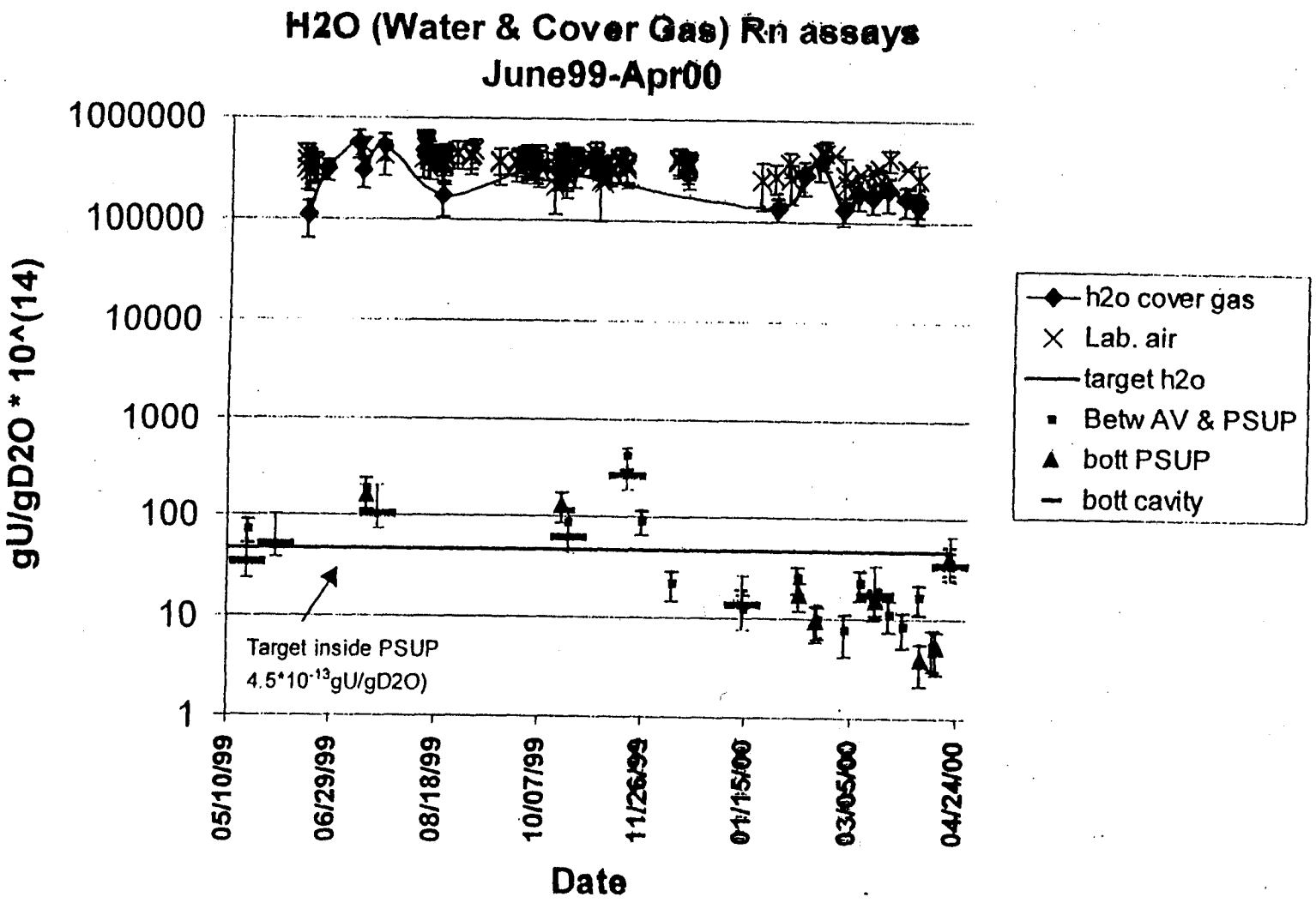
The cover gas data are plotted with both statistical and systematic uncertainties.

H_2O deck needs seal to achieve effective cover gas protection.

All reduction in Rn levels in H_2O due to stagnant thermocline and process degasser.

Water radon levels plotted for several different assay points. Combined statistical and systematic errors shown. See radon in water poster for full presentation.

SNO Preliminary Result



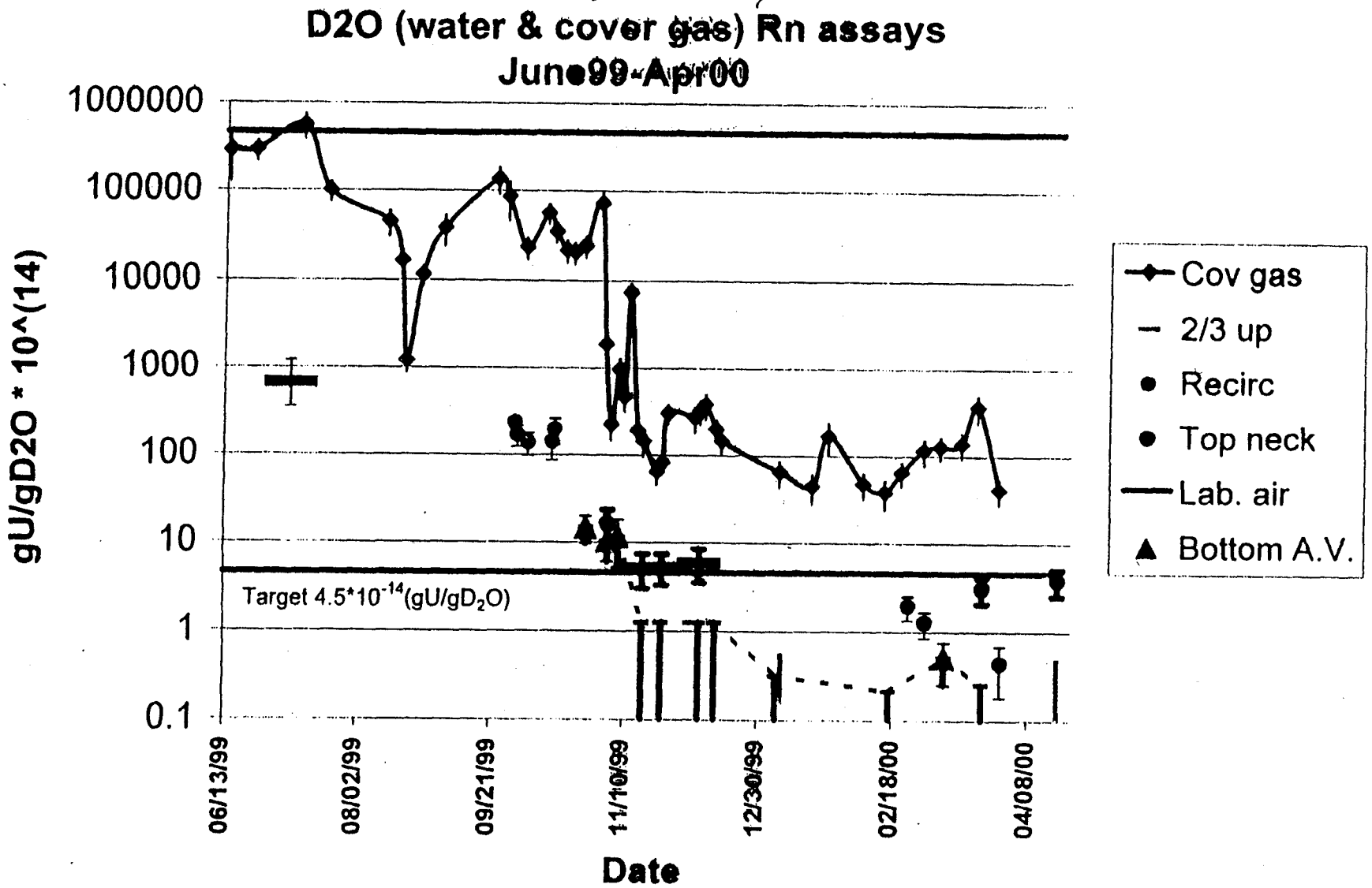
D_2O Cover Gas Radon Assay.

Cover gas data plotted with statistical and estimated systematic uncertainties. The cover gas data target is described assuming secular equilibrium with ^{238}U. The target assumes no benefit of stagnation of the water in the AV neck. Three "dips" in radon levels correspond to flush test, short term flush to test humidification technique, and commencement of large scale humidified flush.

Water radon levels plotted for several different assay points. Combined statistical and systematic errors shown. See radon in water poster for full presentation. Measurements for which only upper limits were derived represented as vertical lines.

The nitrogen flush has been responsible for the dramatic reduction in radon levels above the D_2O and in the D_2O.
Radon in the D_2O can be maintained around target for the neutral current measurement.

SNO preliminary result



Conclusions

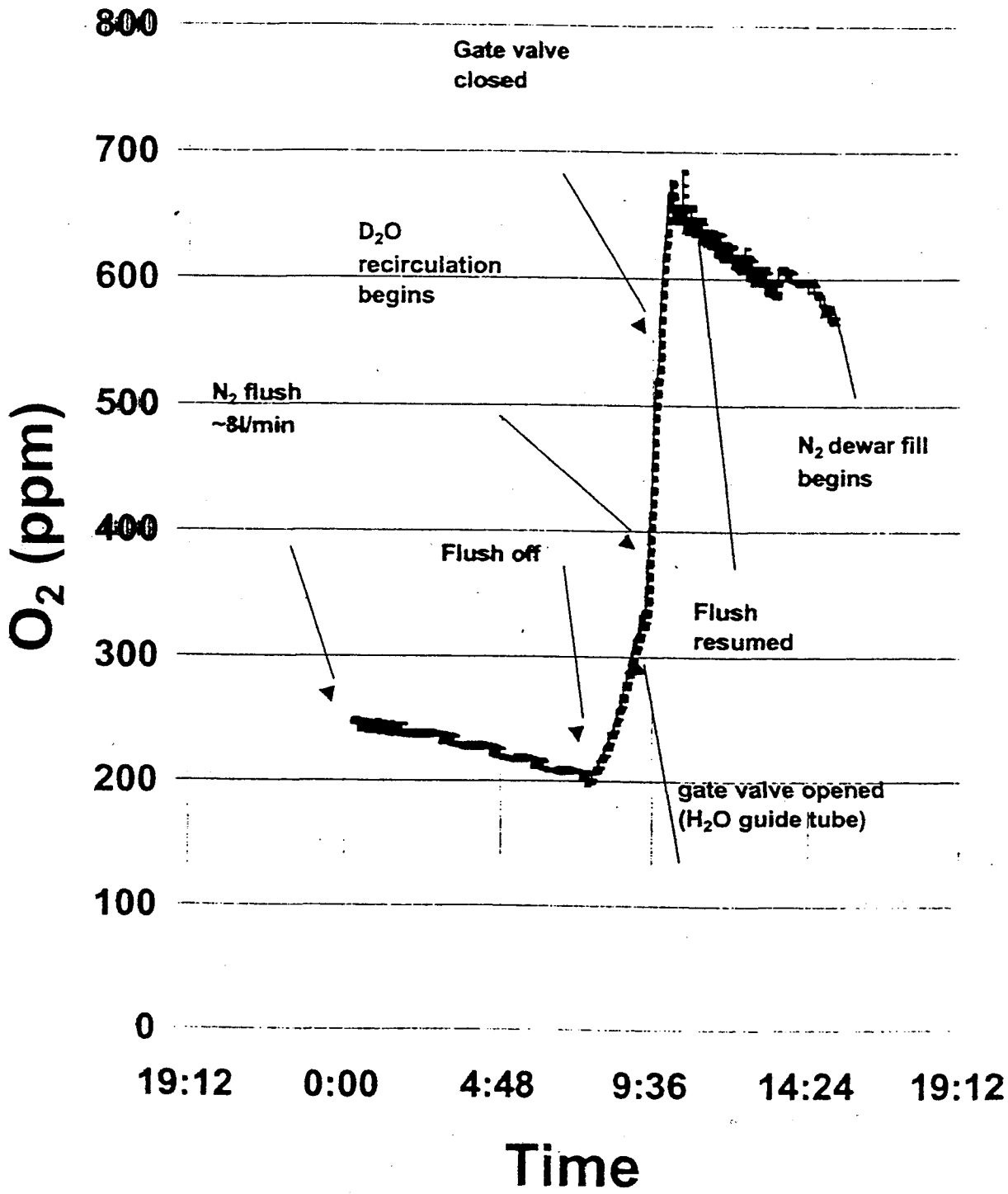
Working to reduce the two largest systematic uncertainties in cover gas radon assays. Lucas cells now undergoing calibration to the ~5% level.

Nitrogen gas supplied by the 1000 liter dewar is sufficiently low in radon to protect the water of SNO. Typically about 0.3 Rn atoms/liter gas (600 $\mu Bq/m^3$ in ^{222}Rn.)

Even with high radon levels above the H_2O, we are around target for radon in H_2O. This is due to a combination of stagnation in the H_2O thermocline and the operation of the light water degasser. Now working to seal the deck to try to reduce radon in H_2O vapor space. Current goal to have H_2O cover gas at least 10 times lower than laboratory air. This may reduce radon in the H_2O comfortably below target.

An active flush of nitrogen gas in the D_2O vapor space has resulted in a dramatic drop in radon levels above the D_2O. The cover gas, in concert with stagnation in the acrylic vessel neck, keeps radon in the fiducial volume at or below target for the neutral current measurement.

6 Dec. '99 D2O Cover Gas



This shows O_2 in D_2O CG when the N_2 flush is stopped. O_2 is a good tracer for Rn.

Radon in the SNO water

H. W. Lee June 2 2K

The underground SNO lab is "dust-free" (Class 10^4 clean-room), the personnel are squeaky clean, the detector components are low radioactivity material and cleaned meticulously, but the environment is "hissing" with radon.

- **Rock Walls**

  (ore body 1.4% Ni 1.4% Cu, 1.8gm/tonne Pt, Pd, R

  (40°C at 6800 ft. because of U, Th

  52 + 47 + 43 MeV Nuclear Geology H. Faul

  $0.015 \frac{Bq}{m^2 sec}$ ⇒ Use Urylon on the walls (J. Farine)

- **Air**

  22°C 45–55% rel. humidity

  110 – 330 Bq/m^3. (week, daily variation → P. Jagam hourly measurem

  I. Levine → cover gas.

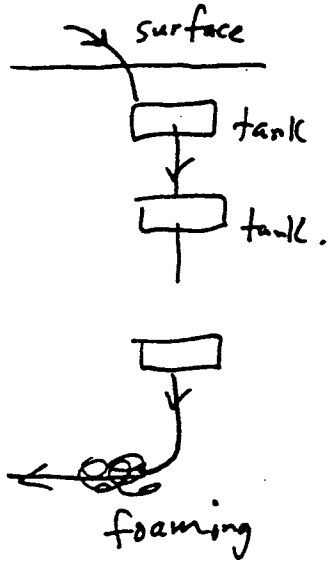
- <u>Water</u>

H_2O

Vermillion River (fishing)

water treatment filtered

chlorinated — E. coli free!



0.02 Bq/l.

D_2O

Bruce Heavy Water Plant

Stored and transfered without cover gas.

SNO storage is sealed but not 100% leak tight tanks

Went into acrylic vessel that was full of mine air. (plus pressure swings)

Radon in the SNO water

H. W. Lee June 2 2K

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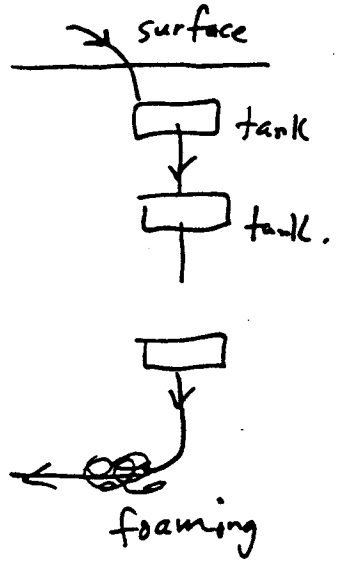


0.02 Bq/l.

D_2O

Bruce Heavy Water Plant

Stored and transfered without cover gas.

SNO storage is sealed but not 100% leak tight tanks

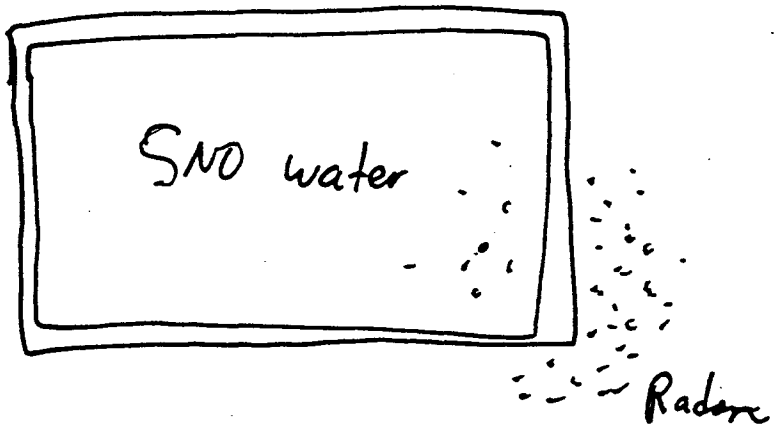
Went into acrylic Vessel that was full of mine air. (plus pressure swings)

SNO Targets

H_2O 6μ Bq/l.

D_2O 0.6μ Bq/l

Note. $0.6\mu Bq/l \Longrightarrow 8\times10^{-27}\ \frac{\text{Rn atoms}}{\text{water}}$!



Internal Sources

- ^{226}Ra

  Removed by water purification to levels below 1.5 μ Bq/l

  (J. Farine poster)

- Emanation

  Submersed materials contain some radium so it "emanates" radon.

  → glass, PMT bases, cables, plastic, Urylon liner, pumps, ...

  We have measured emanation rates of materials (Rn atoms/m^2 hr)

  Identified polypropylene 10 times better than regular grade, O-rings that are 10 times better, ..

External

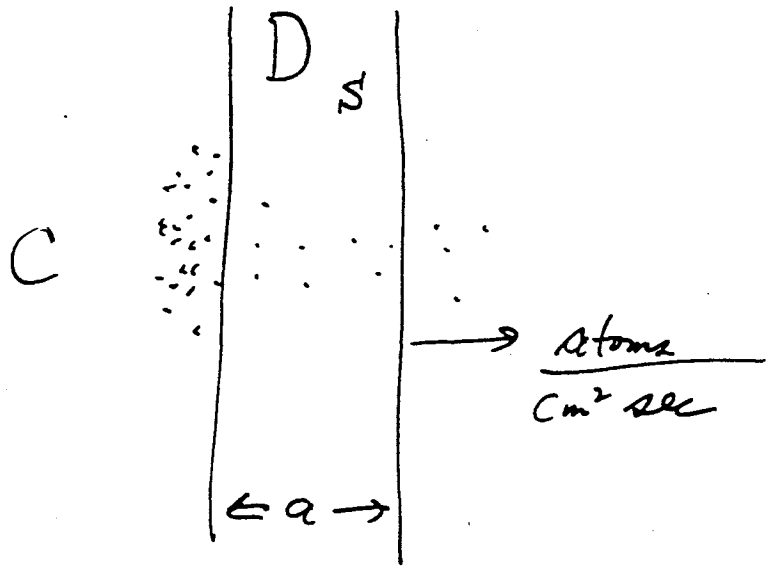
- Leaks

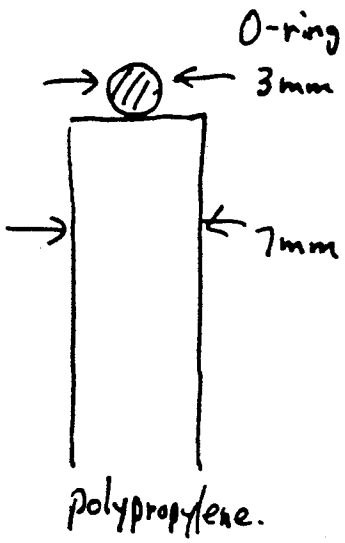
A 10^{-3} cc/sec air leak into a water flow of 20 lpm makes it have a radon concentration of 0.4 μBq/l.

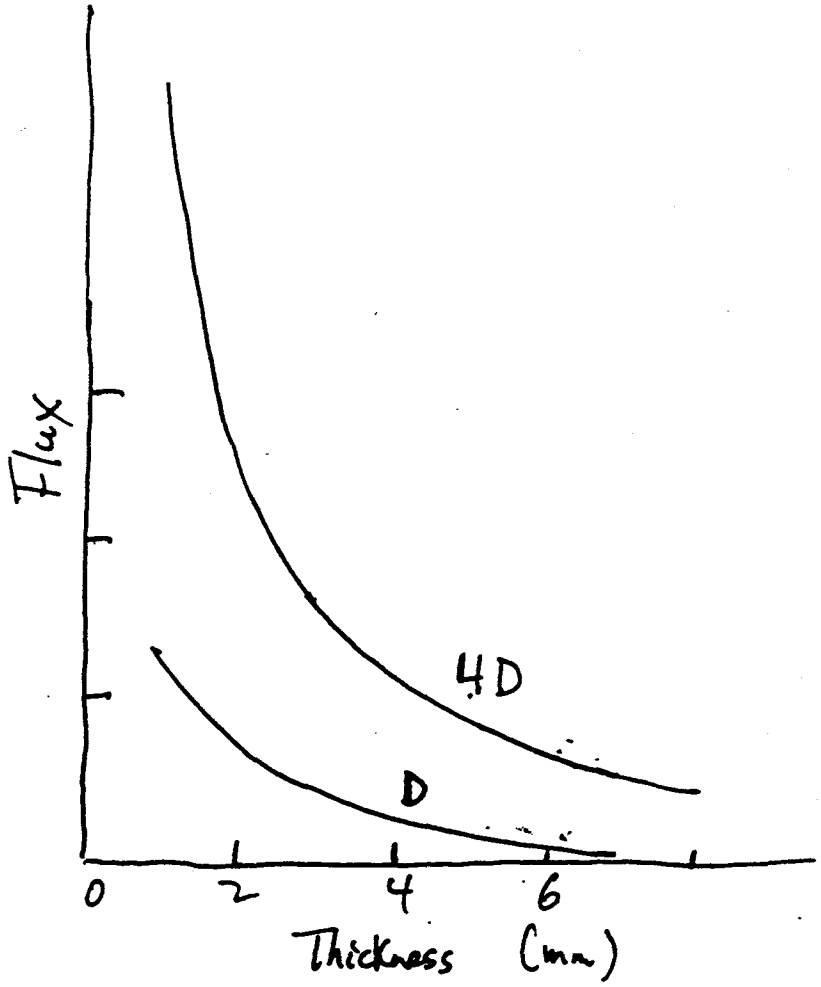
(or 10 leaks of 10^{-4} cc/sec each...)

Extensive Helium leak checking of entire piping system, values, gauges, ...
(unable to do tanks)

- Diffusion



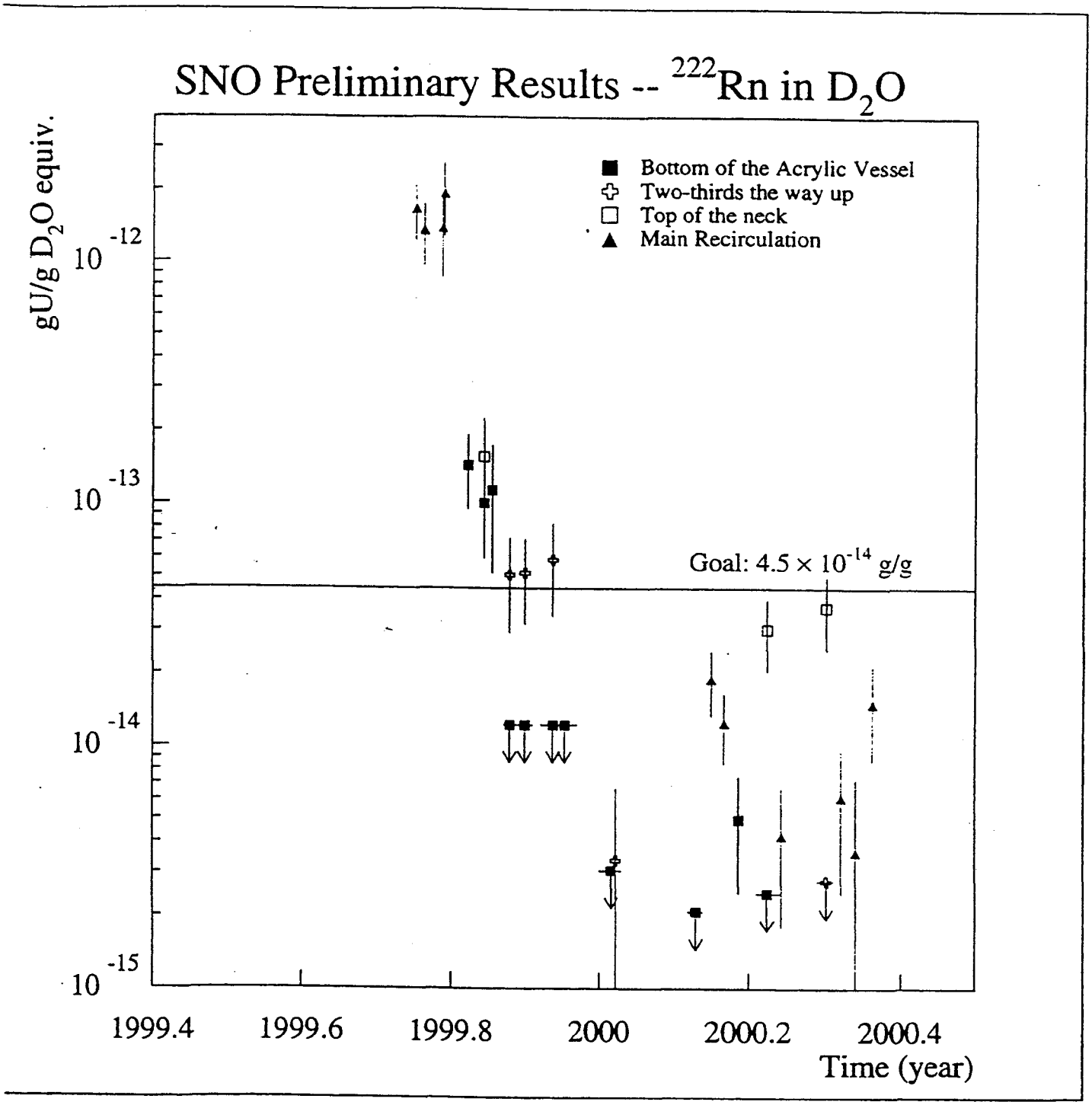


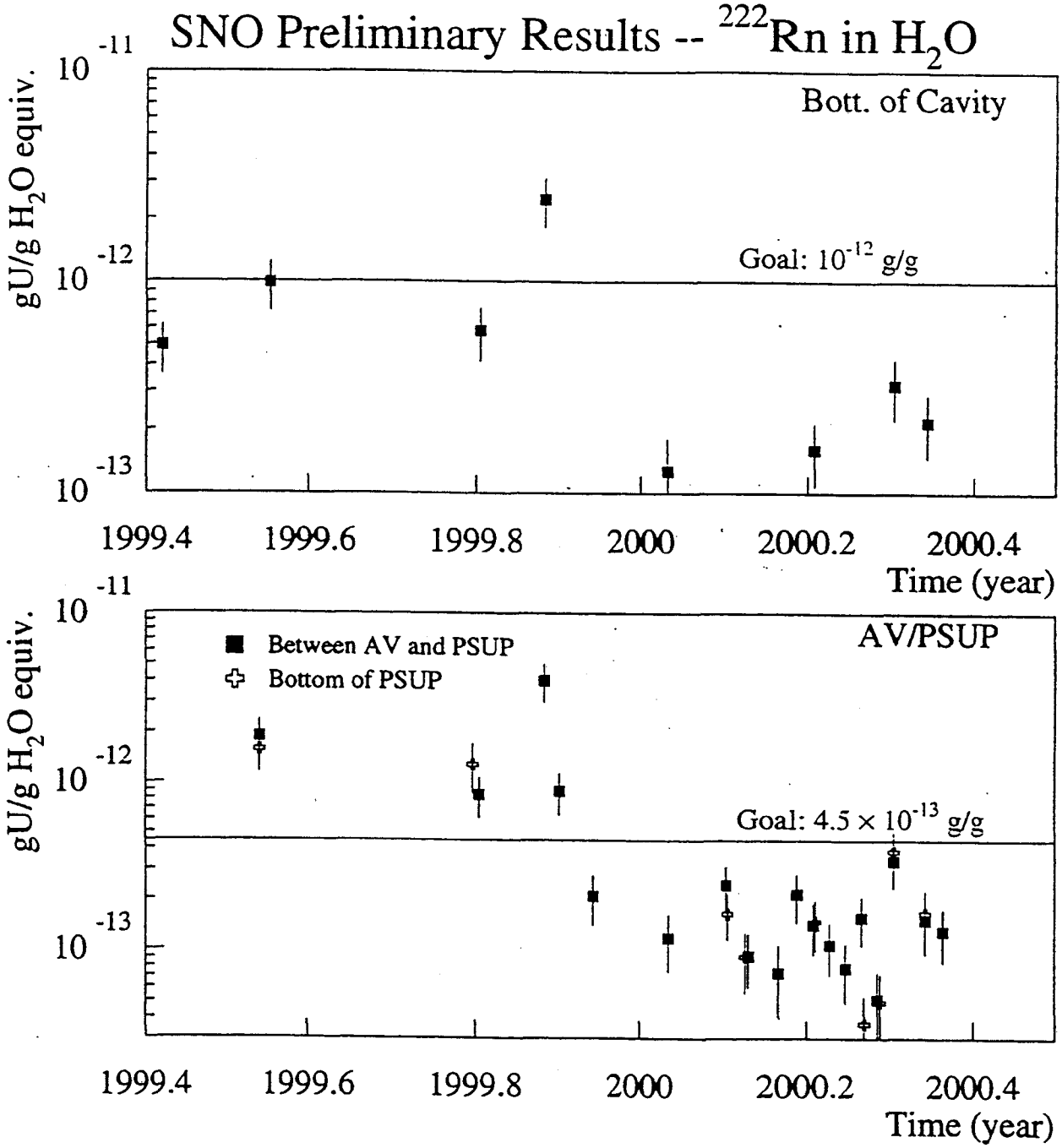


→ Need good O-rings (small D)

Time Dependent Sources?

- Very sensitive to air leaks → a loose nut or value can cause radon background to increase 20-200 times
- O-rings acquire a set/fatigue → increase diffusion rate.


SNO Preliminary Results -- ^{222}Rn in D_2O
gU/g D_2O equiv.
10^{-12}
10^{-13}
10^{-14}
10^{-15}
Bottom of the Acrylic Vessel
Two-thirds the way up
Top of the neck
Main Recirculation
Goal: 4.5×10^{-14} g/g
1999.4
1999.6
1999.8
2000
2000.2
2000.4
Time (year)


SNO Preliminary Results -- ^{222}Rn in H_2O
Bott. of Cavity
Goal: 10^{-12} g/g
gU/g H_2O equiv.
10^{-11}
10^{-12}
10^{-13}
1999.4
1999.6
1999.8
2000
2000.2
2000.4
Time (year)
AV/PSUP
Between AV and PSUP
Bottom of PSUP
Goal: 4.5×10^{-13} g/g
gU/g H_2O equiv.
10^{-11}
10^{-12}
10^{-13}
1999.4
1999.6
1999.8
2000
2000.2
2000.4
Time (year)

Dersch Appl Rad Isotopes 49, 1171 (1998)

Diffusion

7

Mueller 1978 Heidelberg Thesis

Wojcik Radon diffusion and solubility constants
Nucl. Instrum Meth B61, 8 (1991)

J. Bigu, E.D. Hallman and L. Kendrick
Permeability of different materials to radon (222Rn) s
SNO-STR-91-069

J. Bigu and E.D. Hallman
Emanation and permeability studies at the Elliot Lake
SNO-STR-92-018

J. Bigu and D. Hallman
Radon Emanation from materials SNO-STR-92-037

In the Environment

Oliveira Appl. Rad Isotopes 49, 423 (1998)
spring water 222Rn 7 Bq/l 226Ra 4 mBq/l

Al-Masri Appl. Rad Isotopes 50, 1137 (1999)
lake water 222Rn mBq/l 226Ra 6 mBq/l

Cannizzaro Appl. Rad Isotopes 51, 239 (1999)
201Pb in the air 500 microBq/cubic meter

Kuo Appl. Rad Isotopes 48, 1245 (1997)
226Ra in food and water water 226Ra 3 mBq/lit
chicken 0.17 Bq/kg fish 0.04 Bq/kg
pork 0.07 Bq/kg vegetables 0.04 Bq/kg

Man Appl. Rad Isotopes 50, 1131 (1999)
indoor radon in apartment building 170 Bq/cubic mete:
outdoor 30 Bq/cubic meter

Sanchez Appl. Rad Isotopes 50, 1049 (1999)
226Ra in bottled mineral water 0.3 Bq/liter

Martino Appl. Rad Isotopes 49, 49\07 (1999)
radon emanation and exhalation rates from soils

RADON BACKGROUND IN RARE-EVENT EXPERIMENTS

Satellite workshop of 'NEUTRINO 2000'

Radon at SNO

Part III

Jacques Farine

Carleton University, Ottawa

SUDBURY, Canada
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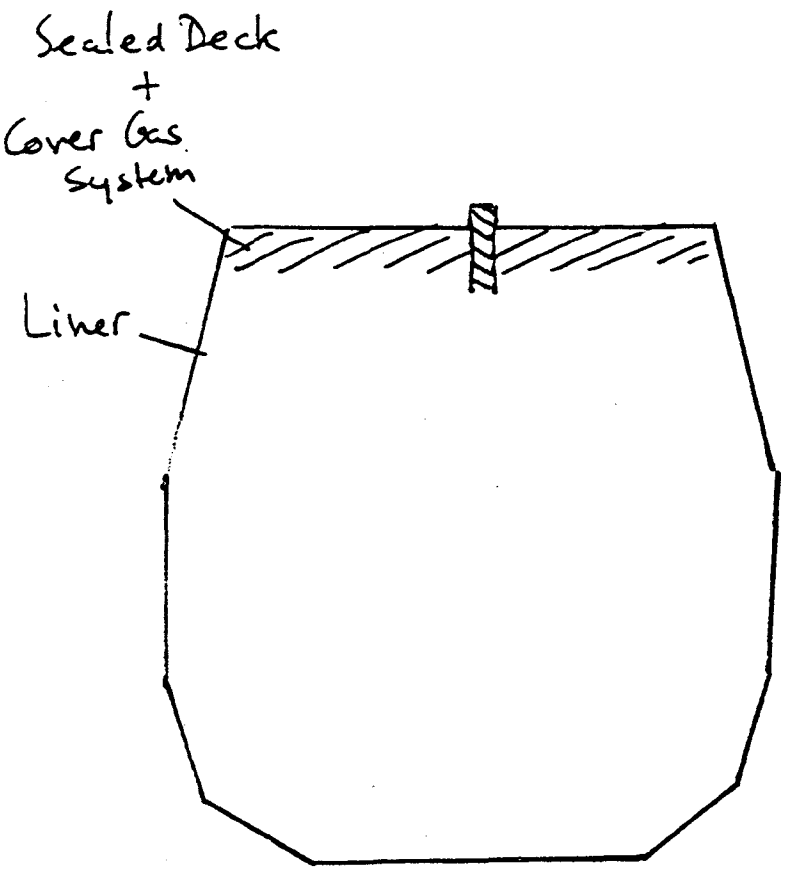
PLAN

1. Radon barriers
   - Cavity liner
   - O-rings
2. Detection of ^{220}Rn
   - Need to assay ^{224}Ra
   - The MnO_x Radium Assay technique
   - The SNO ElectroStatic Counters
   - Efficiencies
   - Sensitivity
   - Results $\longrightarrow$ Hit R. Donald on Friday

Radon suppression in

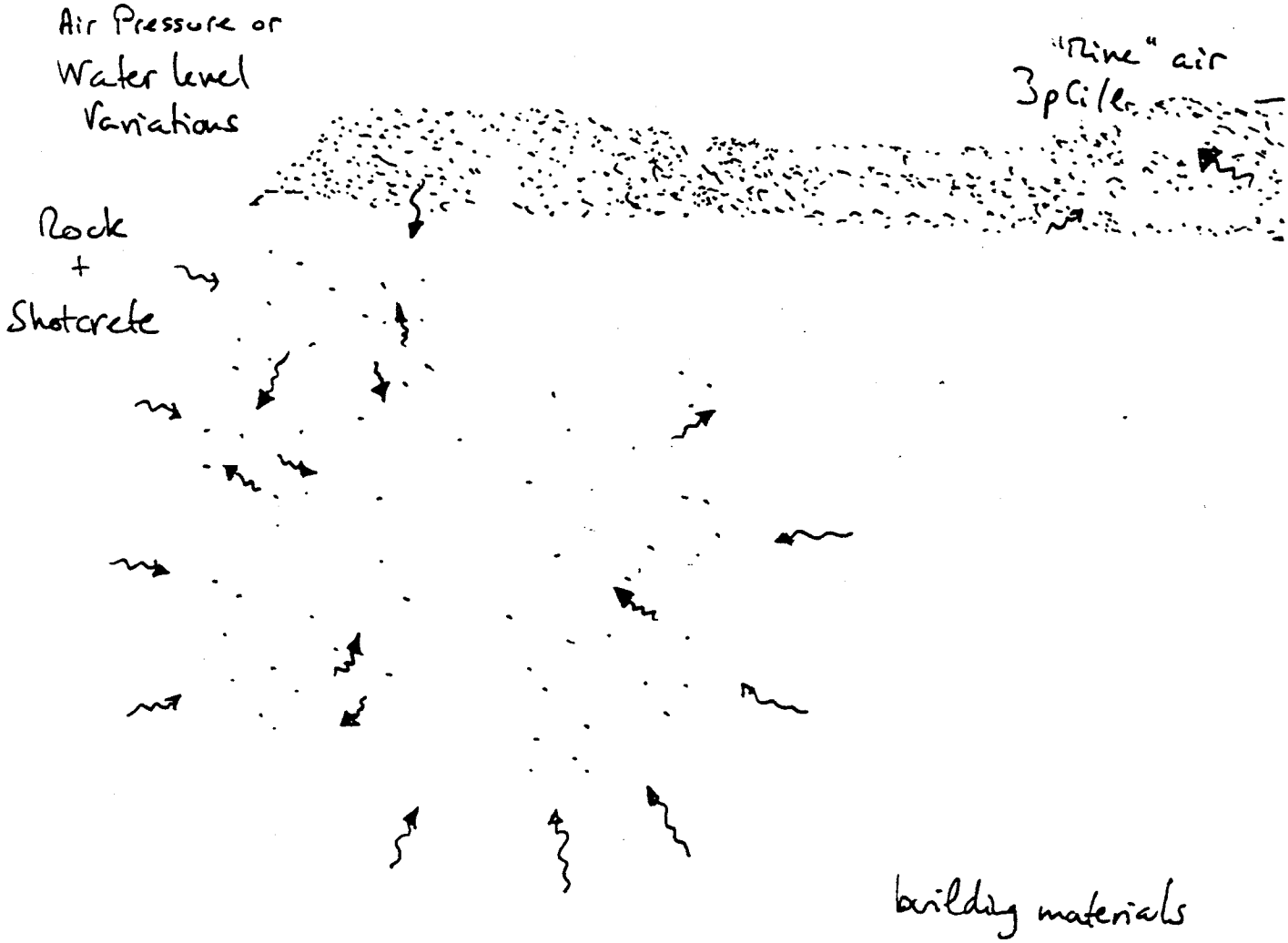


O-ring, piping

⇒ reduce
Rn diffus.

⇒ water
purification
(in addition to low activity
materials selection)

Radon sources and ingress into



The SNO Detector

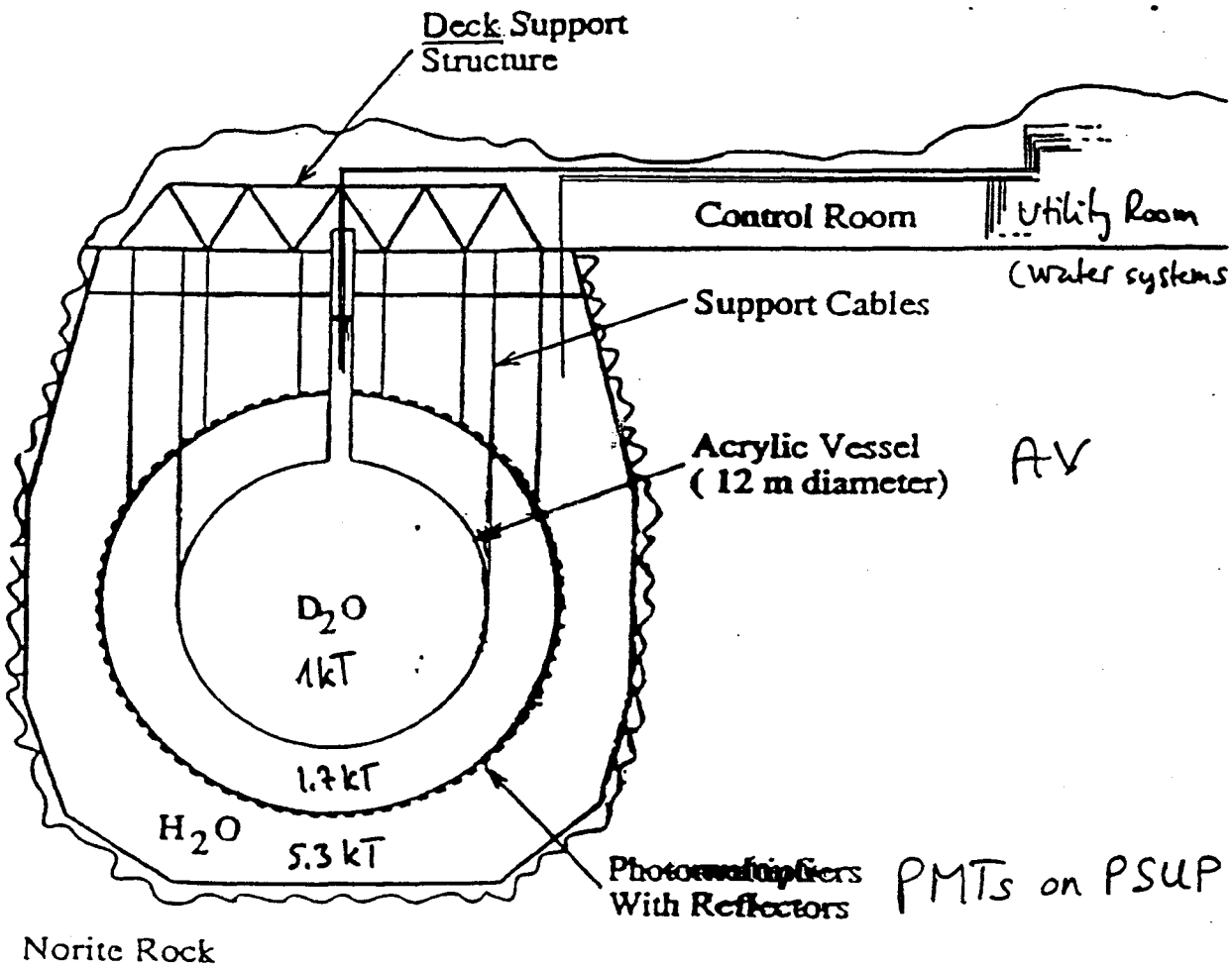


Figure 1.11: The SNO detector

Radon diffusion

Steady-state, one dimensional <u>diffusion equation</u> with decay and source term:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - \frac{C}{\tau} + \varphi = \phi$$

where : C : concentration of Rn at position x (time t)

D : diffusion constant in medium

τ : lifetime ($^{222}Rn = 5.5\,d$)

φ : source term in medium (indep. x)

General solution

$$C(x) = A e^{x/d} + B e^{-x/d} + \varphi \tau$$

A, B from boundary conditions.

define $d := \sqrt{D\tau}$ as "diffusion length".

Application

Solve it for multilayer system

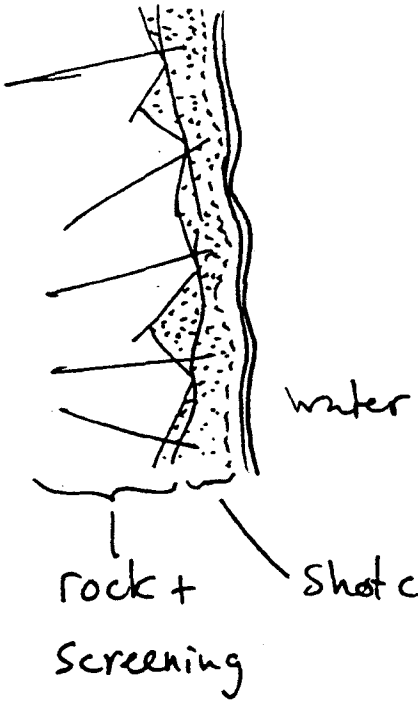
Radon ingress : $j = -D \frac{\partial C}{\partial x}$ at interface

define by $C = S \cdot p$ <u>S = "solubility"</u>, p pressure

→ well-known relation $j = -SD \frac{\partial p}{\partial x}$

<u>k</u> $= SD$ <u>permeability</u>

Cavity Liner



goal: H_2O @ max 10^{-12} eq. gU/g

cavity area: 2000 m^2

water volume: 5800 m^3

$\Rightarrow$ max Rn ingress is $\frac{1}{10}$ a_{water}:

$$\underline{\underline{13 \ m^{-2} h^{-1}}}$$

Cannot be measured! [seal, materials, ...]

measured: shot crete emanation: $1.9 \cdot 10^7 \ m^{-2} h^{-1}$

need reduction factor 10^6 ! (or 14 · d thickness)

"MIROC" or "RINE GUARD" standards used in mines:

polyurethane, hard, show $d \lesssim 0{,}05$ cm

$\Rightarrow$ liner of thickness 9 mm ($\sim 18 \cdot d$)

- applied in 9 layers of different colors
  $\Rightarrow$ visual Quality Control

<u>Rn diffusion through origs</u>

- DEFINE A CLOSED LOOP
- Extract Rn ingress for ≠ modes (VAC, WAT)
  $\rightarrow$ <u>VAC</u>

	urethane	Butyl
Calculated Rn ingress (at/day):	1600	32
Observed:		321 ± 39 . still, leaks

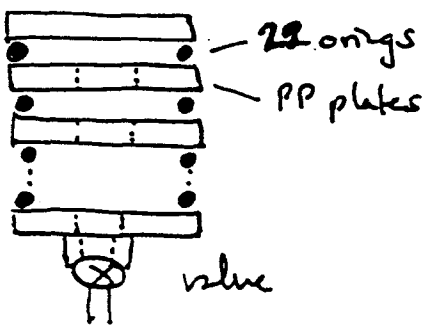
Radon diffusion through o-rings

urethane o-rings were choosen

- low Rn emanation rate : 2.4 Rn / day · meter
- good mechanical properties → proper seals
- low leaching into water

U/G : see high level of Rn

→ build "Oring stack" – assemble
– leak check (He)
– seal : Rn from air diffuses in
– extract Rn daily → >50 $h^{-1}m^{-1}$



→ high $D = 2.8 \cdot 10^{-7}$ cm^2/sec

⇒ contacted
- manufacturers
- Borexino
- Gert Heusser

→ move to butyl rubber
$D = 4.9 \cdot 10^{-9}$ cm^2/sec
also leak! ~ 0.1 Rn/m·h
(urethane < 0.2)

~~Currently exchanging~~ all 7000 orings on water systems.

→ Done!

only ~1/2, i.e. 4000 orings, were critical.

in situ comparison : diffusion through butyl ~0.15 Rn/m·h

BACKGROUNDS TO THE NC REACTION IN SNO

Anticipated rate of NC interactions in SNO (BP98)= 12.6/day

Require that BGND lower or equal to 1n/d

Main BGND: photodissociation of deuteron $\gamma + d \rightarrow p + n$

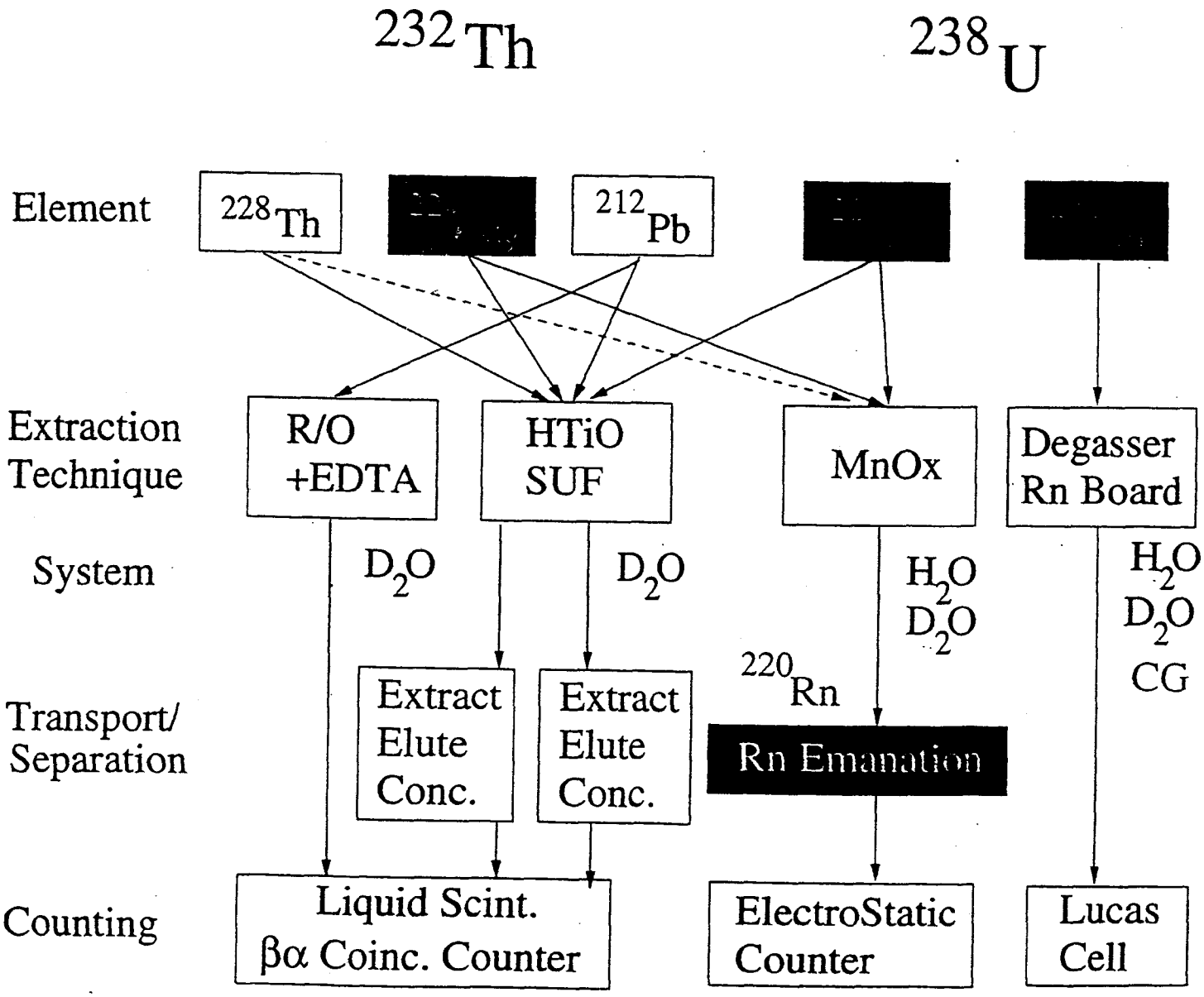
Threshold = 2.2 MeV

Gamma emitters in natural decay chains:	^{214}Bi	^{208}Tl
Energy:	2.4 MeV	2.6 MeV
Number of gamma to produce one neutron	750	470

This implies most stringent limits are in D_2O with:

Concentration	3.7 x 10E−15 gTh/g	4.5 x 10E−14 gU/g
atoms/ton	7	3.8 x 10E7
Decays per day per ton	1.3	45
Bq/kg	1.5 x 10E−8	5.2 x 10E−7

Radioactivity Monitoring at SNO

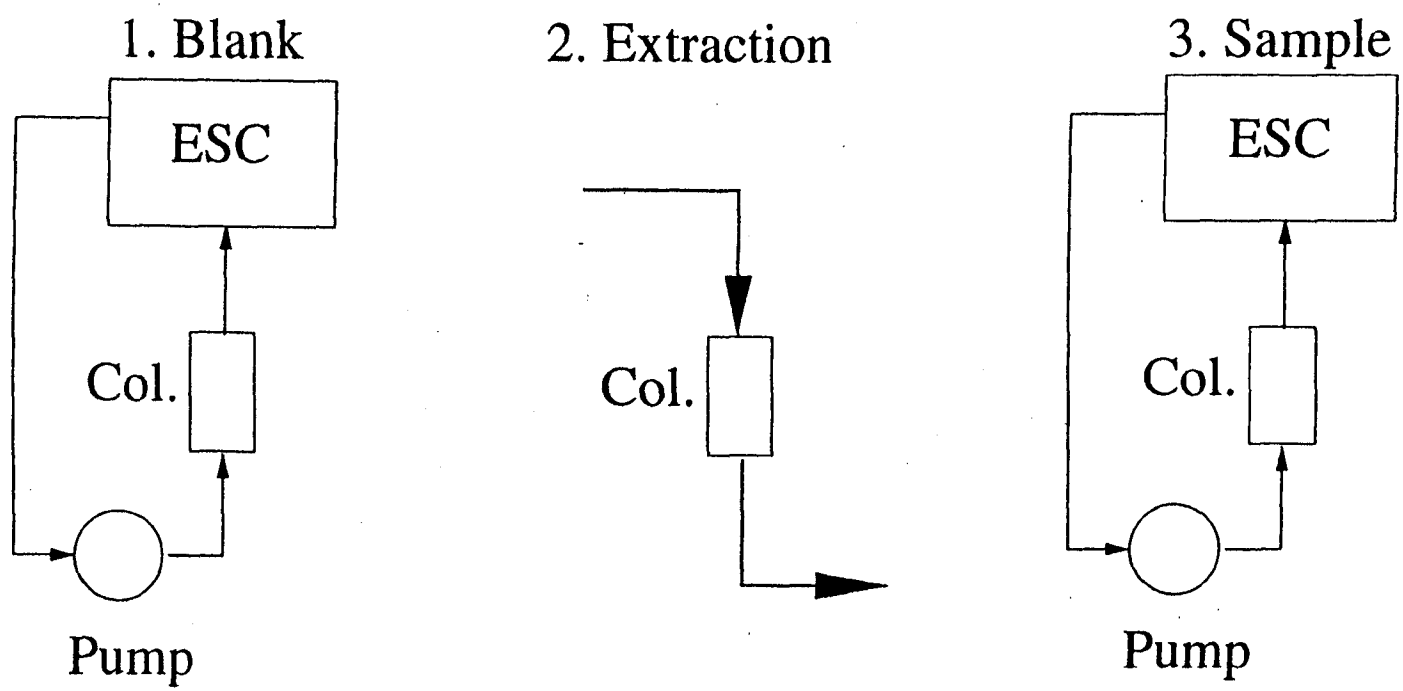


THE MnOx RADIUM ASSAY TECHNIQUE

Radium is extracted on MnOx–coated acrylic beads (spheres)

high affinity for Ra

1. A 1L column is filled with beads and counted on an ESC (Blank)
2. The column is then exposed to water. Typical sample mass = 50T
3. The column is counted again (Sample)
4. The difference Sample–Blank gives the net signal

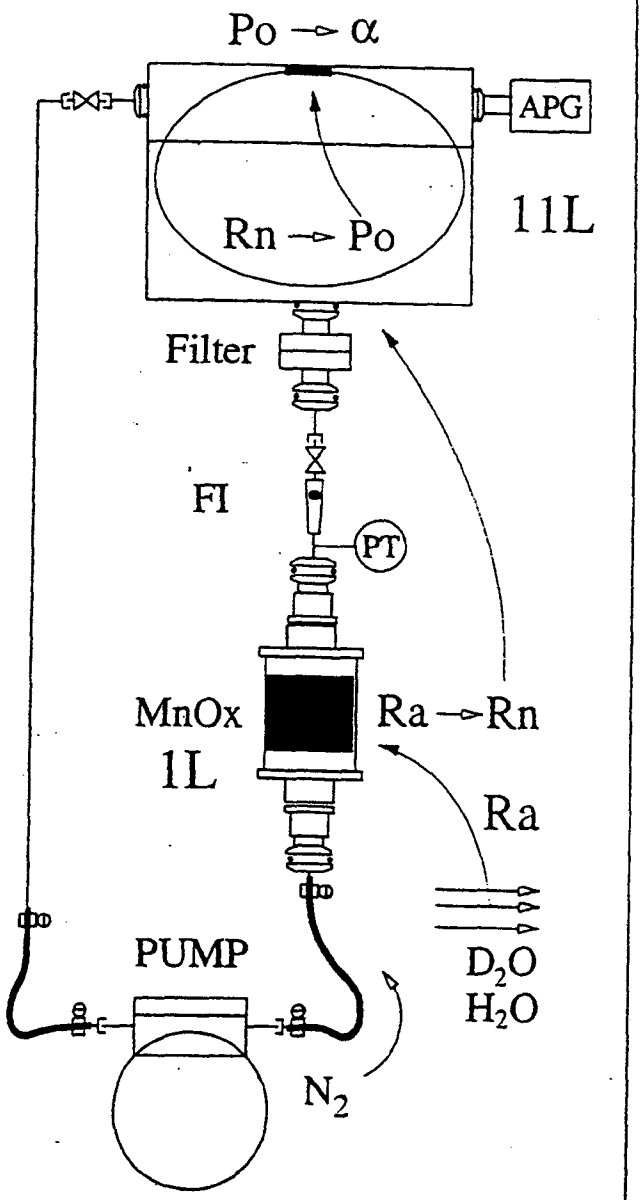


DAQ: ElectroStatic Counter (ESC)

NIM A 421 (1999) 601–609

ESC + recirculation loop

- Ra decays, Rn carried to ESC
- Rn decays, 70% Po daughters charged
- Charged Po collect on Si–PIN diode
- α spectroscopy of Po daughters



8 counters operating on site

Efficiencies	Th	U
$\varepsilon_{detection}$	22%	35%
$= \varepsilon_{geometry}$	50%	50%
$\times\ \varepsilon_{collection}$	71%	100%
$\times\ \varepsilon_{Q}$	70%	70%
ε_{volume}	100%	95%
$\varepsilon_{transport}$	95%	100%
$\varepsilon_{emanation}$	30%	70%
$\varepsilon_{extraction}$	90%	90%
ε_{TOTAL}	6%	22%

MnOx Assay – Analysis

- 4 Energy windows
- coupled through
  - μ lines (^{212}Bi)
  - overlaps (spike-calib'd)
- fit all data w/ single LF
- outputs amplitudes

E_1 E_2 E_3 E_4

Single likelihood fct

^{228}Th ^{224}Ra ^{226}Ra ^{222}Rn

1 spectrum every 3h

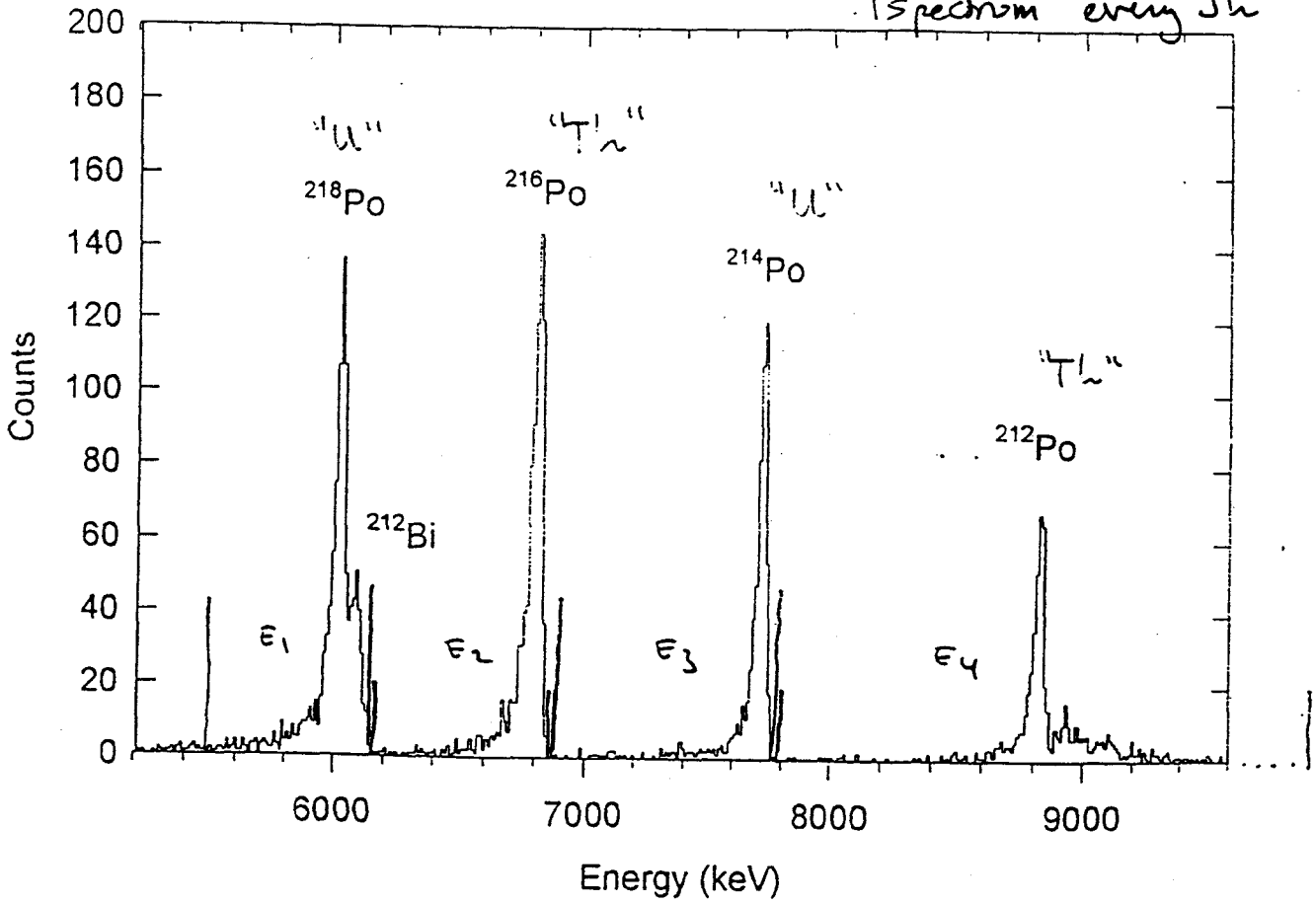


Figure 3

<u>Assume</u> :

- observed activity curves originate from :

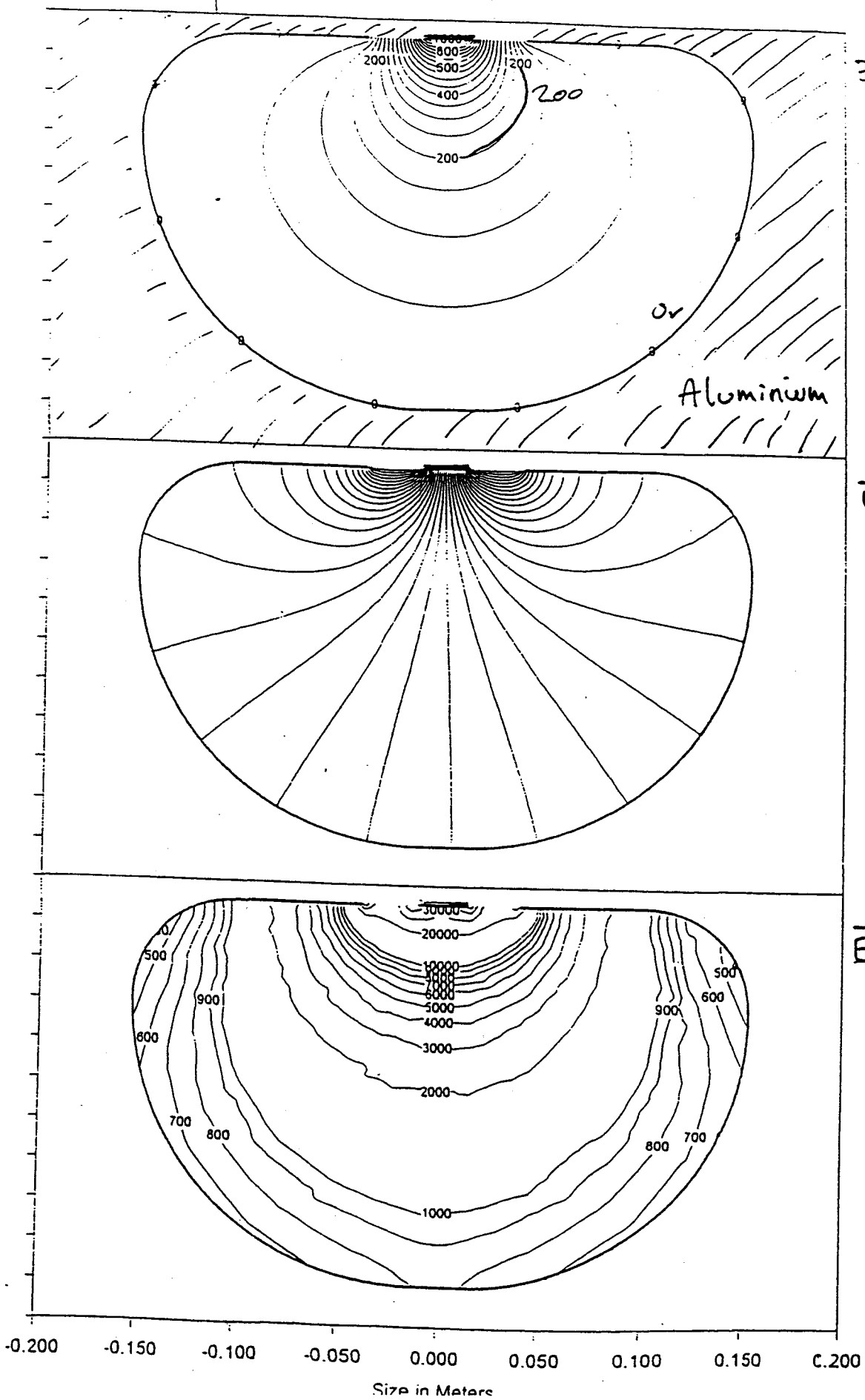
  Th : ^{228}Th ^{224}Ra

  U : ^{226}Ra ^{222}Rn

  "supported" "unsupp."

- blank : [illegible]

Diode 1000V ESC Chamber



Equi Pot.

11 liters

$\vec{E}$ Field Lines

$\vec{E}$ Field Strength

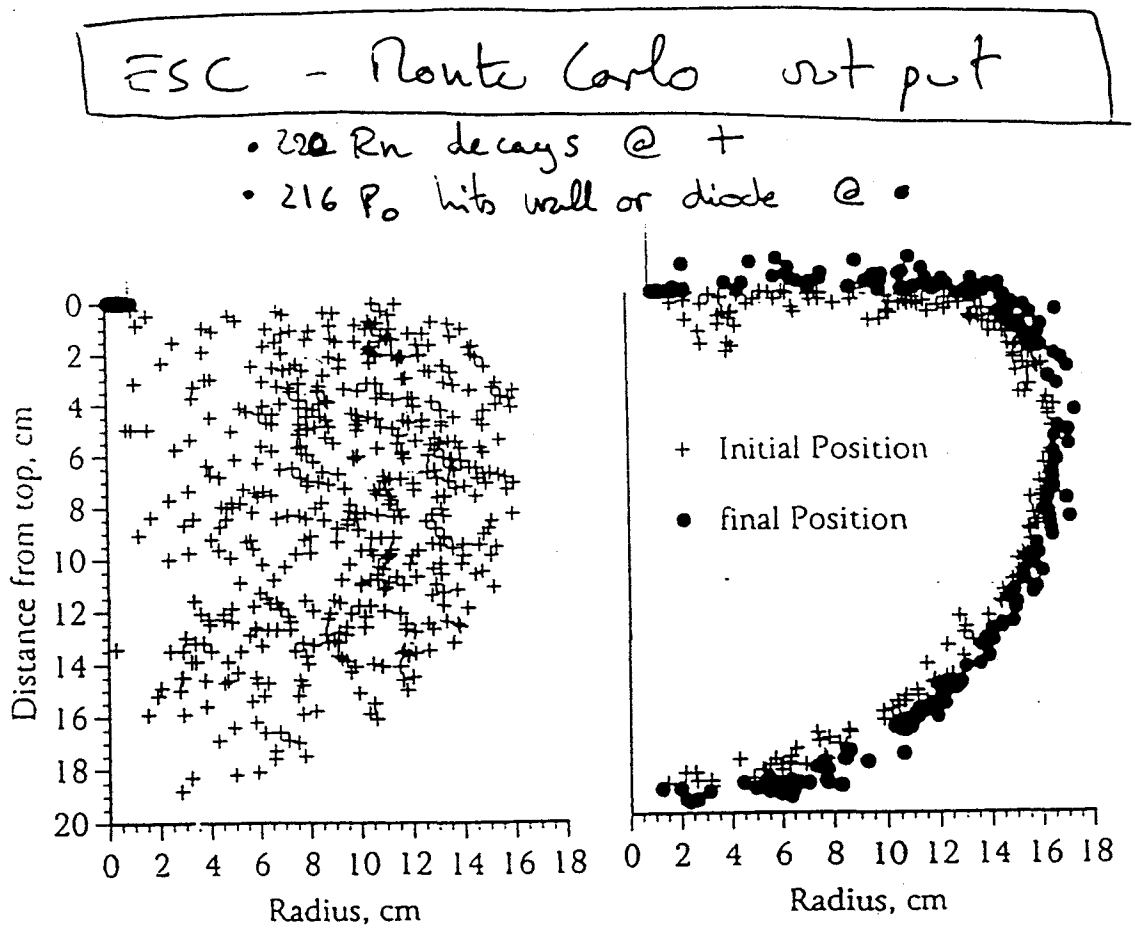
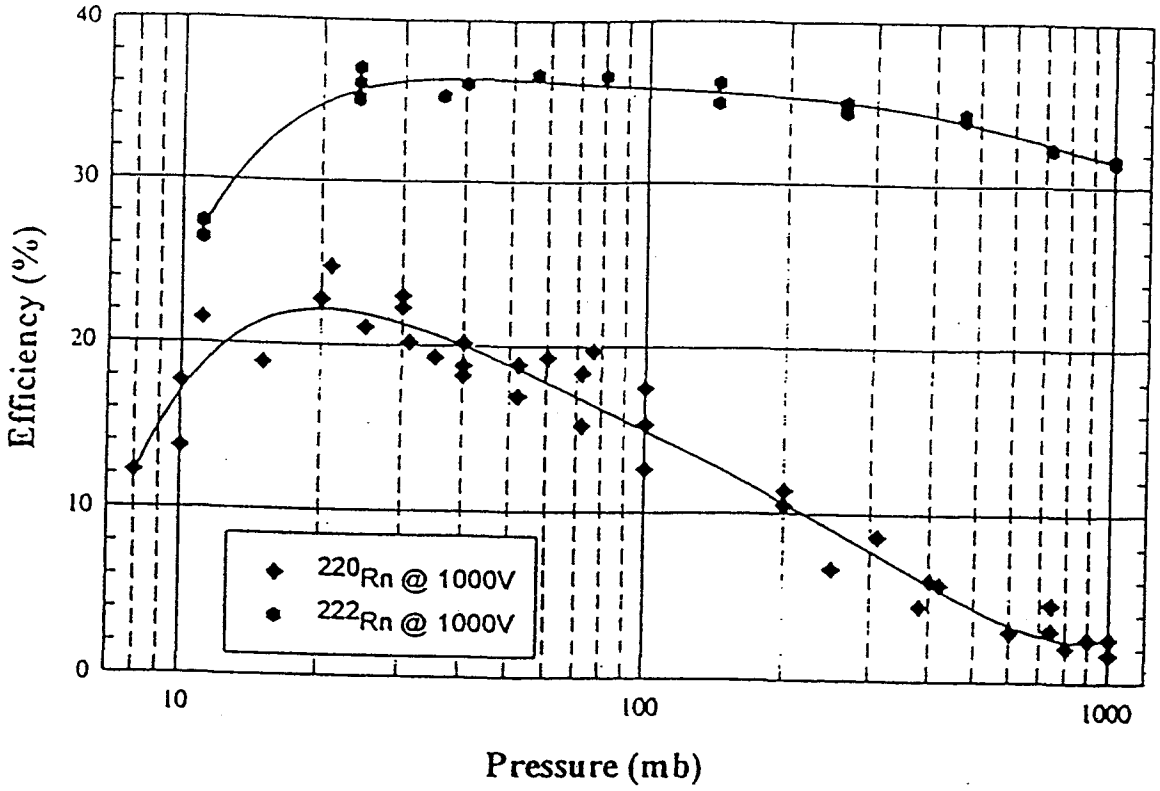
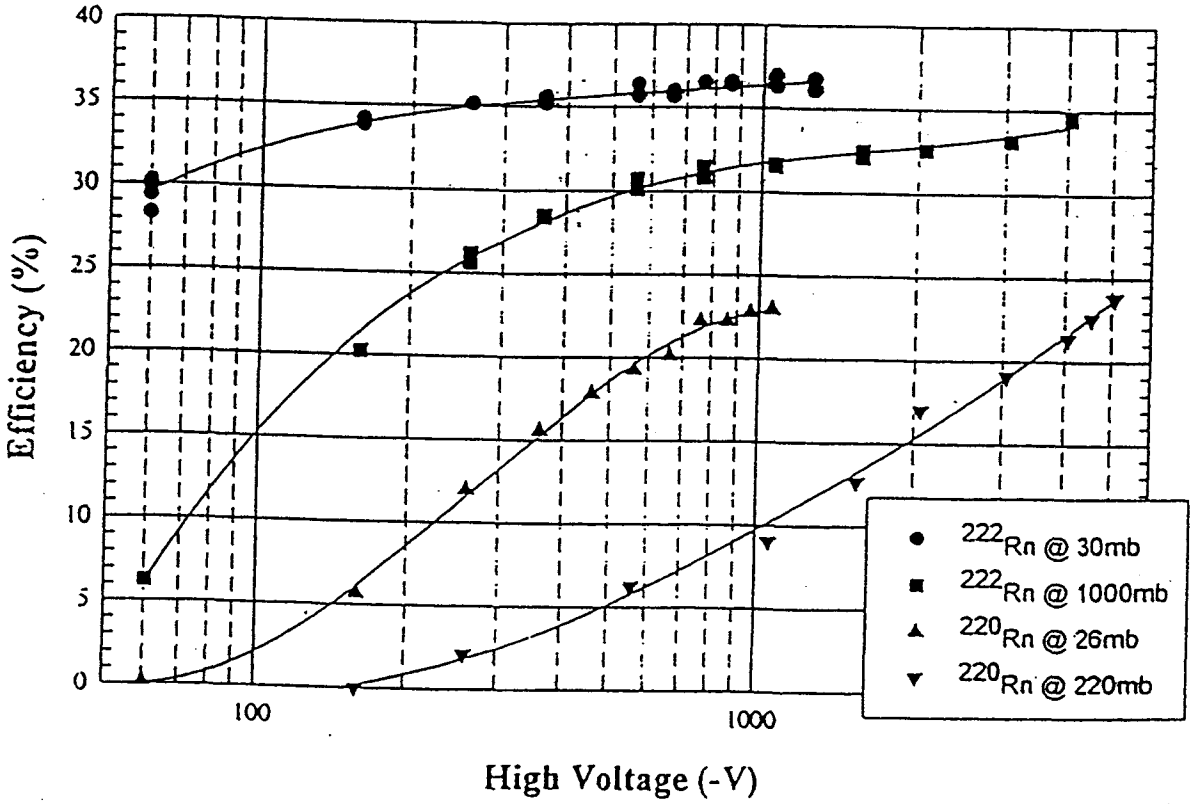


Fig. 2-7. Initial vs Final Position for SNO Electrostatic Chamber
These two graphs show the output of the monte carlo program for a ^{222}Rn simulation showing the initial radius and z coordinate of the Po ions and the final locations.
Right: The initial and final positions for ions that were lost Most of the ions were lost in the initial recoil step. The dead volume can be made out at the top of the detector.
Left: The initial locations of all the ions that made it to the silicon photodiode.

.. HIGH ^{220}Rn DETECTION EFFICIENCY





Background and Sensitivity

- Counter Backgrounds

ESC w/ circ. loop	Date	Thorium cpd (228Th=224Ra)	Uranium cdp (226Ra)
ESC 1	990311	1.3 ± 0.3	43.9 ± 1.2
ESC 2	990311	2.2 ± 0.4	37.1 ± 1.8
ESC 3	990311	7.1 ± 0.8	50.2 ± 2.6
ESC 4	990222	1.7 ± 0.5	54.1 ± 2.7
ESC 5	990402	3.7 ± 0.7	74.7 ± 3.3
ESC 6	990322	1.3 ± 0.3	55.8 ± 1.4

- Sensitivity
  Take D_2O at specs:

Concentration	3.7×10^{-15} gTh/g	4.5×10^{-14} gU/g
Activity	1.3 ddt	45 ddt
In SI units	1.5×10^{-8} Bq/kg	5.2×10^{-7} Bq/kg
Assay 50 tonnes	65 dpd	225 dpd
Correction for decay	55 dpd	225 dpd
Total efficiency	6%	22%
Signal	3.3 cpd	50 cpd
Blank (typical)	3 cpd	50 cpd
S/B ratio	$\simeq 1$	$\simeq 1$

- Possible improvements are
  - double statistics using ^{212}Po – needs relative calibration with ^{216}Po
  - background reduction
  - take larger samples
  - count four columns in parallel on a single counter
- Systematics is 50% for Th, 29% for U, and is dominated by the emanation of ^{220}Rn.

  Observed in a recent spike experiment scaling up to a factor of two, which we will assess at a 15% level in a near future.

BALZER

Conclusions

- Detection of ^{220}Rn
  - counter with low background
  - and high efficiency
  - improvements in sensitivity/background possible
- Associated Assay
  - well practiced
  - re-calibration program running

Radon Workshop

A) Detector Bkgnd Reduction

① Remove U, Th & Ra salts from NaI solution

Final sol'n has $\frac{U, Th, Ra}{I} < 10^{-12}$ gm/g

② Remove Rn from detector (NaI-H_2O & C_2Cl_4) with several He flushes/hr - reduction factor ~ 500.

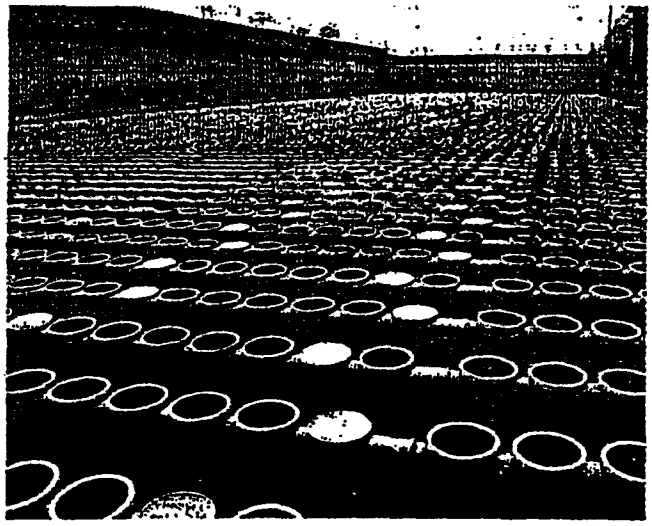
B) Counting Bkgnd Reduction

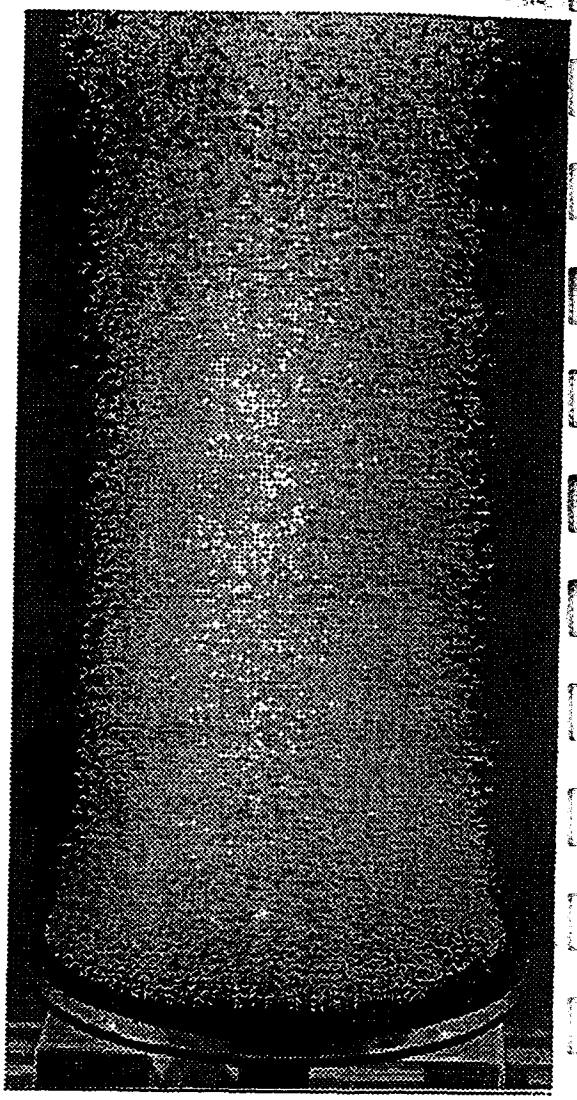
① Separate Rn from $\left\{ \begin{matrix} Ar \\ Xe \end{matrix} \right\}$ by chromatogra

(better for Ar-Rn, poorer for Xe-Rn)

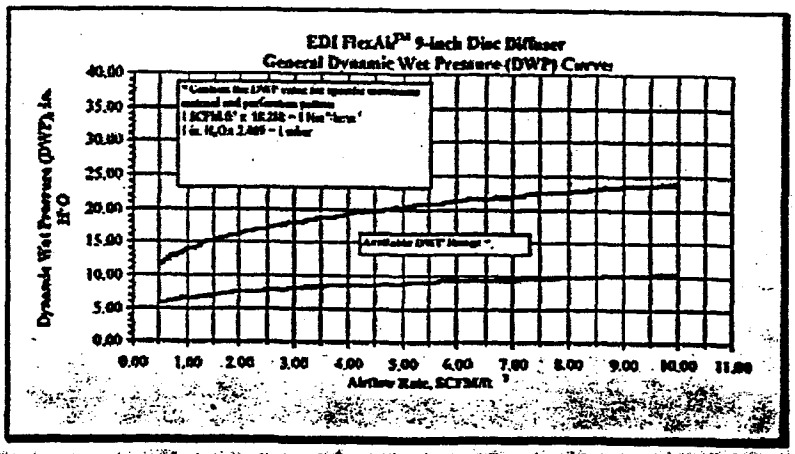
(Do double chromatography for Xe-Rn)

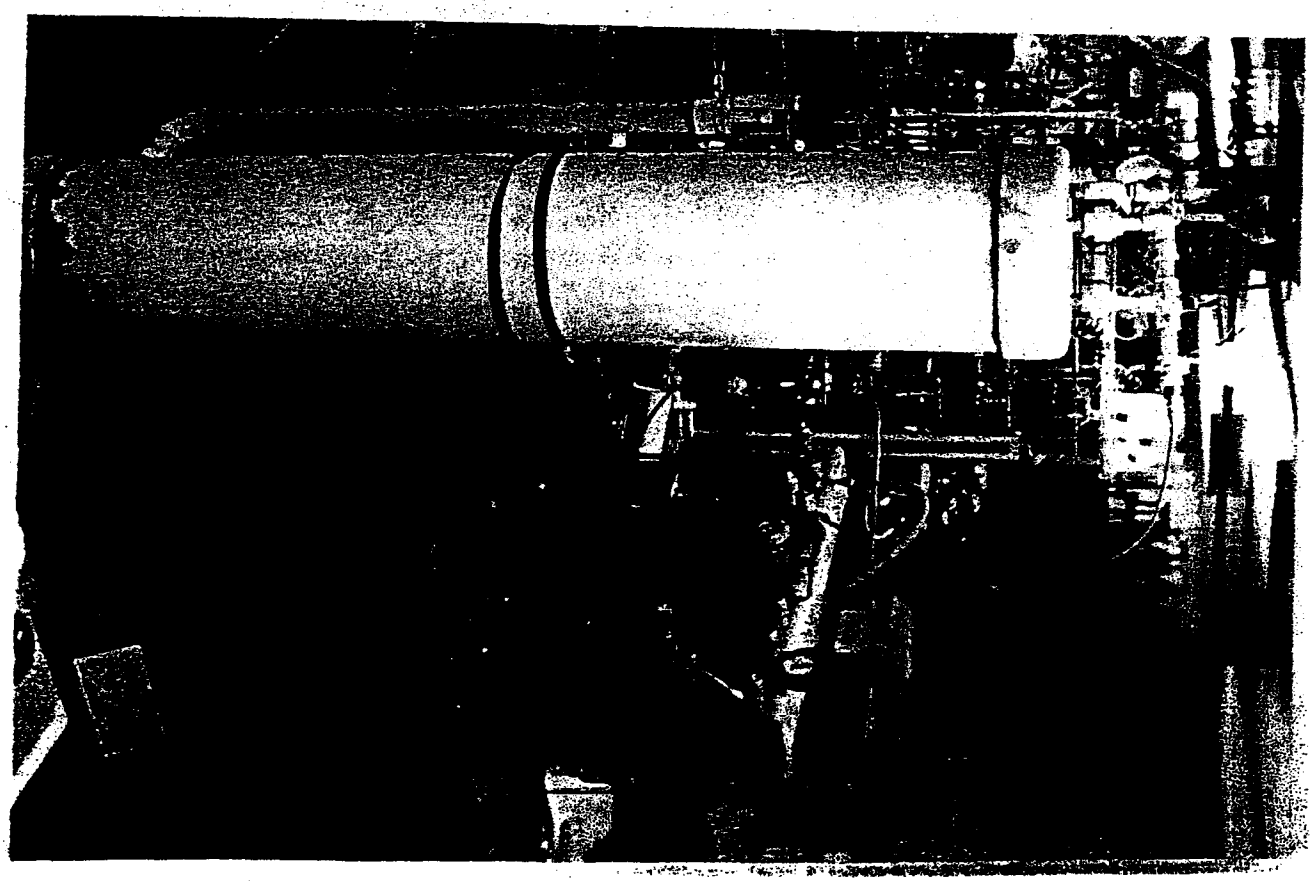
② ^{127}Xe decay - Auger electron - γ coincidence reduces Rn decay bkgnd



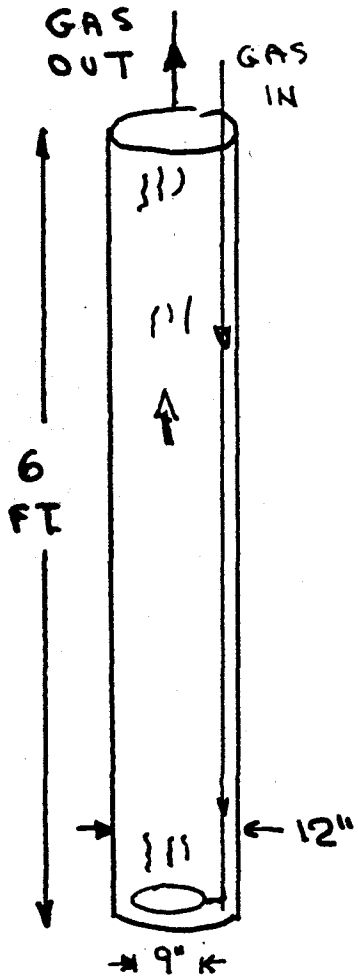


9" Disc Dynamic Wet Pressure (DWP)





GAS DIFFUSER EXTRACTION RATE



MAKE ^{133}Xe WITH Pu-Be SOURCE

1/e EXTRACTION GAS FLOW

= 30 LITERS He @ S.T.P.

GAS COLUMN HEIGHT $= \dfrac{30 \text{ LITERS}}{\text{DIFFUSER AREA } (410 \text{ cm}^2)} = 75 \text{ cm}$

SINCE DIFFUSER CAN EMIT

3 m HIGH COLUMN / min ⇒

$\tau(1/e) \sim 15$ sec

& $1 - e^{-4}$ IN ~ 1 MINUTE

BUBBLE (1mm dia) RISE VELOCITY ~ 20 cm/sec

FOR 8.5 m HIGH DETECTOR - T(RISE) = 40 sec

TOTAL EXTRACTION TIME = 1 min + 40 sec

$\lesssim$ 2 min

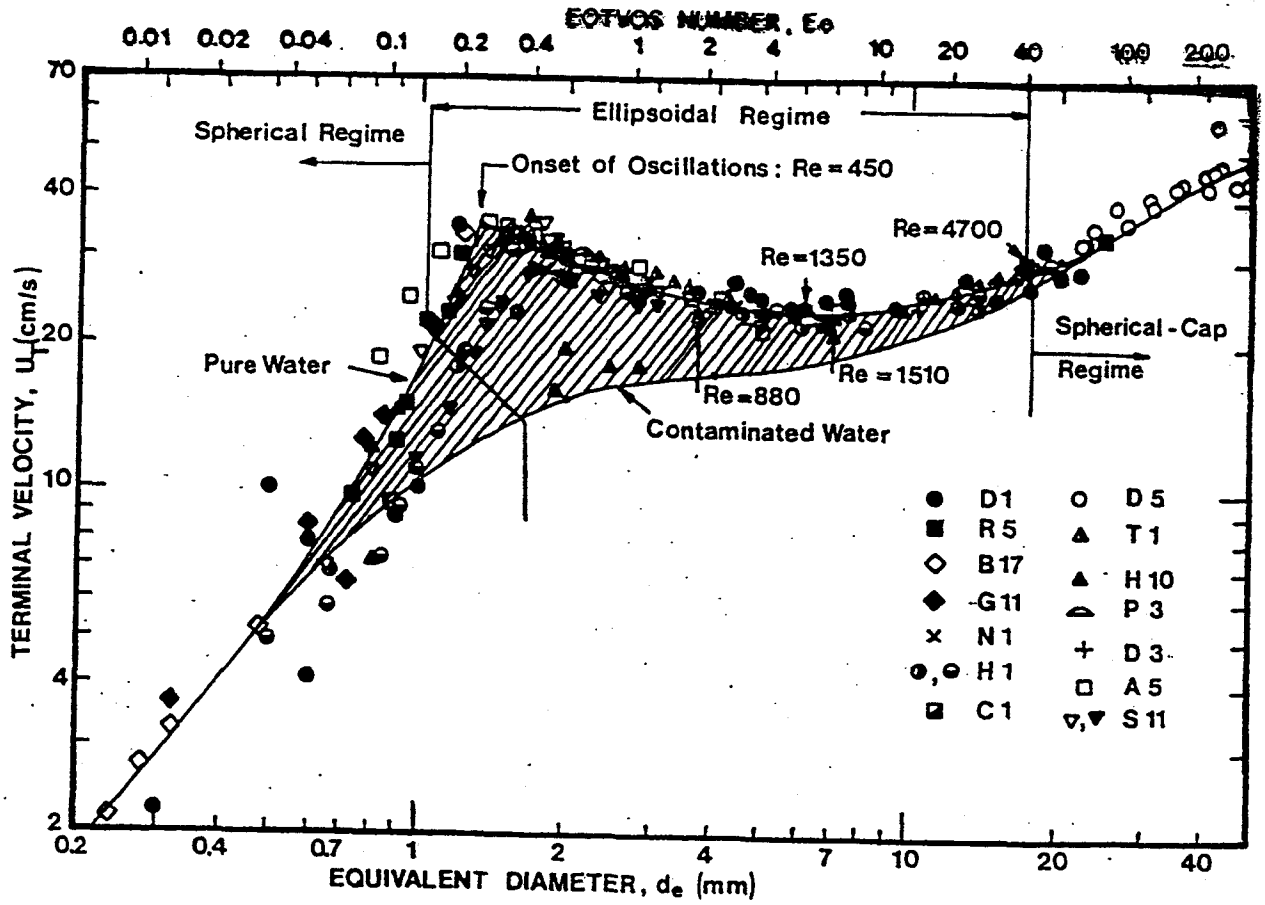


FIG. 7.3 Terminal velocity of air bubbles in water at 20°C.

TABLE 7.1

Motion of Intermediate Size Air Bubbles Through Water at 28.5°C[a]

d_e (mm)	Re	E	Path
<1.3	<565	>0.8	Rectilinear
1.3 to 2.0	565 to 880	0.8 to 0.5	Helical
2.0 to 3.6	880 to 1350	0.5 to 0.36	Plane (zig-zag) then helical
3.6 to 4.2	1350 to 1510	0.36 to 0.28	Plane (zig-zag)
4.2 to 17	1510 to 4700	0.28 to 0.23	Rectilinear but with rocking

[a] After Aybers and Tapucu (A5).

HAXTON (1988)

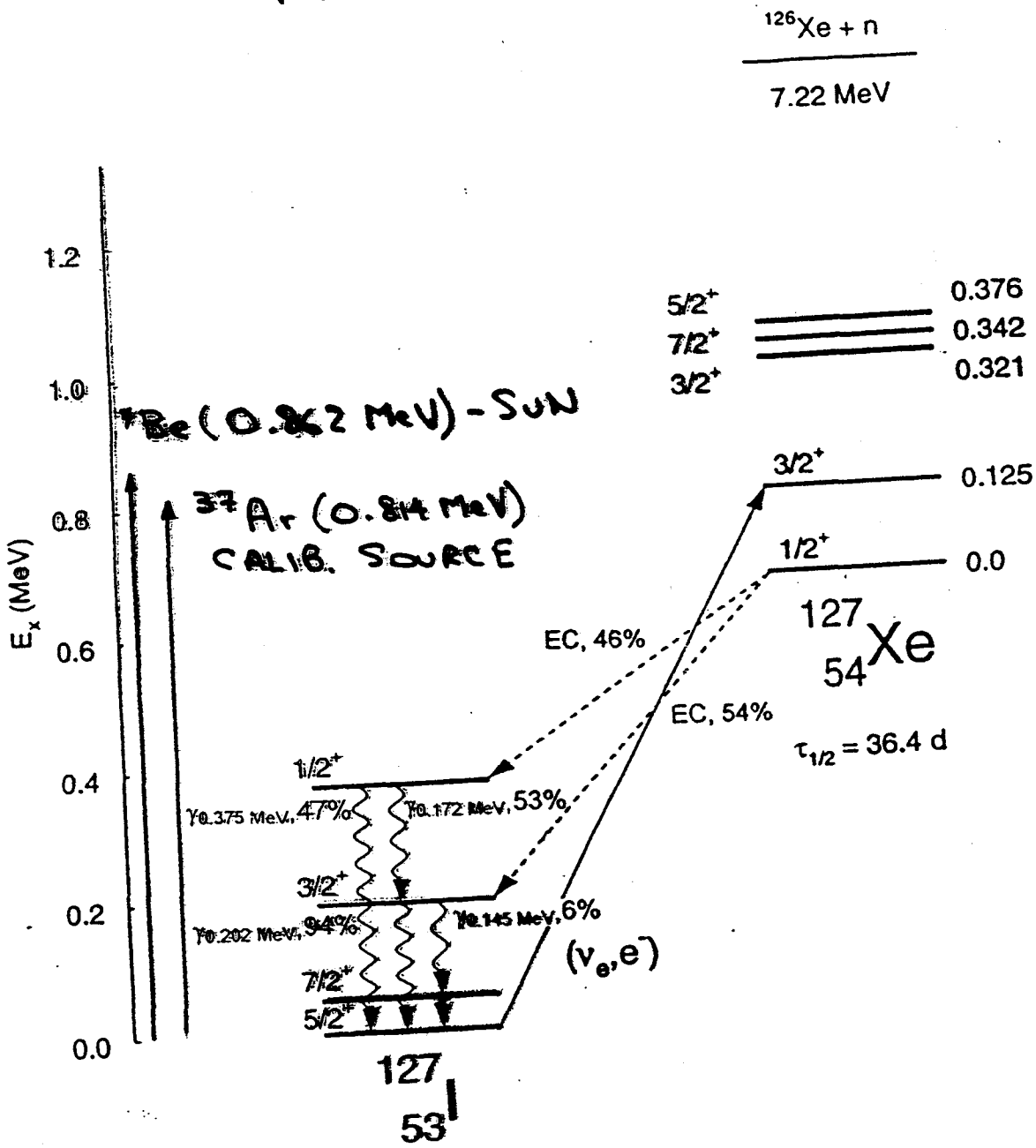


Figure 1.4 Decay scheme for $^{127}Xe \rightarrow {}^{127}I$. The decay proceeds via electron capture. The absence of a ground state to ground state transition means that ^{127}Xe decays to an excited state of ^{127}I. Deexcitation of the ^{127}I nucleus results in nuclear gamma rays in the proportions and energy shown.

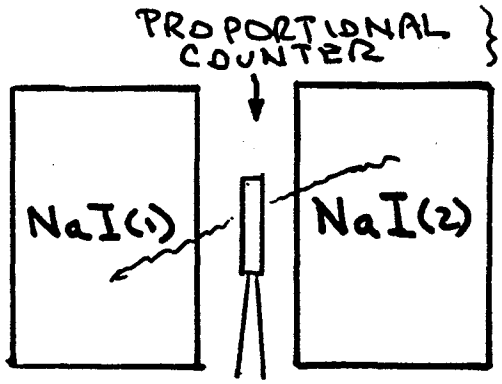
COUNTER EFFICIENCY DETERMINATION

$$^{127}Xe \rightarrow {}^{127}I^{*}$$

$$\xrightarrow{19\%} {}^{127}I + \gamma(375\ keV)$$

$$\xrightarrow{27\%} {}^{127}I + \gamma(203\ keV) + \gamma(172\ keV)$$

$$\xrightarrow{54\%} {}^{127}I + \gamma(203\ keV)$$



FILL WITH ^{127}Xe
(MADE AT UNIV. OF WASHING
VAN de GRAFF
-DEREK STORM

$$Eff(PC) = \frac{(PC) + (NaI(1)) + (NaI(2))}{(NaI(1)) + (NaI(2))}$$

$$Eff(NaI(1)) = \frac{(PC) + (NaI(1)) + (NaI(2))}{(PC) + (NaI(2))}$$

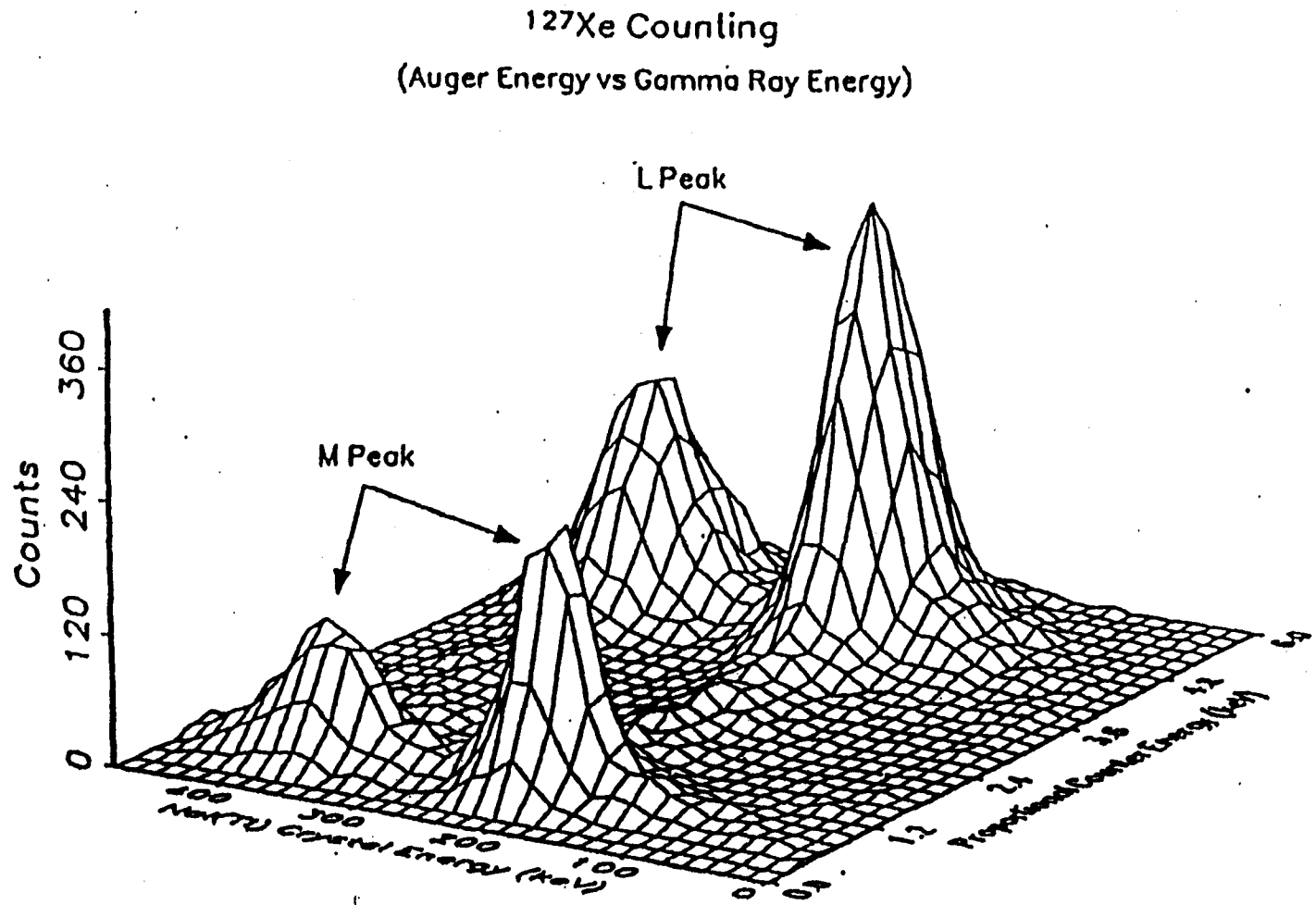


Figure 7: AUGER – GAMMA RAY COINCIDENCE COUNTING IN ^{127}Xe. This graph shows the coincidences observed between Auger electrons (observed in a miniature proportional counter)

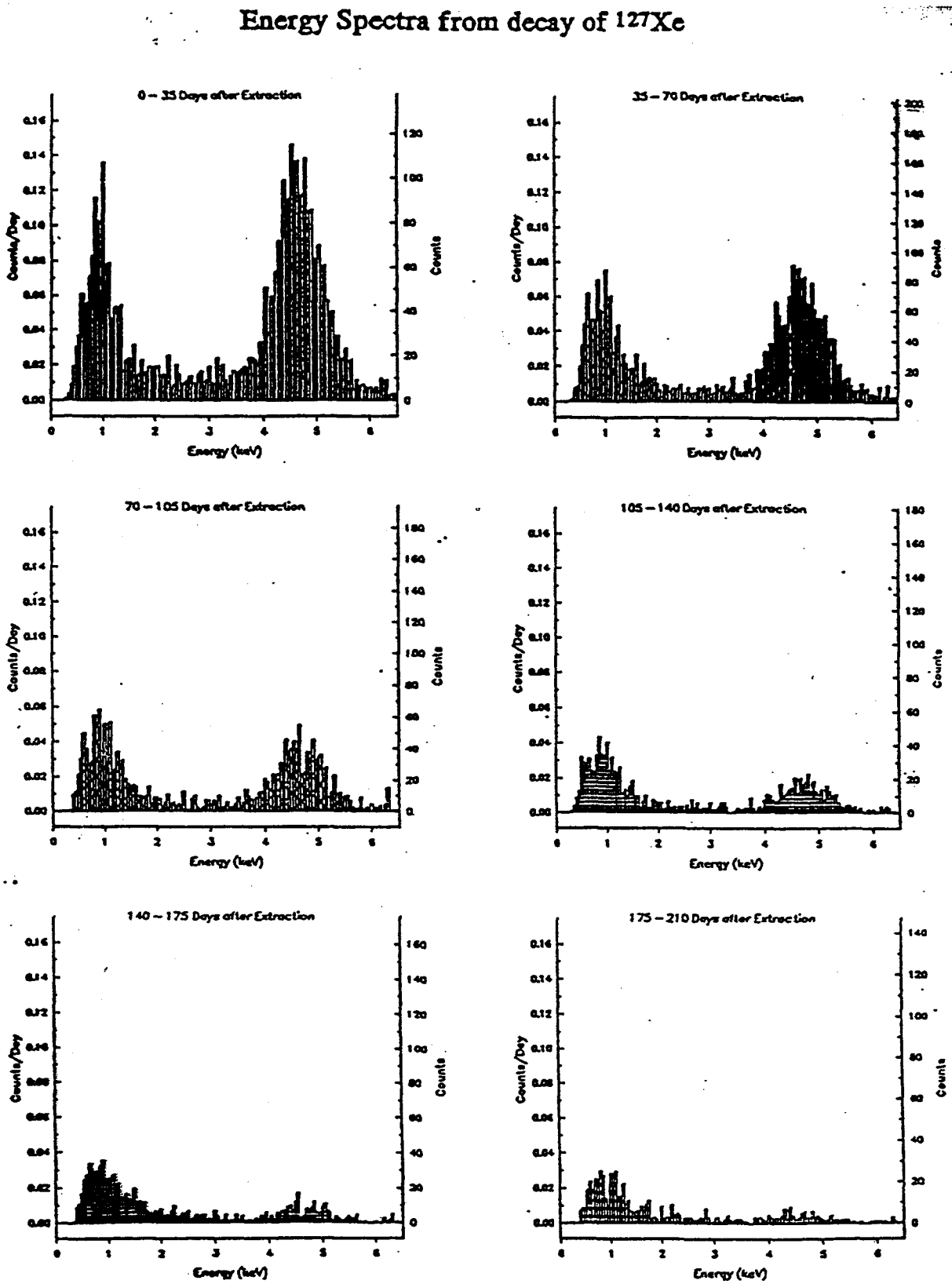


FIGURE 5. ENERGY SPECTRA IN PROPORTIONAL COUNTERS. Shown are the energy spectra for the first six half lives for all runs of the LAMPF calibration experiment combined. The peak at 4.7 keV is due to Auger electron emission from the L shell, and the peak at 0.9 keV from the M shell. Events shown in this histogram are selected as "fast", from the forward peak in the Inverse Risetime spectrum.

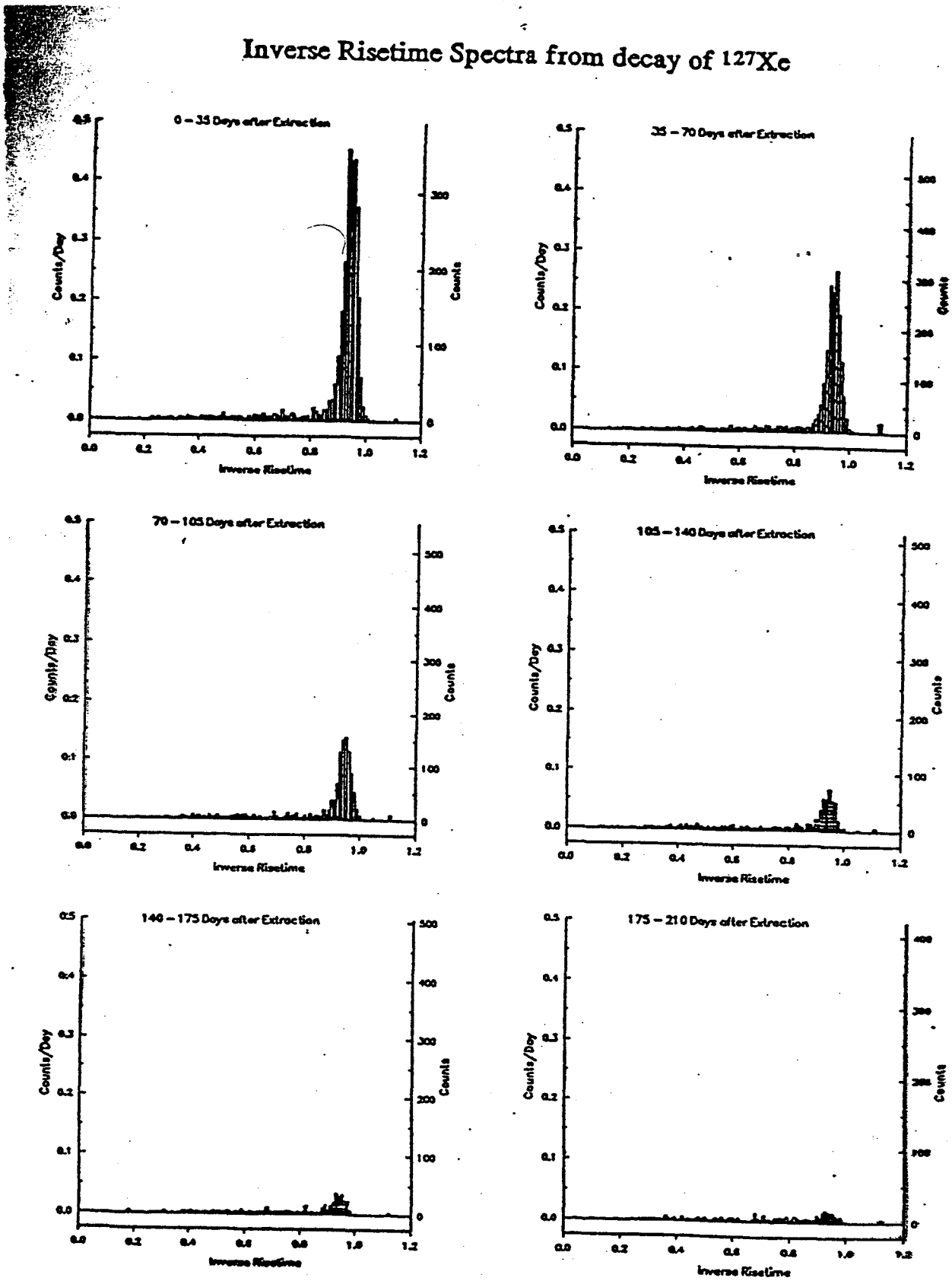


FIGURE 6. INVERSE RISETIME SPECTRA IN PROPORTIONAL COUNTERS. Shown are the inverse risetime spectra for the first six half lives for all runs of the LAMPF calibration experiment combined. The peak between 0.9 and 1.0 represents the "fast" localized events in the counter; the slower tail is from distributed events (e.g. Compton scattered electrons). Events shown in this histogram are selected from the region 3.3 - 6.1 keV, approximately 2 FWHM about the L Auger peak.

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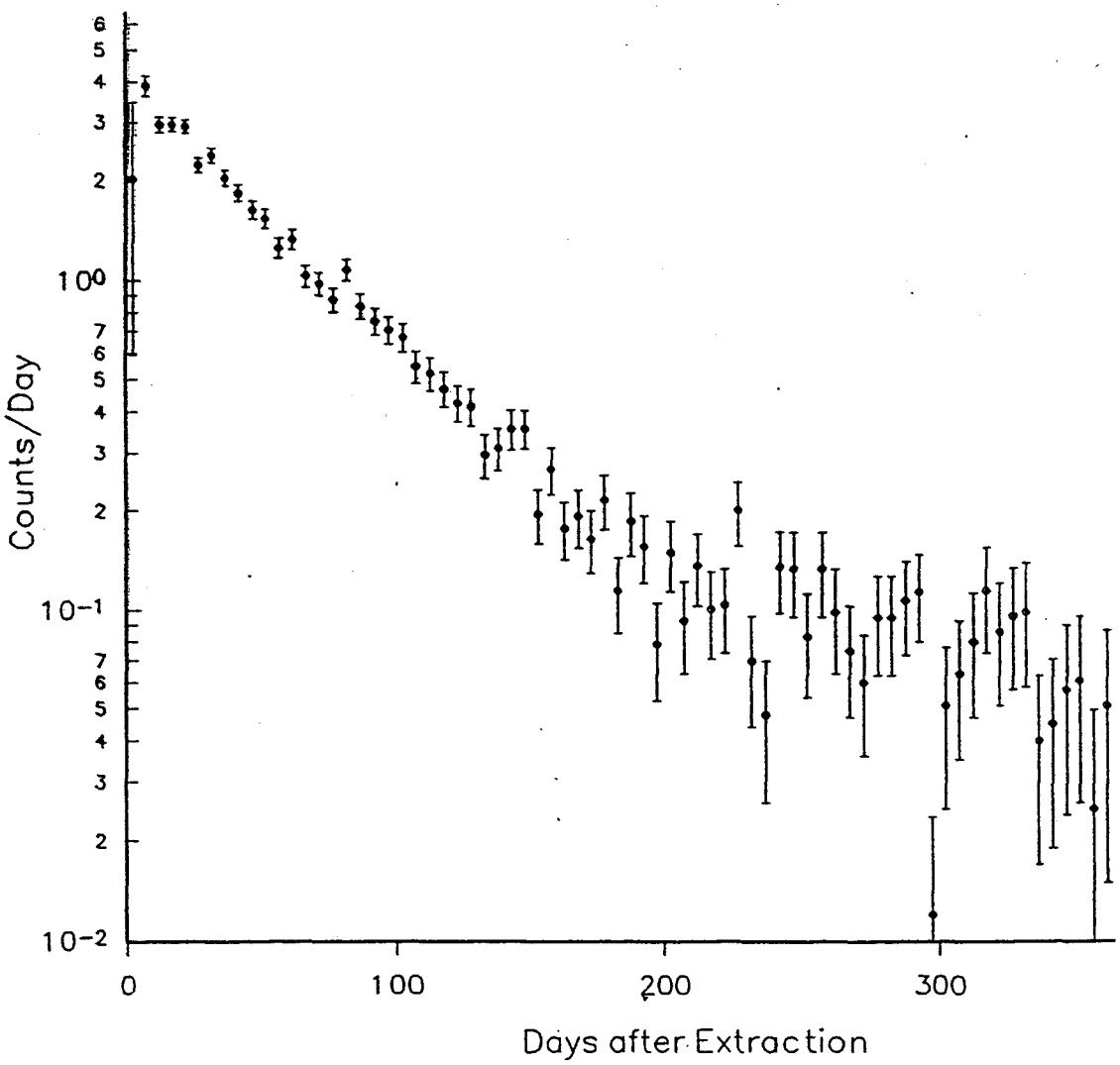
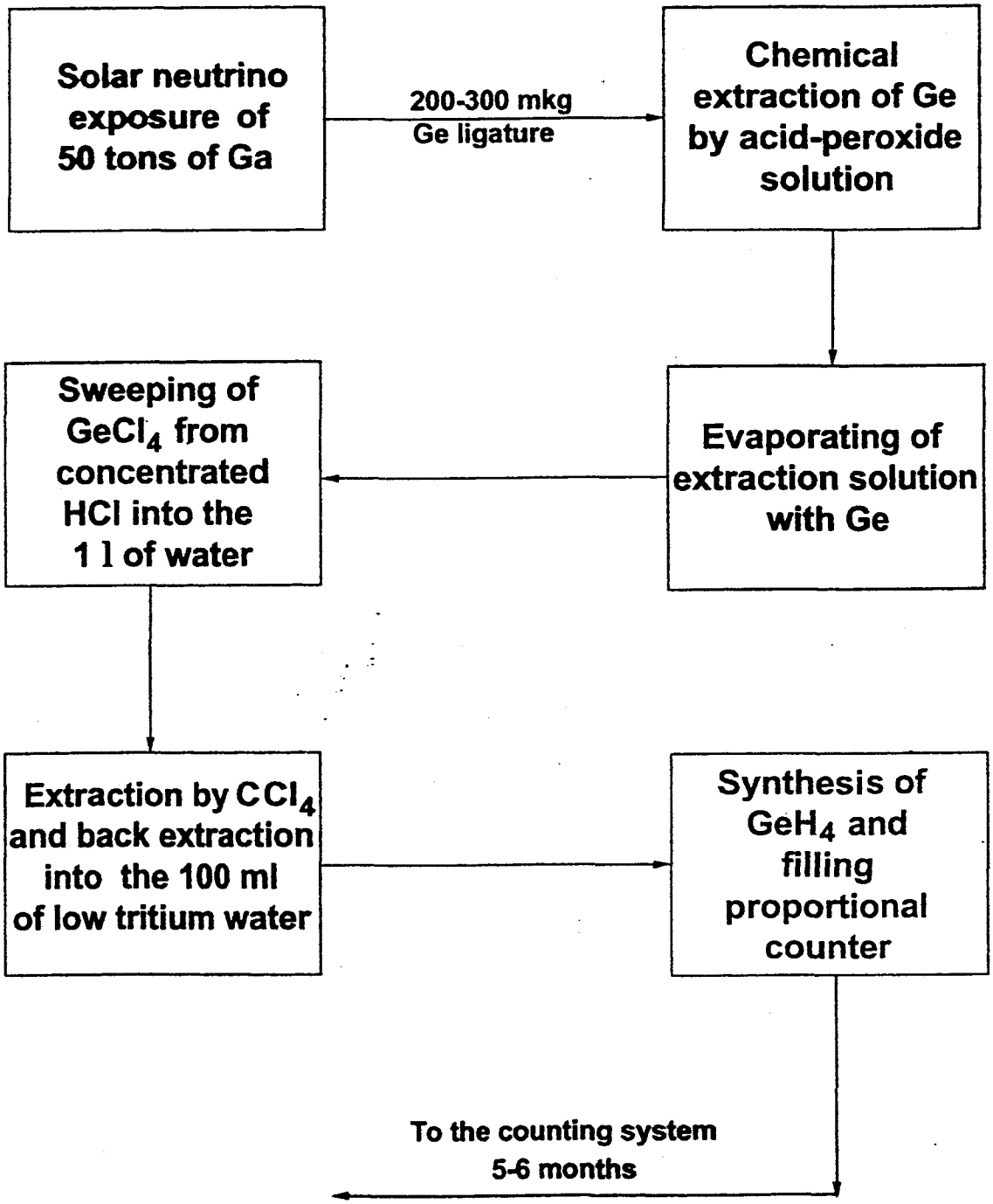
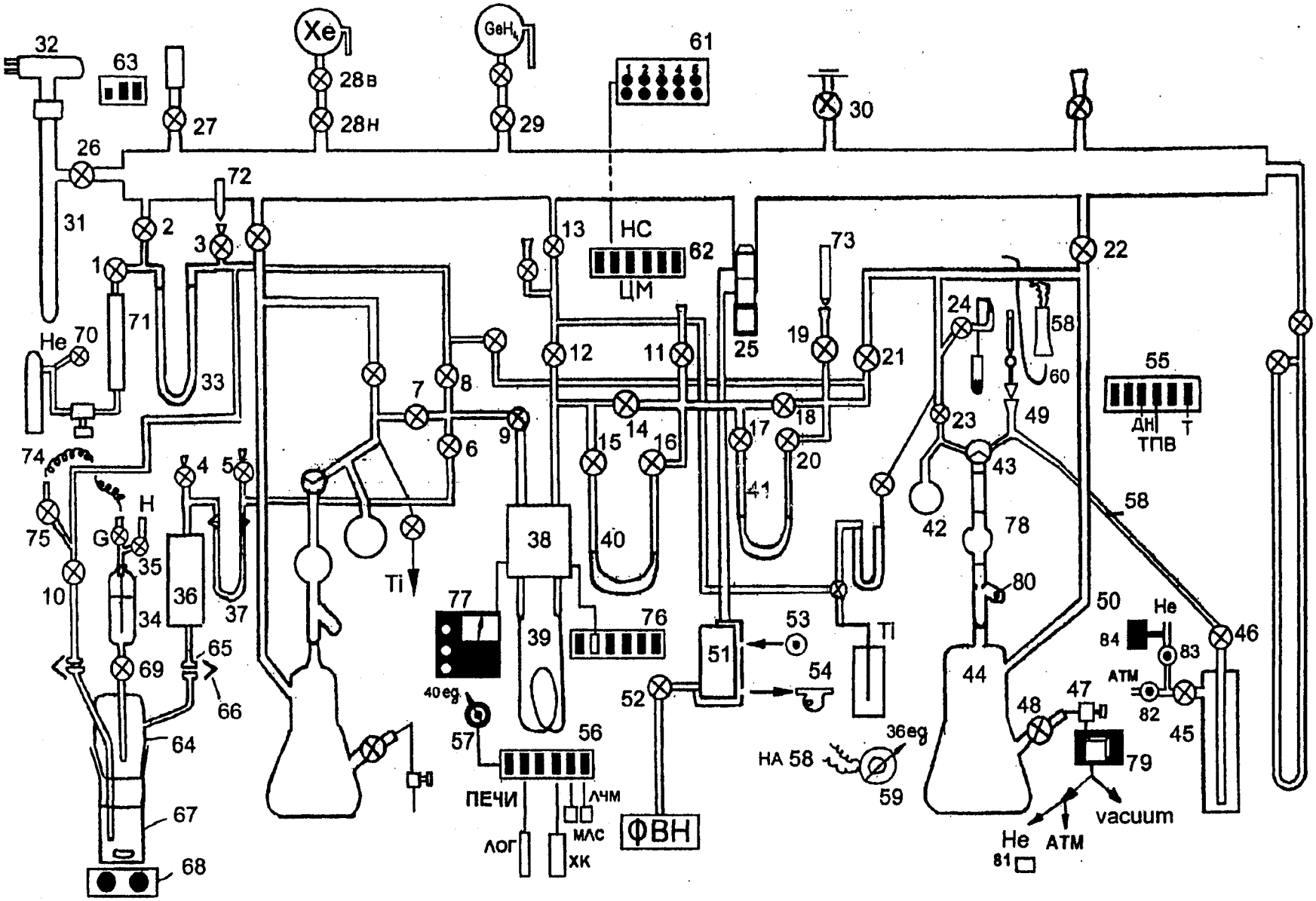
Iodine Data Rate Plot



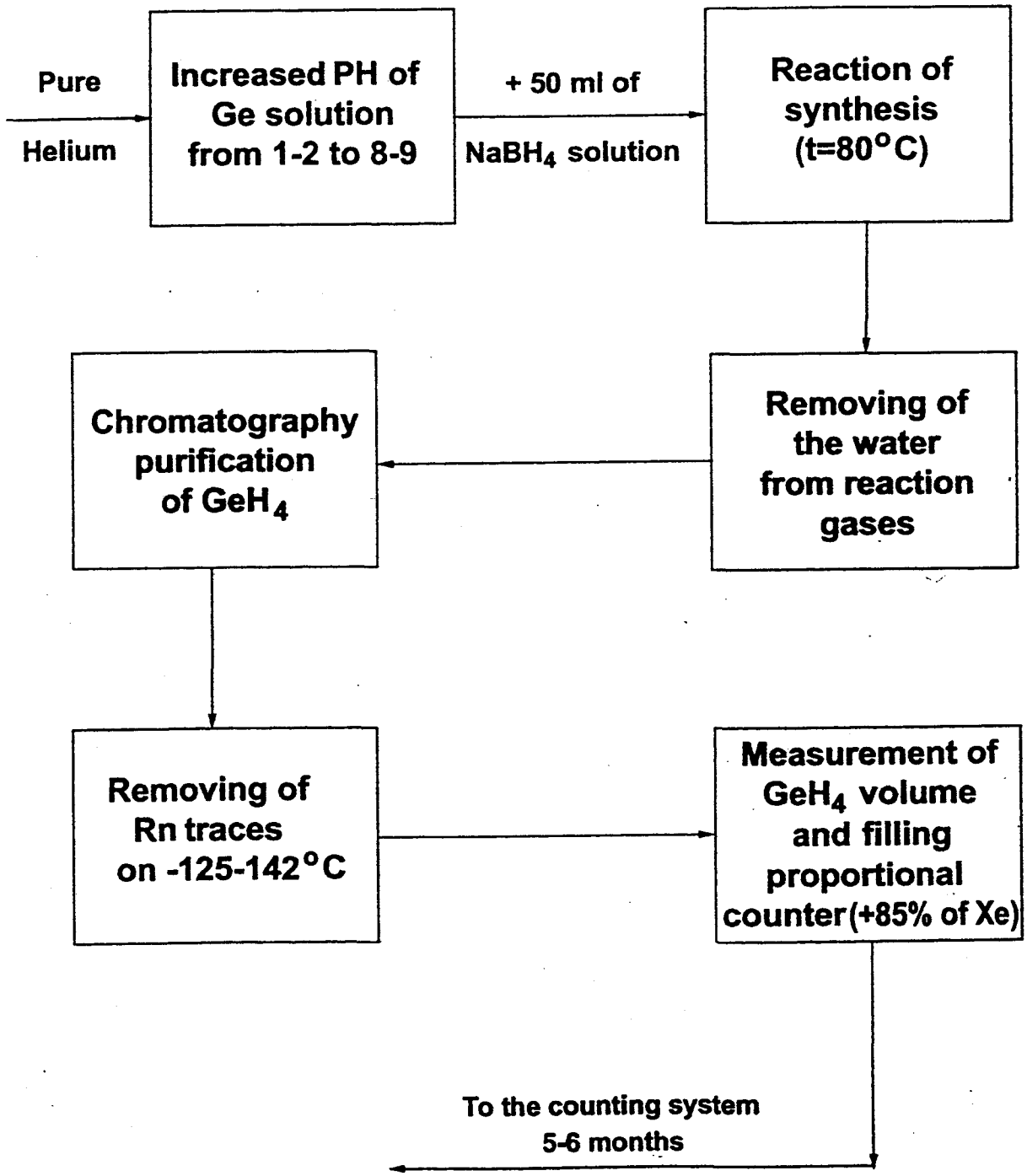
Chemical Technology of SAGE



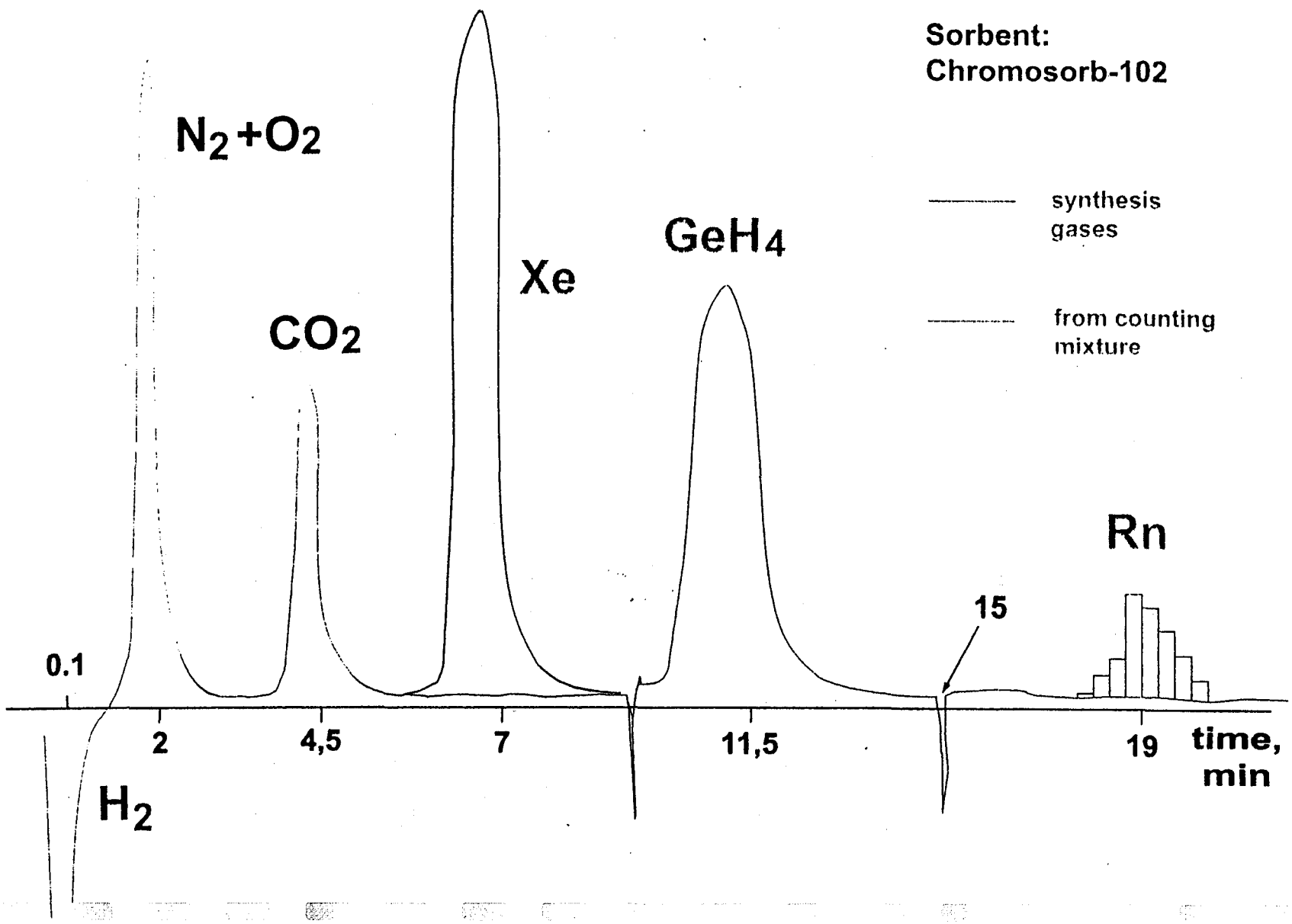


T Mirmov SAGE

Synthesis of Germane (GeH_4)



Chromatography purification of germane



I. Mizmov, SAGE

Sources of Rn in Synthesis of Germane (GeH_4)

Atmospheric Air in reaction part of installation

Atmospheric Air dissolved in reactives

Removed by chromatography

Rn emanate from chromatography sorbent materials

Rn emanate from constructional materials of synthesis installation (viton o-rings, glass)

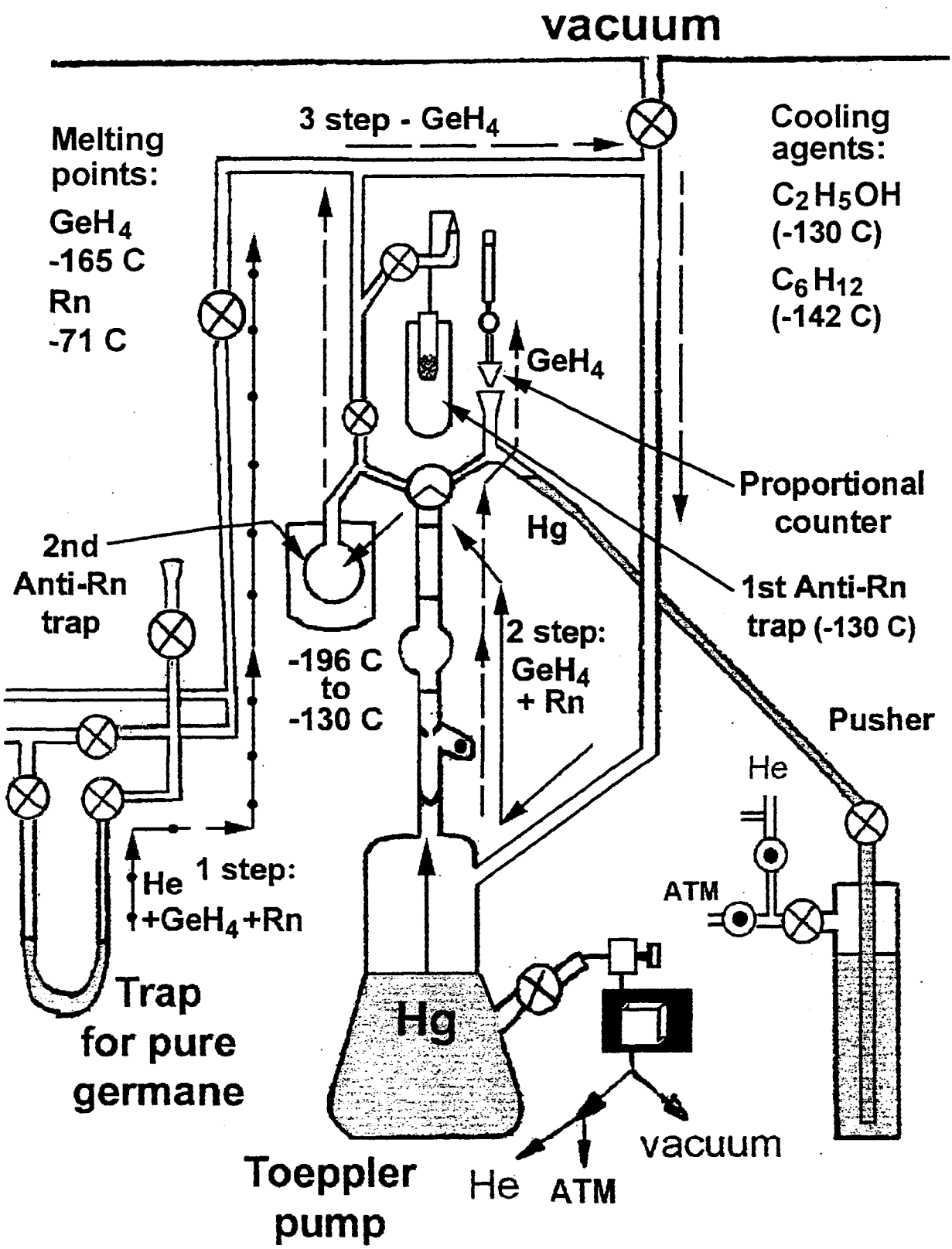
Removed by low temperuture sorbing

Rn sorbed on internal surfaces of the system and dissolved in grease and Hg

Removed by evaporation

Rn Emanation Abilities

Silica gel	~ 0.4 atoms/h/g
Chromosorb-102	~ 0.07 atoms/h/g
Viton o-rings	$(28\pm4)\cdot10^{-2}$ atoms/h
Viton o-rings from SNO	$(13\pm6)\cdot10^{-3}$ atoms/h
Laboratory Air	2 pCi/l ~ 30 atoms/cm^3 $\sim 10^{-1}$ Bq/l


vacuum
3 step - GeH4
Melting points:
GeH4
-165 C
Rn
-71 C
Cooling agents:
C2H5OH
(-130 C)
C6H12
(-142 C)
GeH4
Proportional counter
Hg
2nd Anti-Rn trap
1st Anti-Rn trap (-130 C)
-196 C to -130 C
2 step: GeH4 + Rn
Pusher
He
ATM
He
1 step: +GeH4+Rn
Trap for pure germane
Hg
Toeppler pump
He
ATM
vacuum

INTERNAL RADON IN SOLAR NEUTRINO RUNS

Average per run

Without antiradon procedures:	**~ 11 atoms/run**
Runs with antiradon procedures:	**~ 2 atoms/run**

RADON systematic in SAGE.

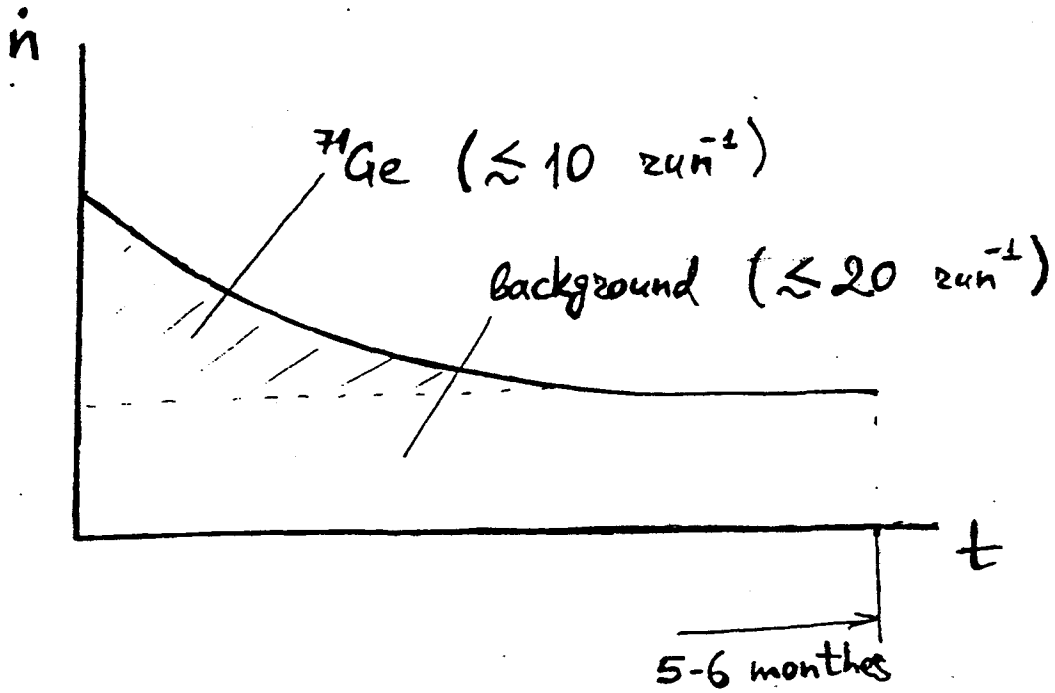
Gorbachev V.,
Institute for Nuclear Research, Moscow.

^{71}Ge:

$T_{1/2}$=11.43 days
e-capture
Auger-electrons –

M	L	K
0.1 keV	1.2 keV	10.4 keV

Time analysis:



Proportional Counter.

quartz cylinder
cathode: Fe, Ø = 0.4 cm, L = 5 cm
anode: W, Ø = 12 mkm
counting gas: Xe+GeH_4 ($\lesssim$ 30%), P ~ 1 atm.

Counting System.

time, energy, ...
Digitizing oscilloscope – waveform
risetime (T_N) – "pointlike" (short tracks)
"extended" (long tracks)

Energy range – 0.3-16 keV

Active shield (NaI)

^{222}Rn – mimic ^{71}Ge pulses

"external" – in air

γ of β-decays

liquid nitrogen evaporation (for purge)

constant counting rate – statistical error

"internal" – gas filling

high counting efficiency

$T_{1/2} = 3.8$ days – systematic error

Measurements:

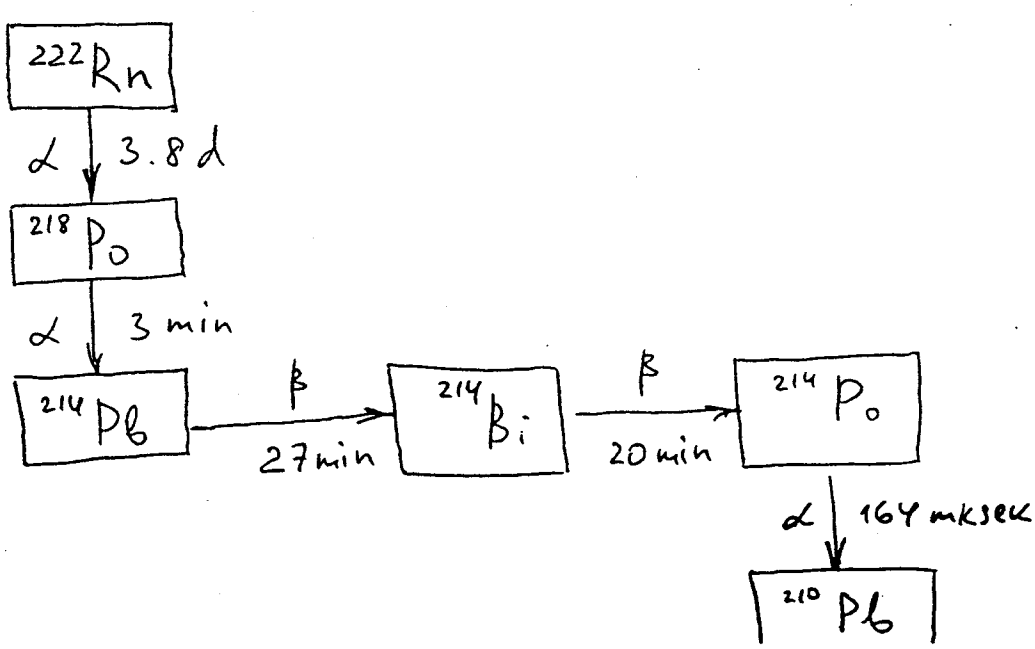
3200 Rn atoms

RST – method of waveform analysis

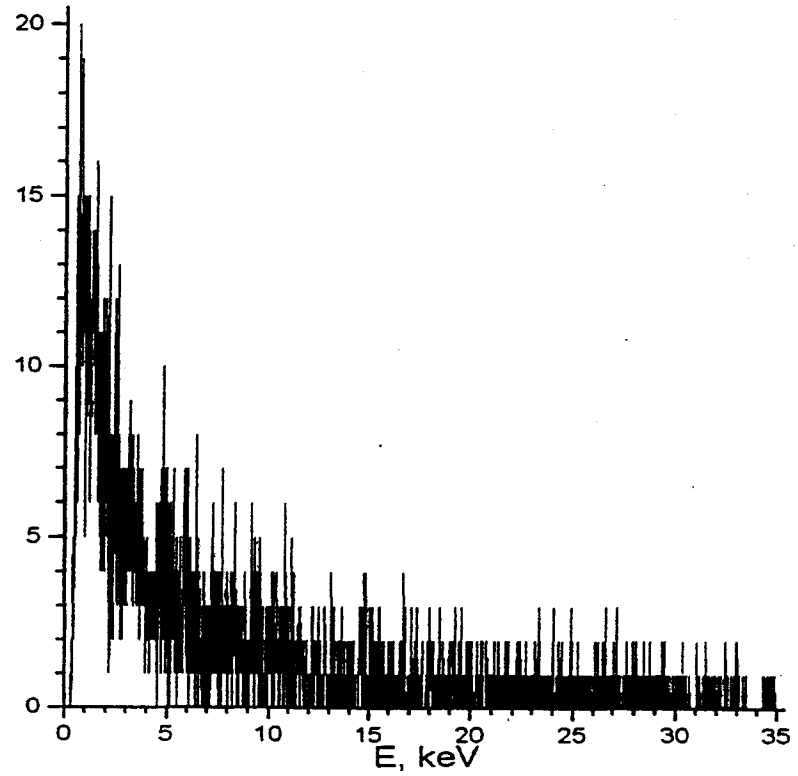
– $E \lesssim 30$-35 keV

(ordinary analysis – $E \lesssim 16$ keV)

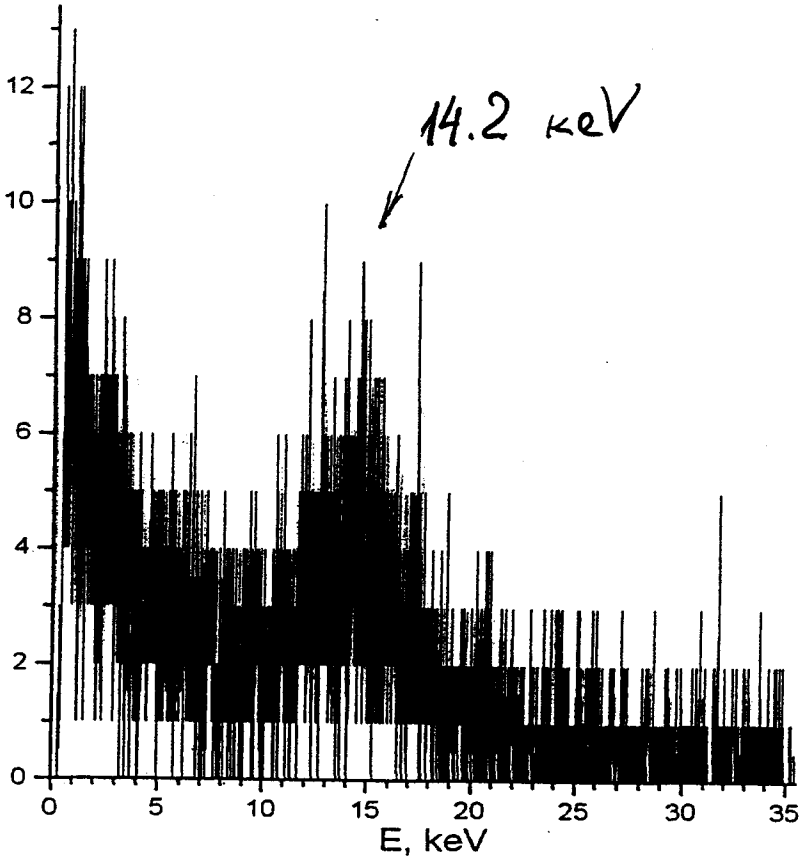
^{222}Rn chain:



Spectrum of events coincident with NaI pulses (β).



Spectrum of events without NaI coincidences. ($\alpha+\beta$)



~1250eV

resolution ~ 41%

- "pointlike"
- recoil nuclei

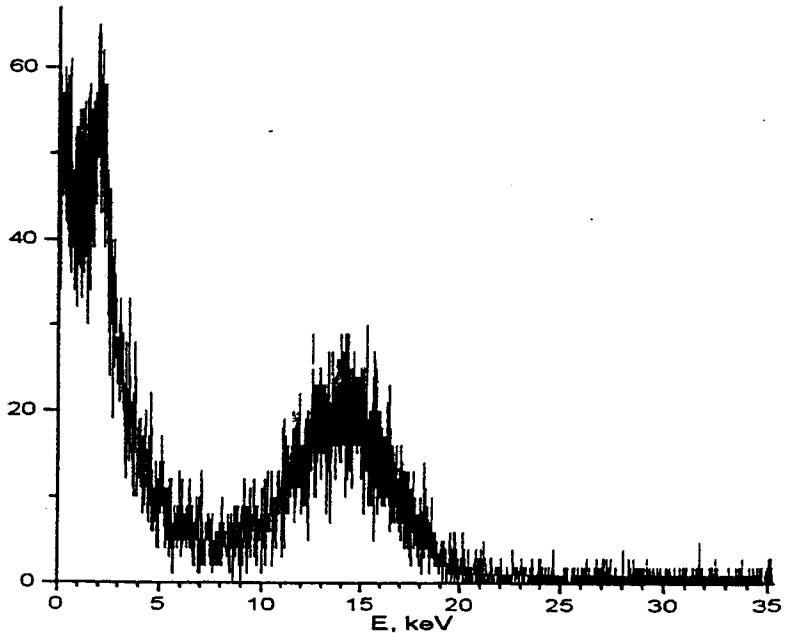
α-decays: α + r. n.
linear tracks
uniform energy loss
- uniform energy distribution
(E < 16 keV L < 0.35 mm)

β-decays: complex energy spectra
nonlinear tracks
noncontinuous energy loss
bremsstrahlung
reflection from walls

for our geometry complex models
do not change spectra

Reflect coefficient: K = 0.3

MC calculated spectrum.



Pulse Selection.

α - energy

β - 1) energy,
2) NaI: ^{214}Pb – 50 %
^{214}Bi – 75 %
3) risetime (TN): L-peak – 83 %,
K-peak – 60 %

Probability of 71Ge pulse mimic:

	^{222}Rn	^{218}Po	^{214}Pb	^{214}Bi	^{214}Po
L:	0	$0.42 \cdot 10^{-2}$	$1.10 \cdot 10^{-2}$	$0.72 \cdot 10^{-2}$	$0.16 \cdot 10^{-2}$
K:	$4 \cdot 10^{-5}$	$4.90 \cdot 10^{-2}$	$0.61 \cdot 10^{-2}$	$0.18 \cdot 10^{-2}$	$1.86 \cdot 10^{-2}$

Overflow events (E>16 keV) are main indicators of radon.

Time cuts for overflows.
15 min befor
3 hours after
- 1.1 % ^{214}Pb,
3.5 % ^{214}Bi, ^{214}Po

1 atom of ^{222}Rn:

L-peak - $4.3 \cdot 10^{-4}$ events
K-peak - $7.8 \cdot 10^{-4}$ events

Counter energy resolution correction:

x 1.3 - L
x 1.5 - K

1 run:	2.1	- L
(75 SNU)	2.5	- K
	7.7	- overflow

L - 0.2% (0.2 SNU)
K - 0.4% (0.3 SNU)

1 hour decreases the radon systematic on factor ~3

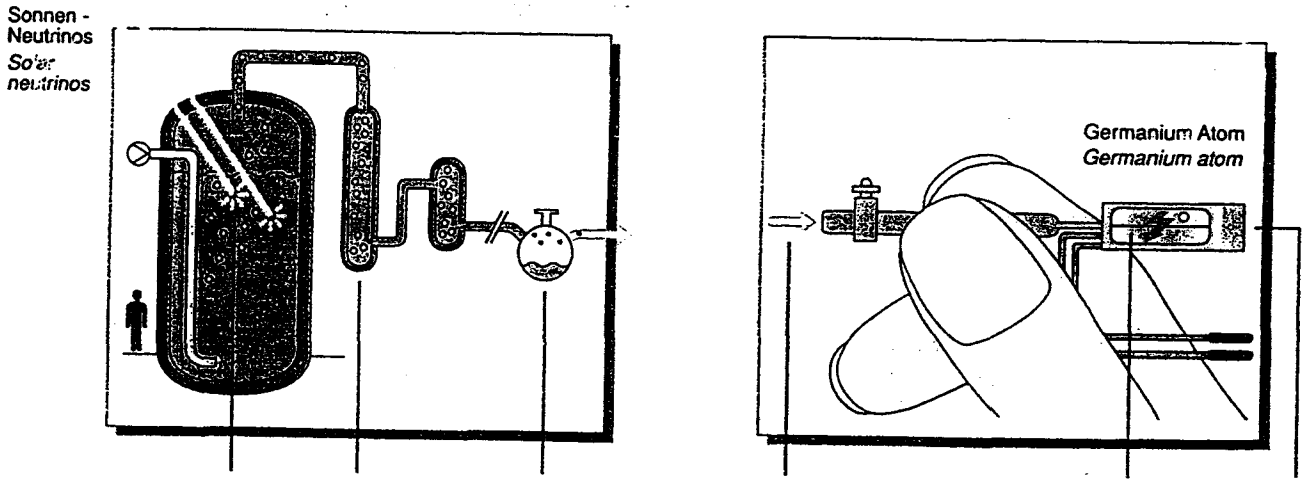
Sudburry
Rn workshop
14 - June - 2000

Suppression of Rn in the GALLEX / GNO counting system

G. Heusser
Max-Planck-Institut für Kernphysik
Heidelberg

M. Wojcik
Jagellonian University
Cracow

detection method

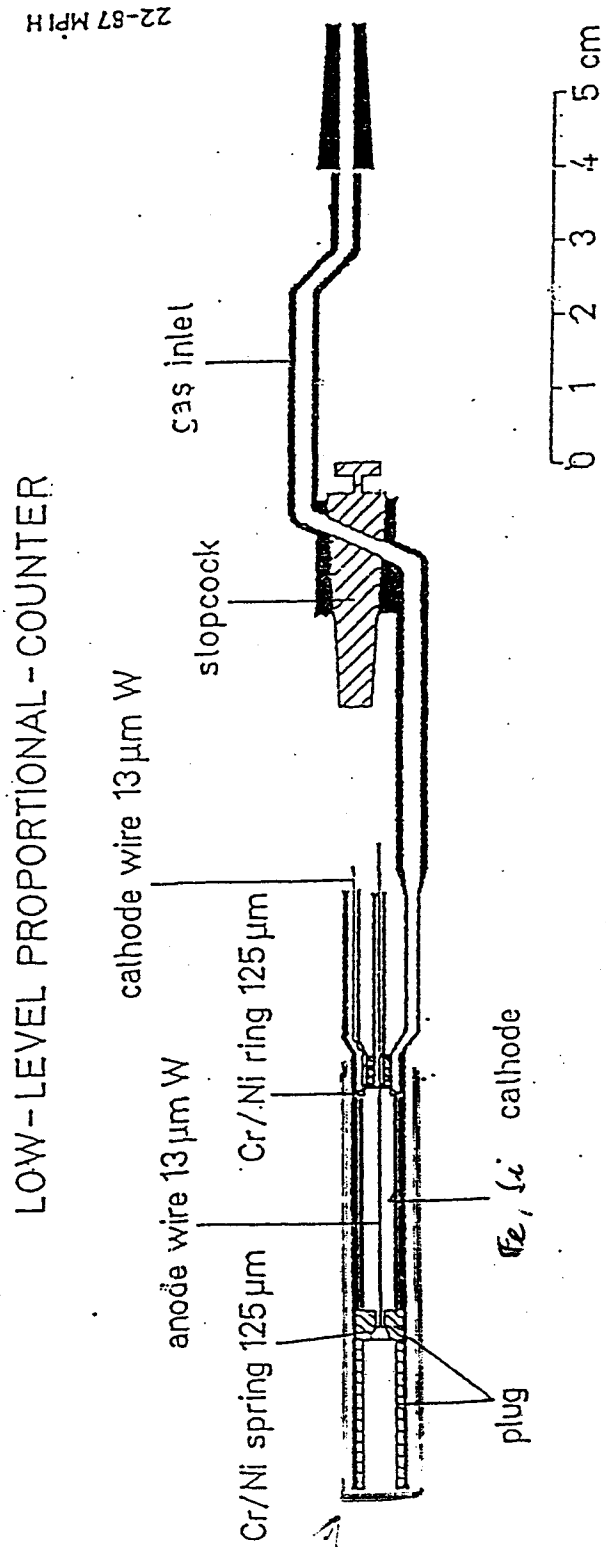


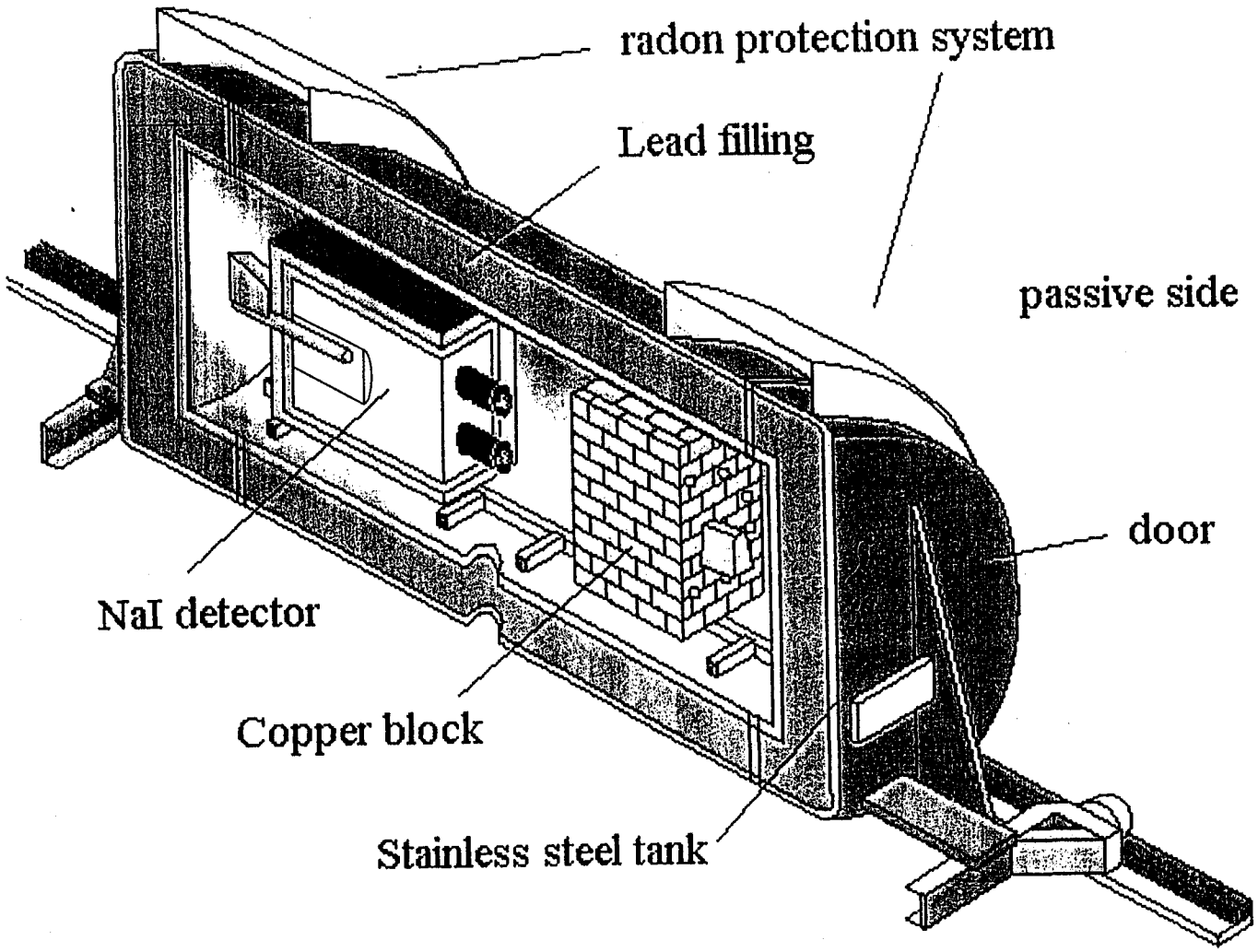
Starting activity ~ 0.5 counts per day of ^{71}Ge

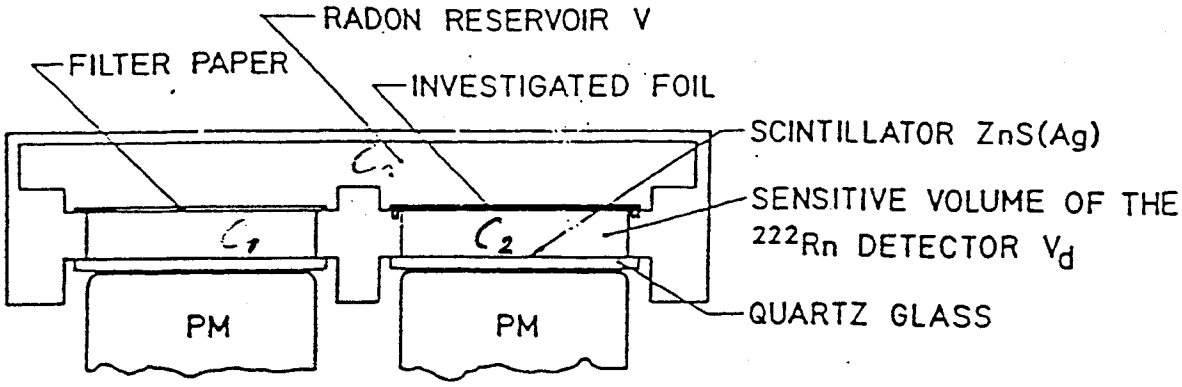
radon problems :

1. Rn in counting gas
2. plate out activity on countersurface
3. Rn in spectrometer shield

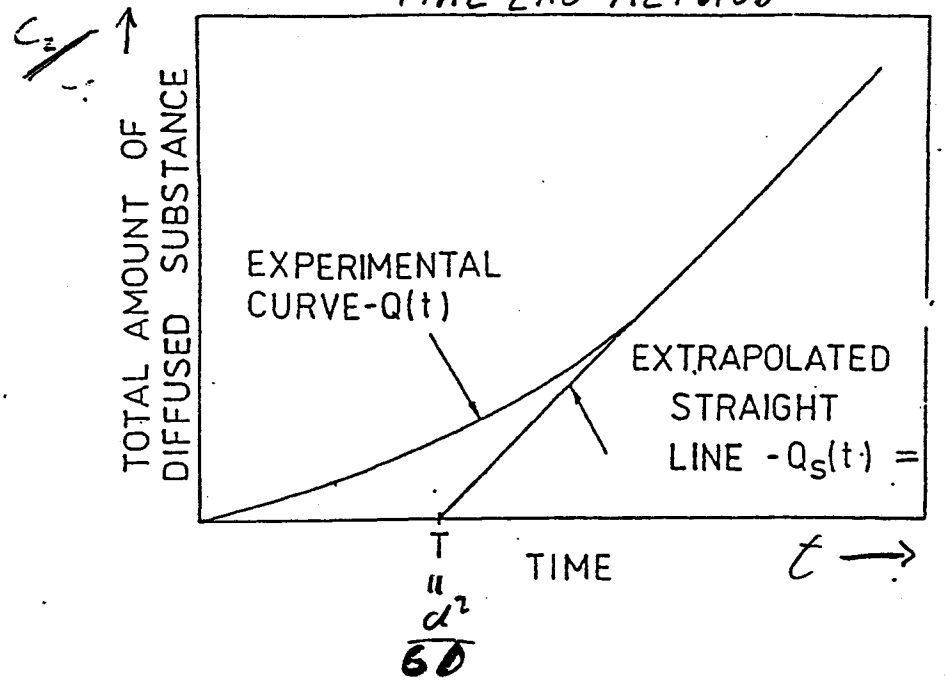
discussion)


22-87 MPIH
LOW-LEVEL PROPORTIONAL-COUNTER
gas inlet
stopcock
cathode wire 13 µm W
Cr/Ni ring 125 µm
anode wire 13 µm W
Cr/Ni spring 125 µm
cathode
Fe, Si
plug
0 1 2 3 4 5 cm


active side
radon protection system
Lead filling
passive side
door
NaI detector
Copper block
Stainless steel tank



TIME-LAG METHOD



$$\frac{SDC_1}{d}\left(t-\frac{d^2}{6D}\right)$$

S = SOLUBILITY

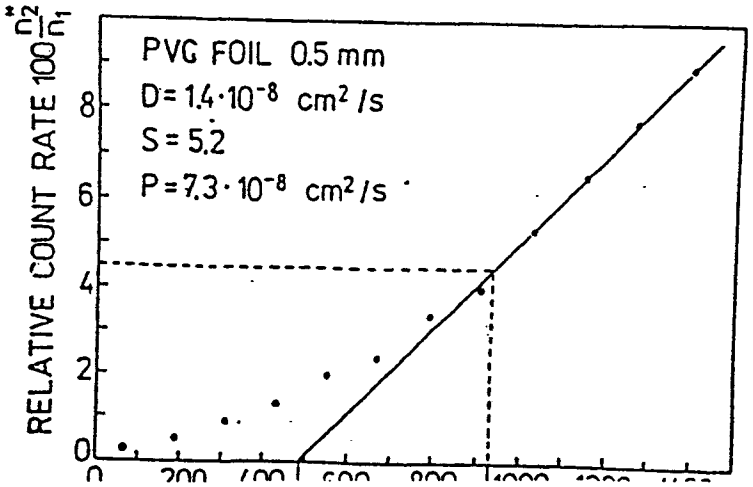
D = DIFFUSION COE.

C_1 = RN CONCENTR.

d = THICKNESS OF FOI

PERMEABILITY $P = D \cdot S$

RATE OF TRANSFER $F = D \cdot S \cdot (C_1 - C_2)/d$

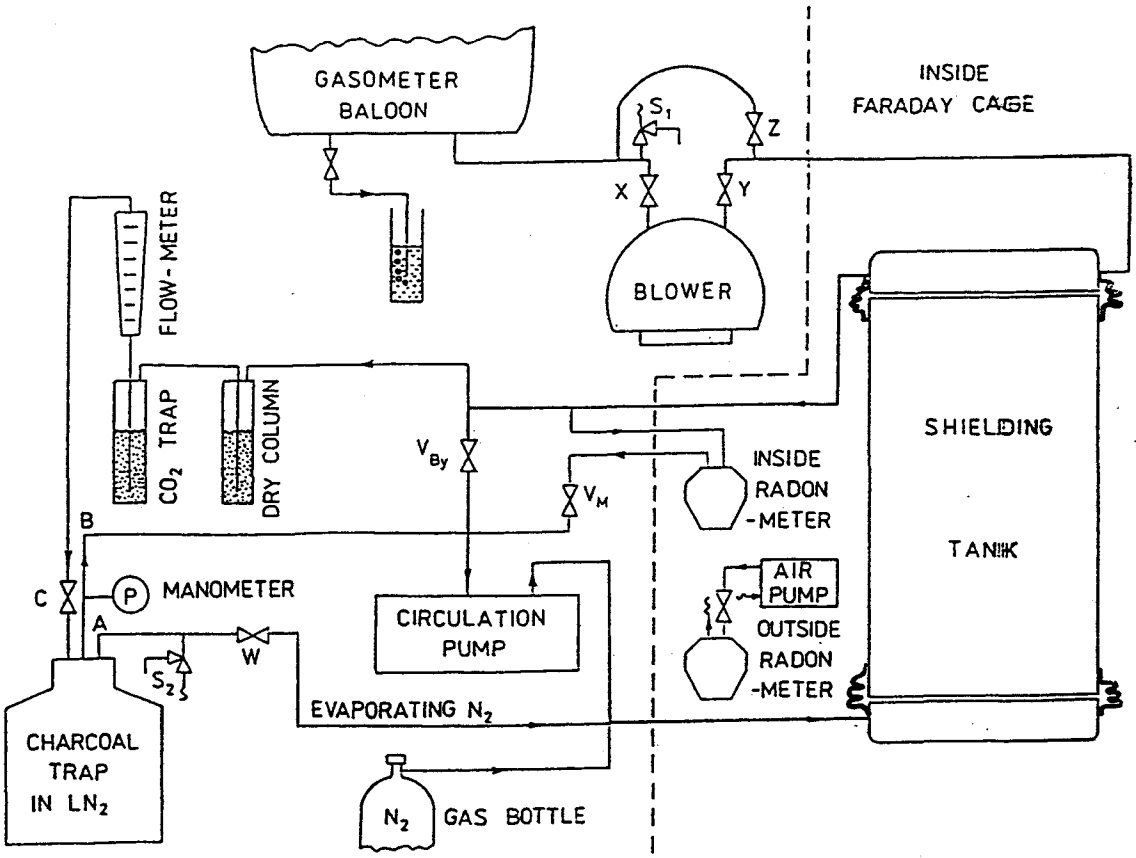


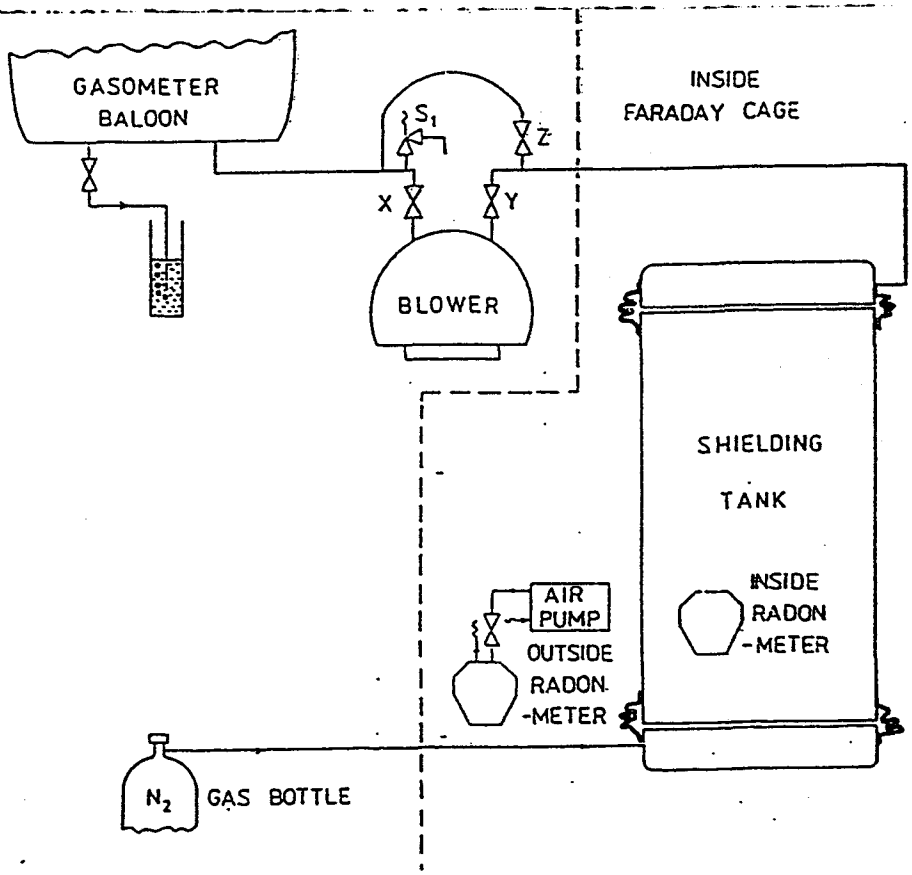
M. WOJCIK

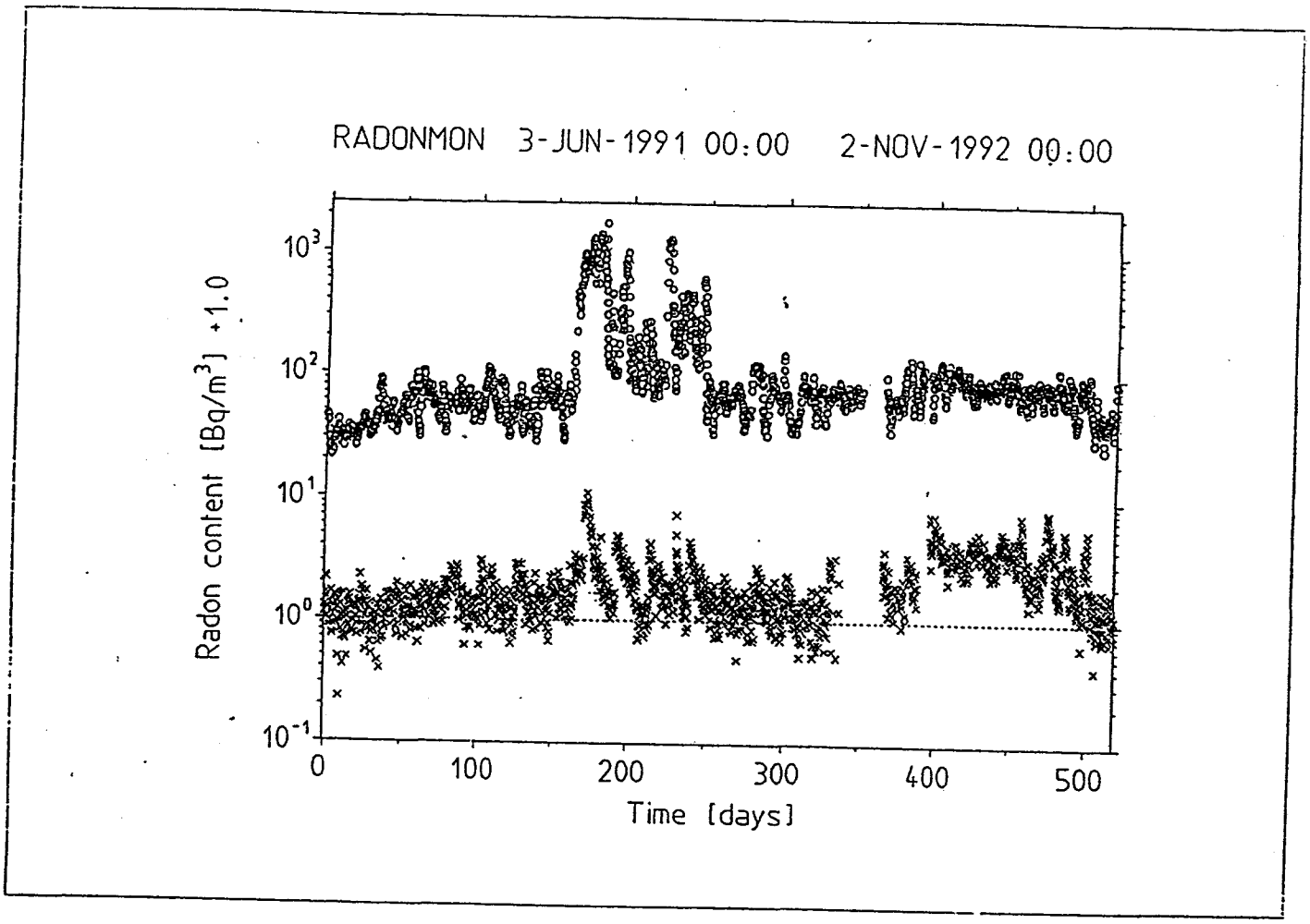
NIM B61 (1991) p.

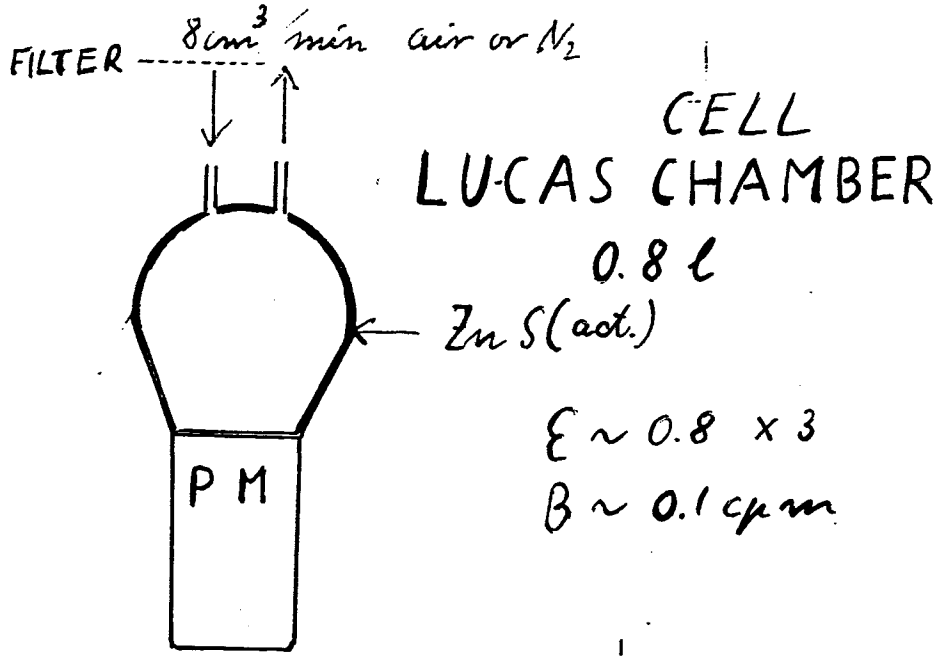
Table 1
The diffusion-, solubility-, permeability coefficients and diffusion lengths of Rn in some membranes

Material	Diffusion coefficient D [10^{-10} cm^2/s]	Solubility S	Permeability P [10^{-8} cm^2/s]	Diffusion length d_e [mm]
Rubber soft	1000	12	120	2.2
Butyl rubber	49	4.4	2.1	0.48
PU soft	408	5.6	23	1.4
PU hard	88	7.9	6.9	0.65
PVC soft	420	10	42	1.4
PVC hard	140	5.2	7.3	0.82
PA Supronyl	6.1	3.4	0.2	0.11
Plexi	6.2	8.2	0.5	0.11

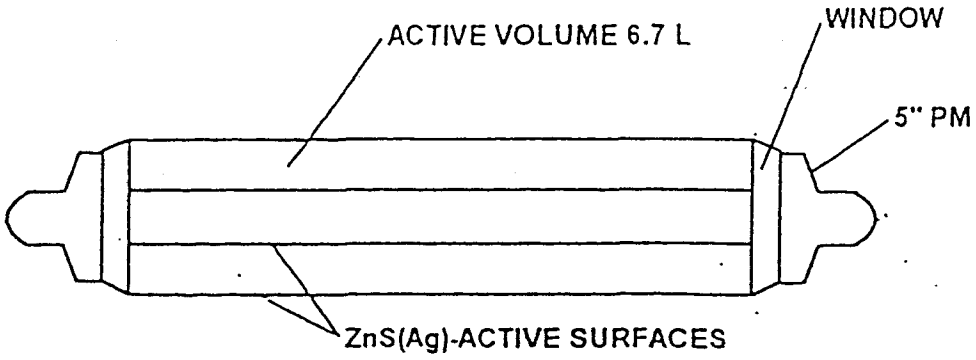

GASOMETER BALOON
INSIDE FARADAY CAGE
S_1
Z
X
Y
BLOWER
FLOW-METER
CO_2 TRAP
DRY COLUMN
V_{By}
V_M
INSIDE RADON -METER
SHIELDING TANK
B
C
P
MANOMETER
A
W
S_2
CIRCULATION PUMP
AIR PUMP
OUTSIDE RADON -METER
EVAPORATING N_2
CHARCOAL TRAP IN LN_2
N_2
GAS BOTTLE


GASOMETER BALOON
INSIDE FARADAY CAGE
S_1
Z
X
Y
BLOWER
SHIELDING TANK
INSIDE RADON -METER
AIR PUMP
OUTSIDE RADON -METER
N_2
GAS BOTTLE


RADONMON 3-JUN-1991 00:00 2-NOV-1992 00:00
Radon content [Bq/m³] +1.0
10³
10²
10¹
10⁰
10⁻¹
0
100
200
300
400
500
Time [days]

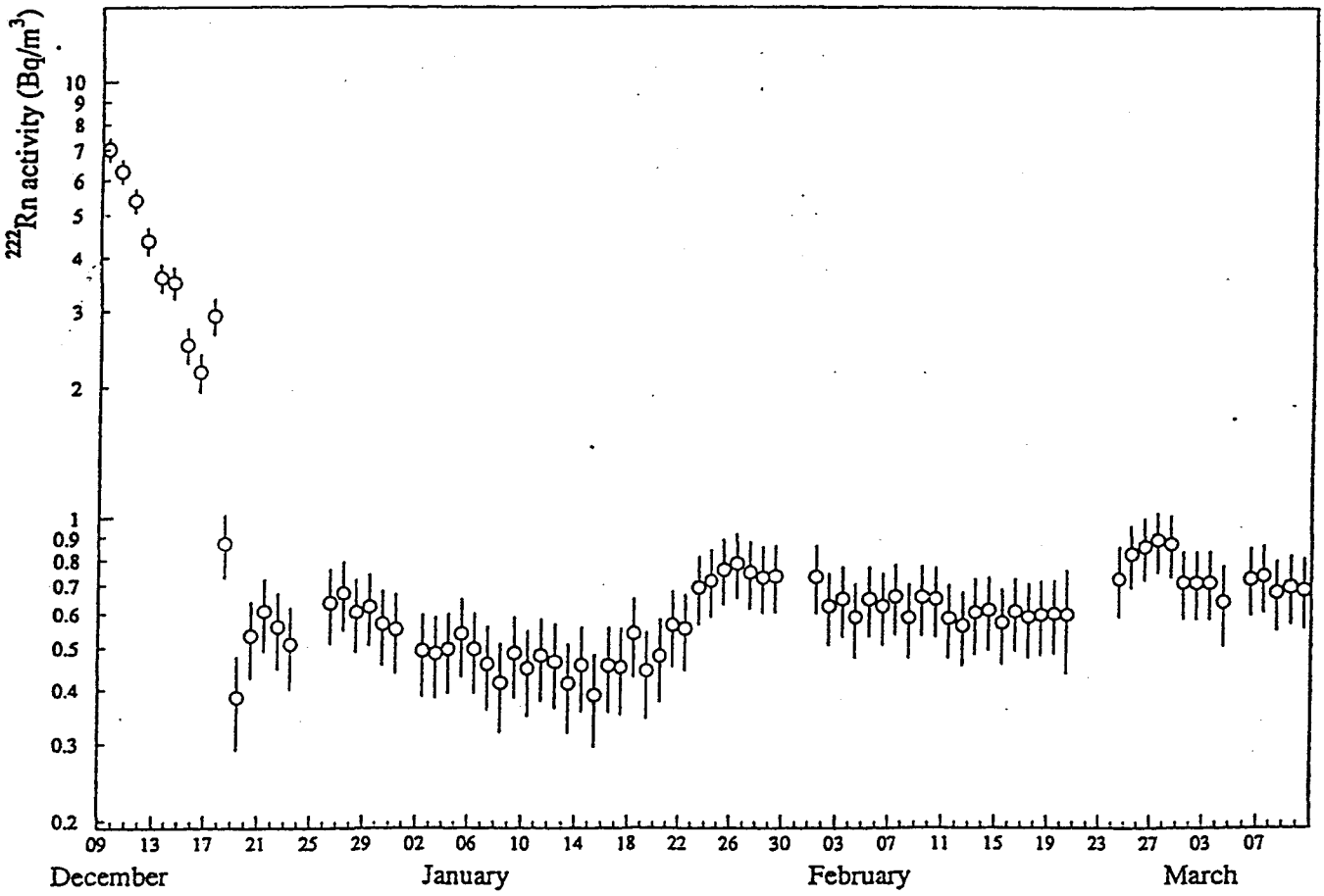

FILTER
8 cm³/min air or N₂
CELL
LUCAS CHAMBER
0.8 l
ZnS(act.)
PM
ε ~ 0.8 x 3
B ~ 0.1 cpm

coincidence Lucas chamber

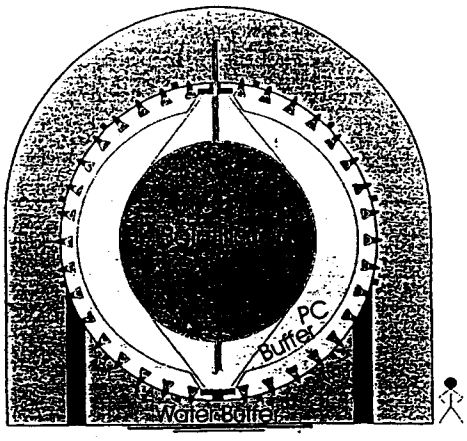


ϵ_{Rn} - 130% B = 0.3 cpm

MARCIN WOJCIK



RADON PROTECTION IN BOREXINO



1. MATERIAL SELECTION

by Rn measurements with preconcentration and proportional counting.

0.5 μBq/m³ in N_2
10 μBq/m² surface emanation

0.1 mBq/m³ in H_2O
50 μBq/kg emanation from Nylonfoil.

by Ge-γ spectrometry : *0.1m Bq ^{226}Ra/kg*

by permeation measurements of barrier foils: *$<10^{-12}$ cm²/s*

2. DETECTOR DESING AND SCINTILLATOR PURIFICATION

sealings of low permeability and low Ra content

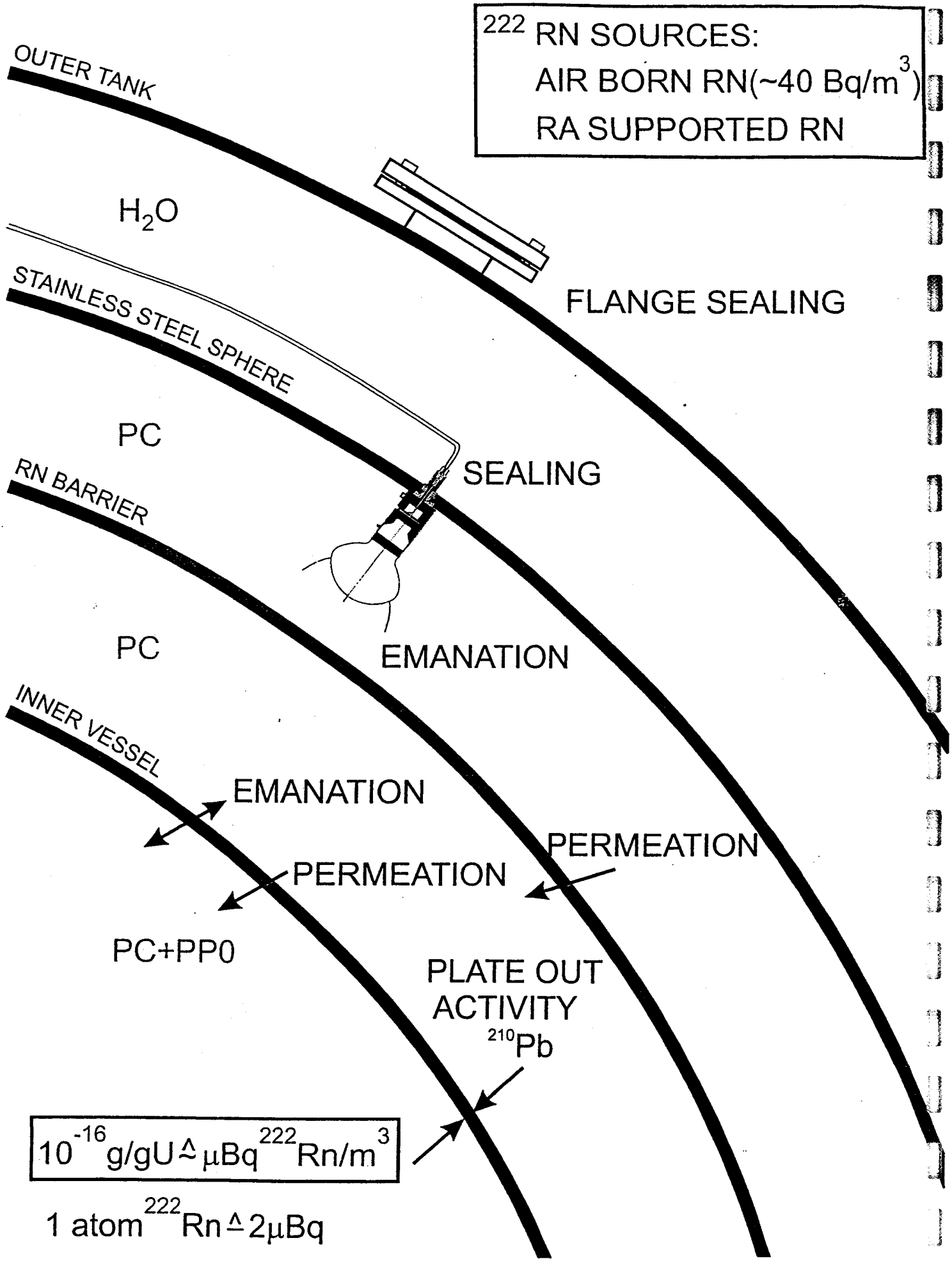
Rn barriers of Nylon foil in the inner detector

Rn filtered or synthetic air during inner vessel construction and installation

sparging of scintillator with high purity N_2 *(<μBq ^{222}Rn/m³)*

3. ACTIVE SUPRESSION

background cuts by tagging delayed coincidence events of the Th/U decav chaines in the detector.

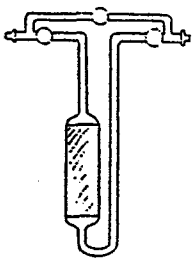

222 RN SOURCES:
AIR BORN RN(~40 Bq/m3)
RA SUPPORTED RN
OUTER TANK
H2O
STAINLESS STEEL SPHERE
FLANGE SEALING
PC
SEALING
RN BARRIER
PC
EMANATION
INNER VESSEL
EMANATION
PERMEATION
PERMEATION
PC+PP0
PLATE OUT
ACTIVITY
210Pb
10-16 g/gU ≙ μBq 222Rn/m3
1 atom 222Rn ≙ 2μBq

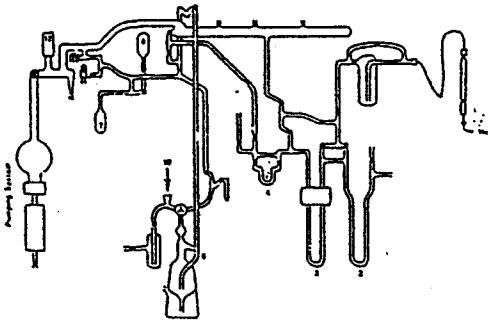
Measuring principle

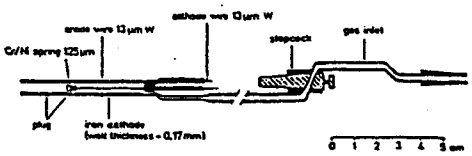
1
concentration

2
desorption
purification
transfer to counter

3
counting in
proportional counter







charcoal trap at -196° C
(gas chromatograhy)
$\alpha \sim 2.4 \times 10^7$

blank activity ⟷ ***sensitivity***
(radon leaks +Ra contamination)

metal sealed high vacuum technique

high purity charcoal (0.33 ± 0.05 mBq (Ra)/kg)

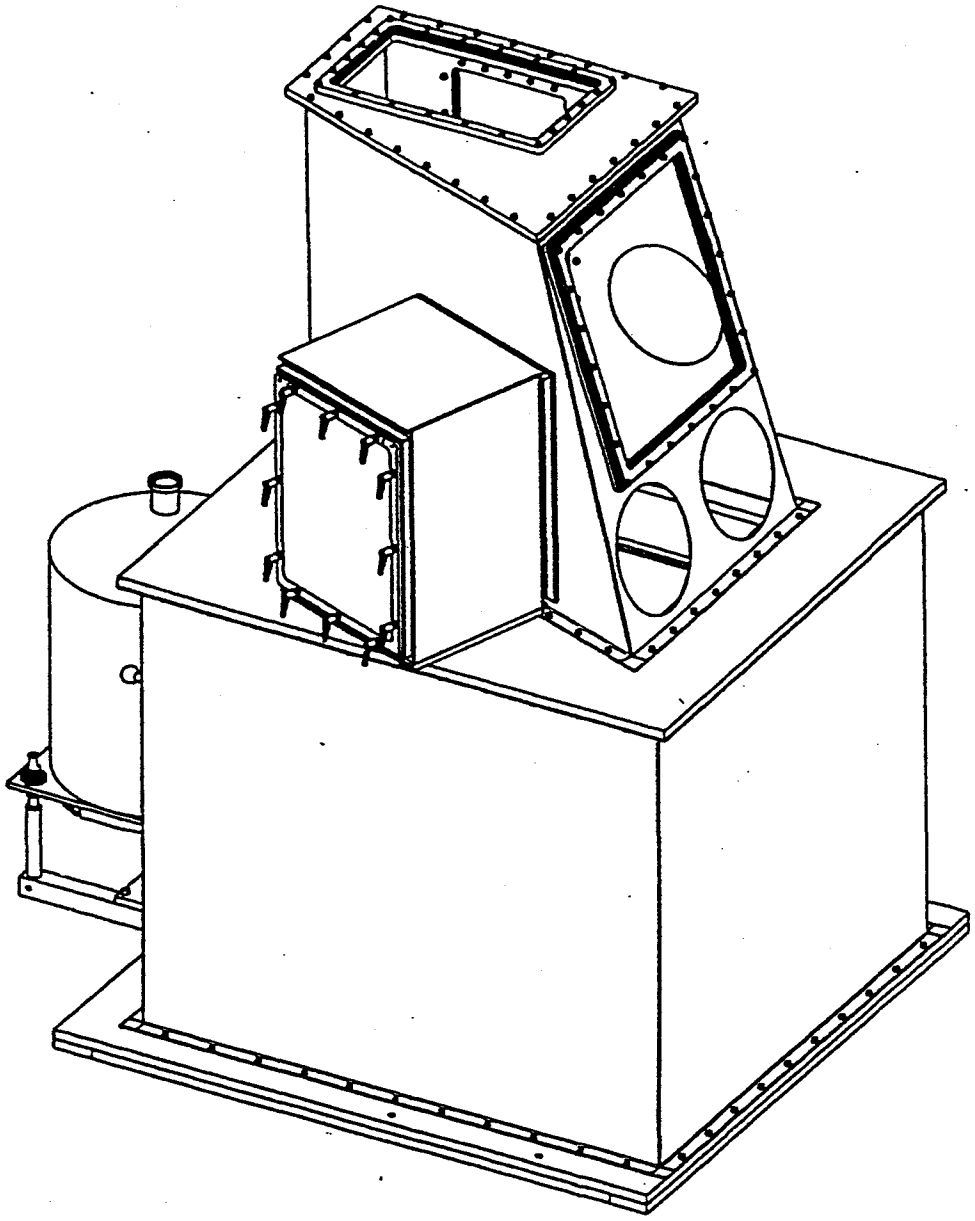
adsorption/desorption studies to optimize size of charcoal

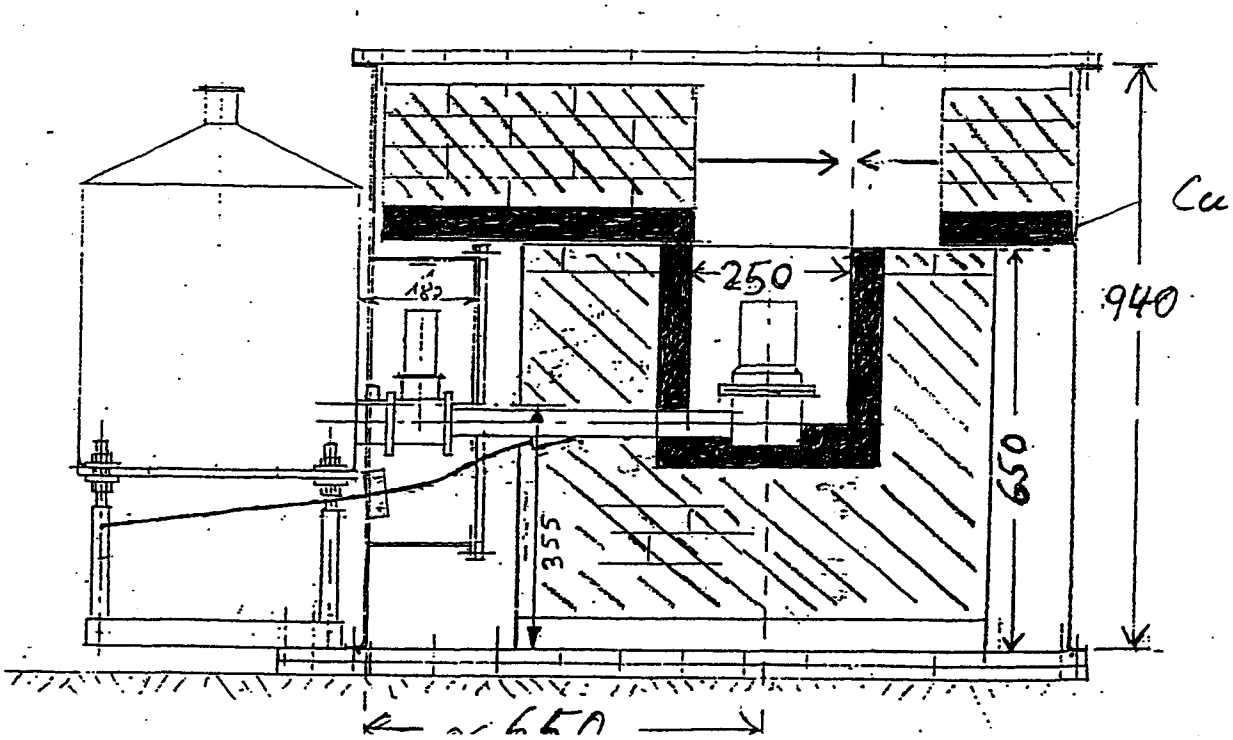
Radioimpurity [Bq/kg]

in charcoal

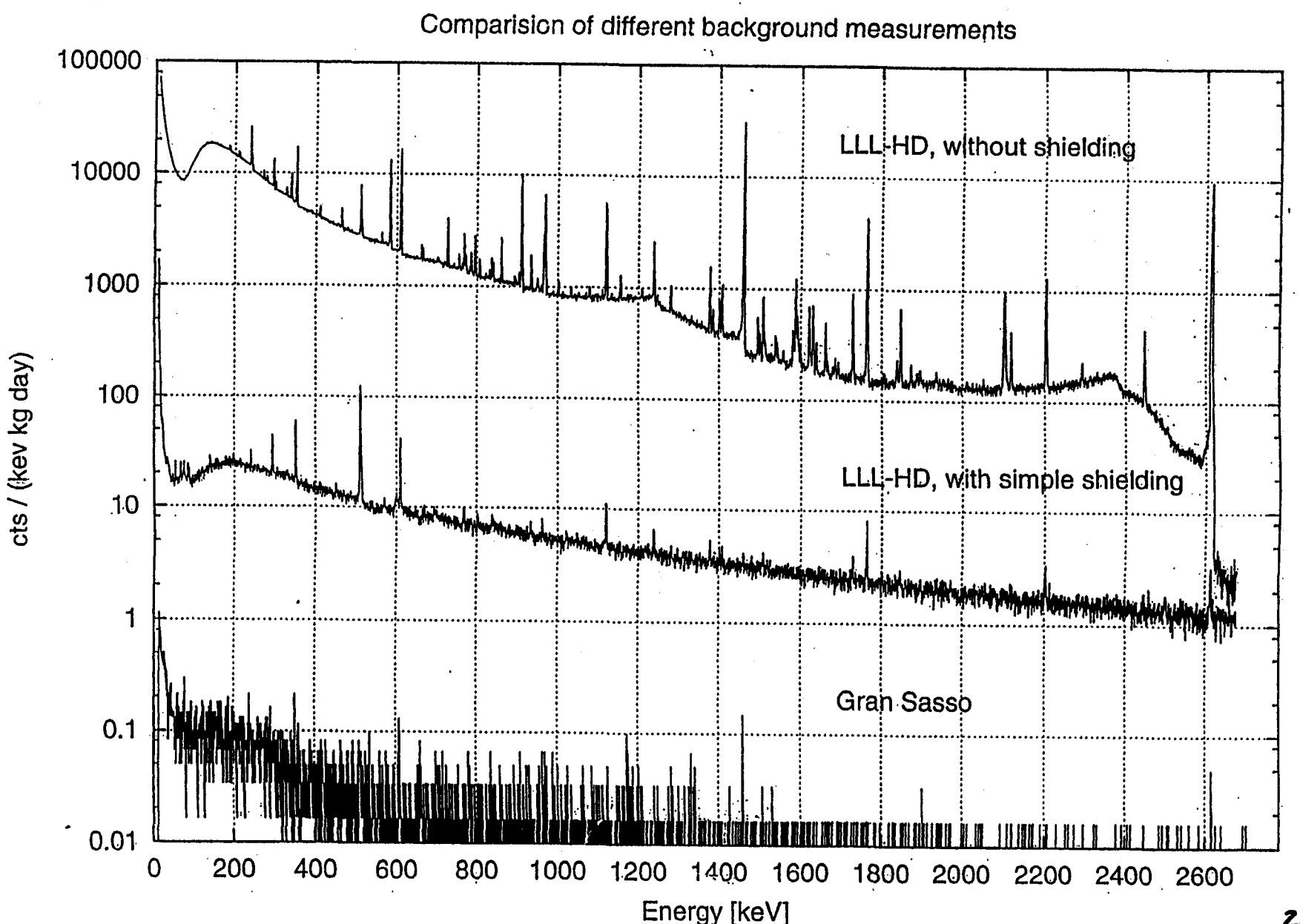
Type	^{137}Cs	^{228}Th	^{40}K	^{226}Ra	$^{222}Rn(Ra)$ *)
Silkarbon K48	<1	0.5	10	0.35	0.28±0.05
Silcarbon C46	1.2	1	380	1.0	
Silcarbon Sil40	<1	28	80	28	
Hydraffin UV43	3.4	0.7	1130	0.5	
Hydraffin CC8x30	1.3	1.2	275	1.0	0.33±0.02
ECN (CARBO ACT)	<0.5	<0.5	<2	<0.3	0.0003 ± 0.0001
Alcarbon 7x16	1.5	<0.4	690	<0.3	0.10±0.02
Alcarbon 12x20	0.1	<0.3	590	<0.3	0.17±0.02
Model PCB 616	5.3±0.3	0.18±0.12	120±7	0.37±0.09	
Model 1193	0.6±0.08	<0.25	360±20	0.20±0.1	

*) extracted from charcoal and measured




Cu
250
940
650
355
182

total >40 keV



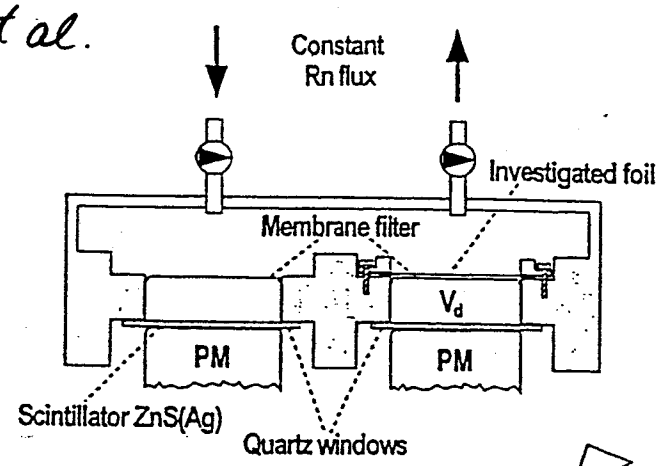
9015 cpm

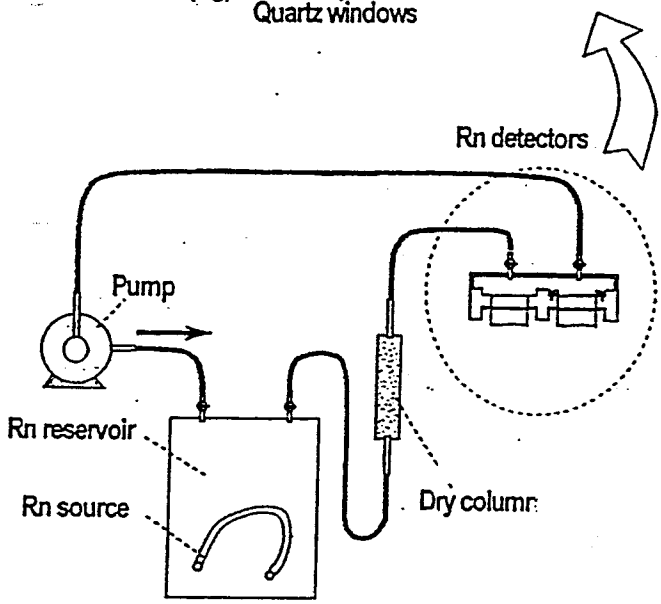
17.8 cpm

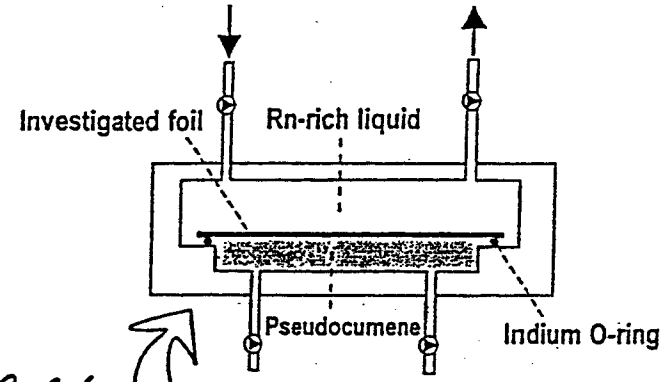
0.071 cpm

large sample capacity + low background ⇒ ~ 0.2 m Bq ^{226}Ra/kg

M. WOJCIK et al.



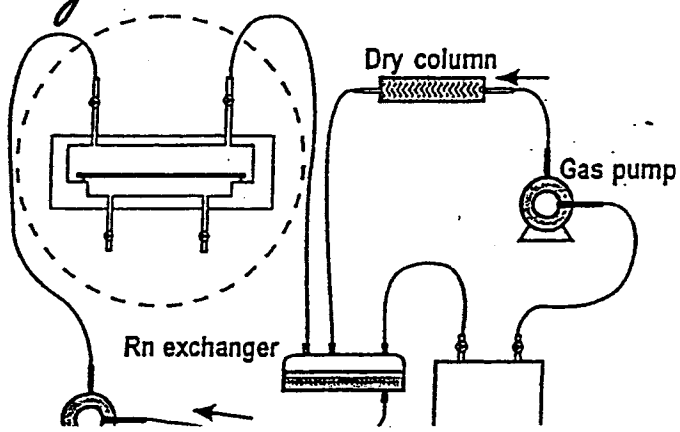




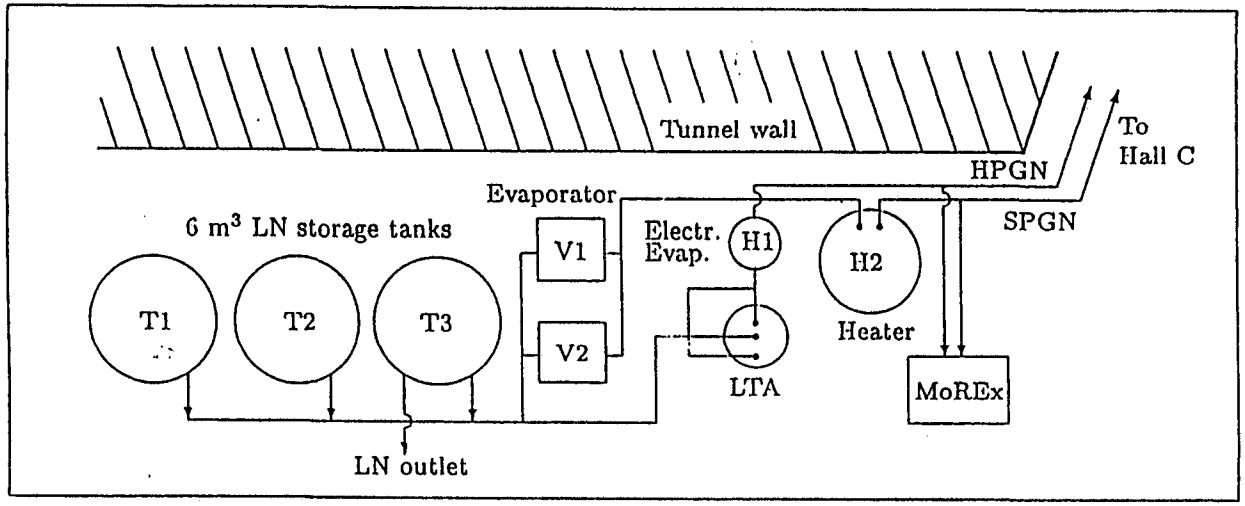
effective Permeability

Dry Nylon ~ 6×10^{-12} cm²/s

Wet Nylon ~ 1×10^{-9} cm²/s



Nitrogen Plant



R N_2 ($< 100\ \mu Bq/m^3$ Rn) 250 m^3/h

HP N_2 ($\leq 1\ \mu Bq/m^3$ Rn) 100 m^3/h

SYNTHETISCHE LUFT ($\sim 1\ mBq/m^3$ Rn) 200 m^3/h

Rn-222 Measurements with the Counting Test Facility (CTF) of BOREXINO

(St. Schönert, MPI-Hd, Borexino-Colla

A) Internal Rn-222 contamination:
(Rn in liquid scintillator)

- If Rn decays away: build-up of Pb-210 (→ Bi-210 → Po-210)
- If const. Rn decay rate: diagnostic: for Ra-226 (and U-238, if in equil.

Borexino requirement:

U-238 (in equil. with daughters) $\lesssim 10^{-16}$ g/g

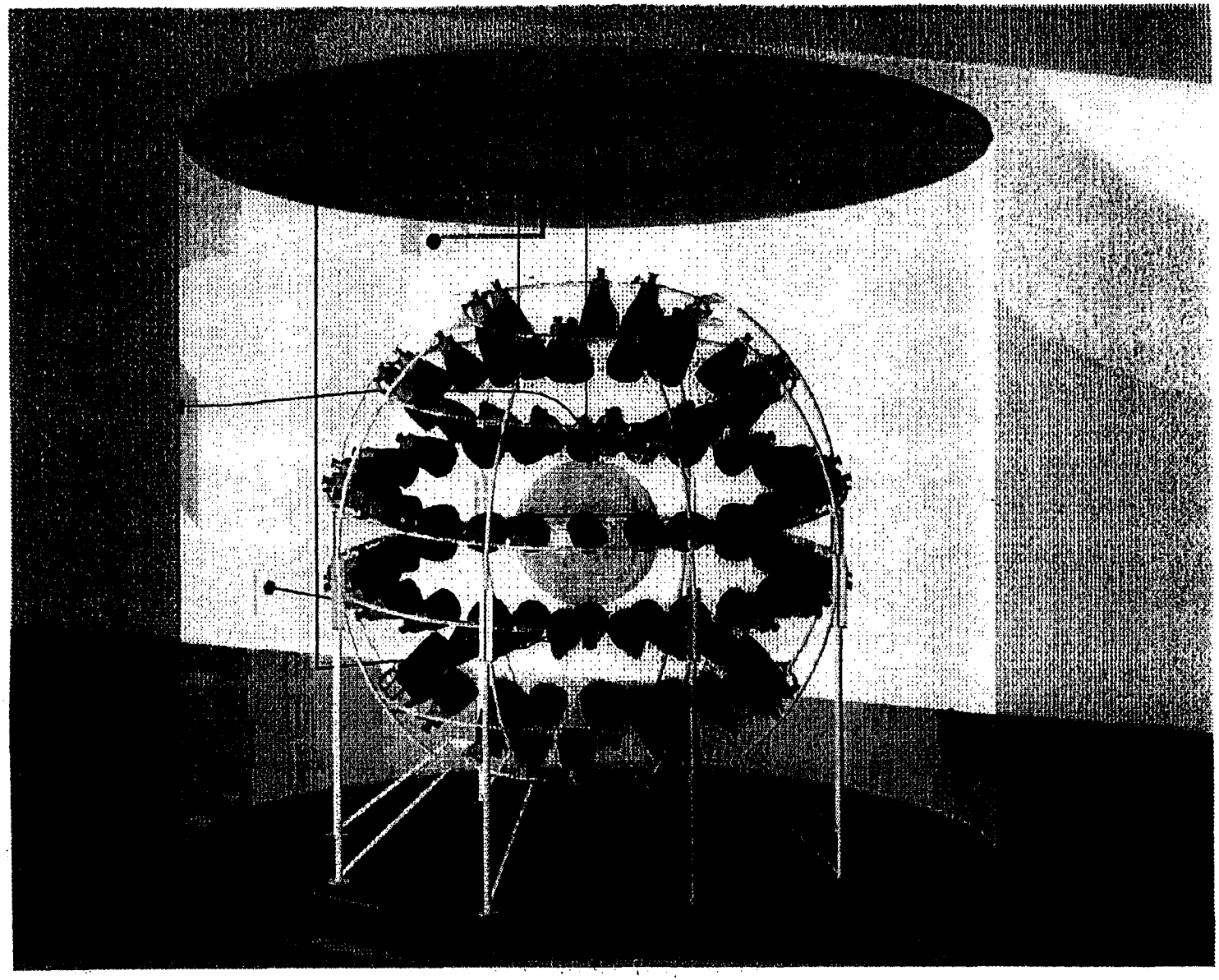
$\hat{=}$ 1.2 μBq / 10^3 kg Rn-222 activity

$\hat{\approx}$ 0,1 decays / d / 10^3 kg

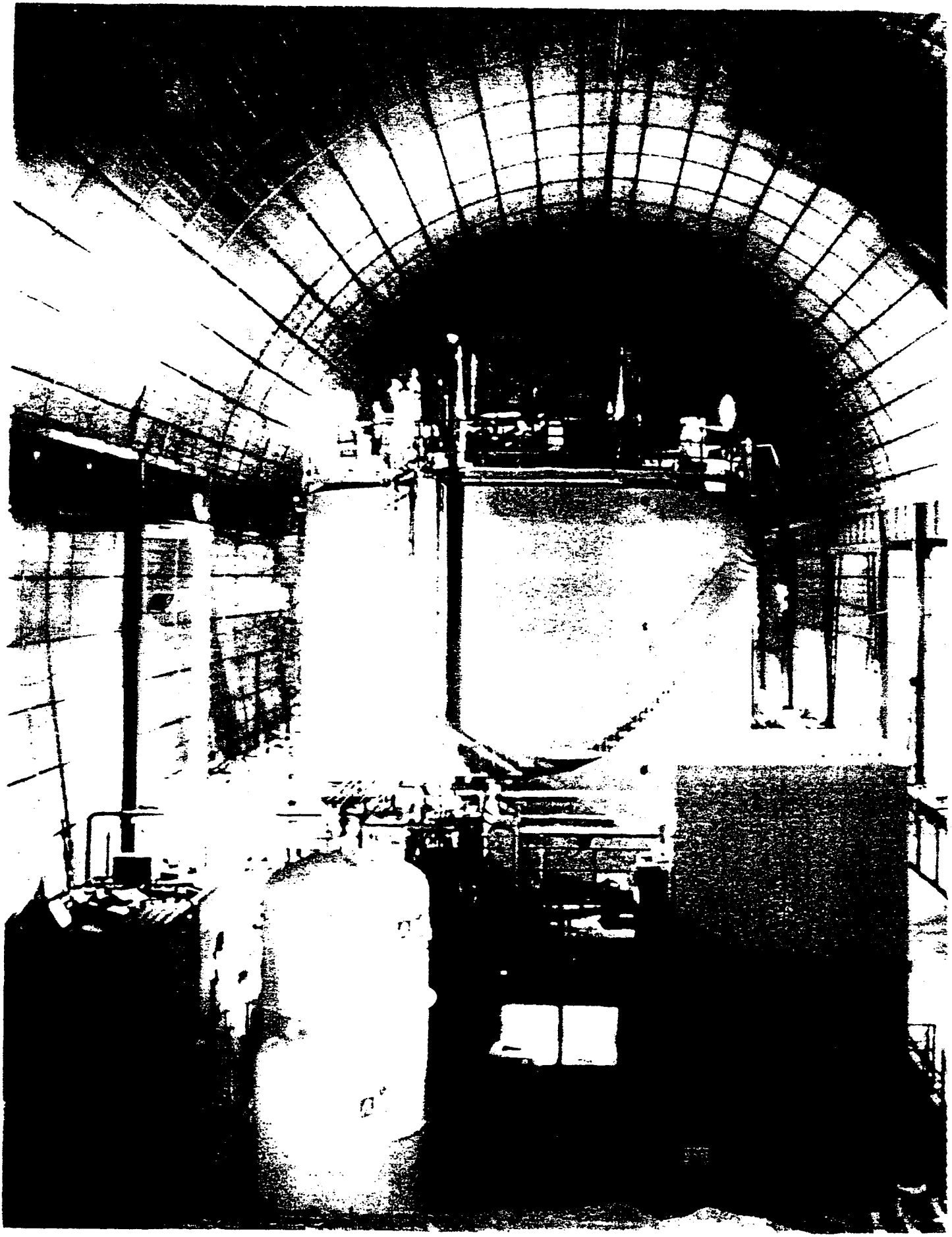
B) External Rn-222 contamination: (Rn in bu

- γ's from Bi-214
- Rn diffusion through membranes (if Rn

BX requirement: $\mathcal{O}$ (1 mBq / m^3)



1000t H_2O 100 PM's



Summary

- Radon is a dominant BG for the current very low-energy solar ν data in Super-Kamiokande.

➡ **Need further radon reduction!!**

- Real-time Rn monitoring has been carried out at SK site by using several 70L and 950L radon detectors.

Radon concentration in

purified air:	2~3mBq/m^3	
purified water:	10~20mBq/m^3	(re-estimated, preliminary)
air in SK tank:	10~20mBq/m^3	
water in SK tank:	1~2mBq/m^3 (upper half)	
	~5mBq/m^3 (bottom part)	

- In order to reduce radon in purified water, a hollow fiber membrane degassing module was tested.

➡ **Succeeded to remove ~93% of remaining radon by the test module**

Future plan

- Locate radon source in water purification system (~summer, 2000)
- Install hollow fiber membrane degassing system

Sudburry
Rn workshop
14 - June - 2000

Radon off-line monitoring program in BOREXINO, some general remarks

G. Heusser

Max-Planck-Institut für Kernphysik

Heidelberg

Detection of ^7Be ν (861 keV) by elastic ν-e^- scattering $\boxed{\nu + e^- \longrightarrow \nu + e^-}$

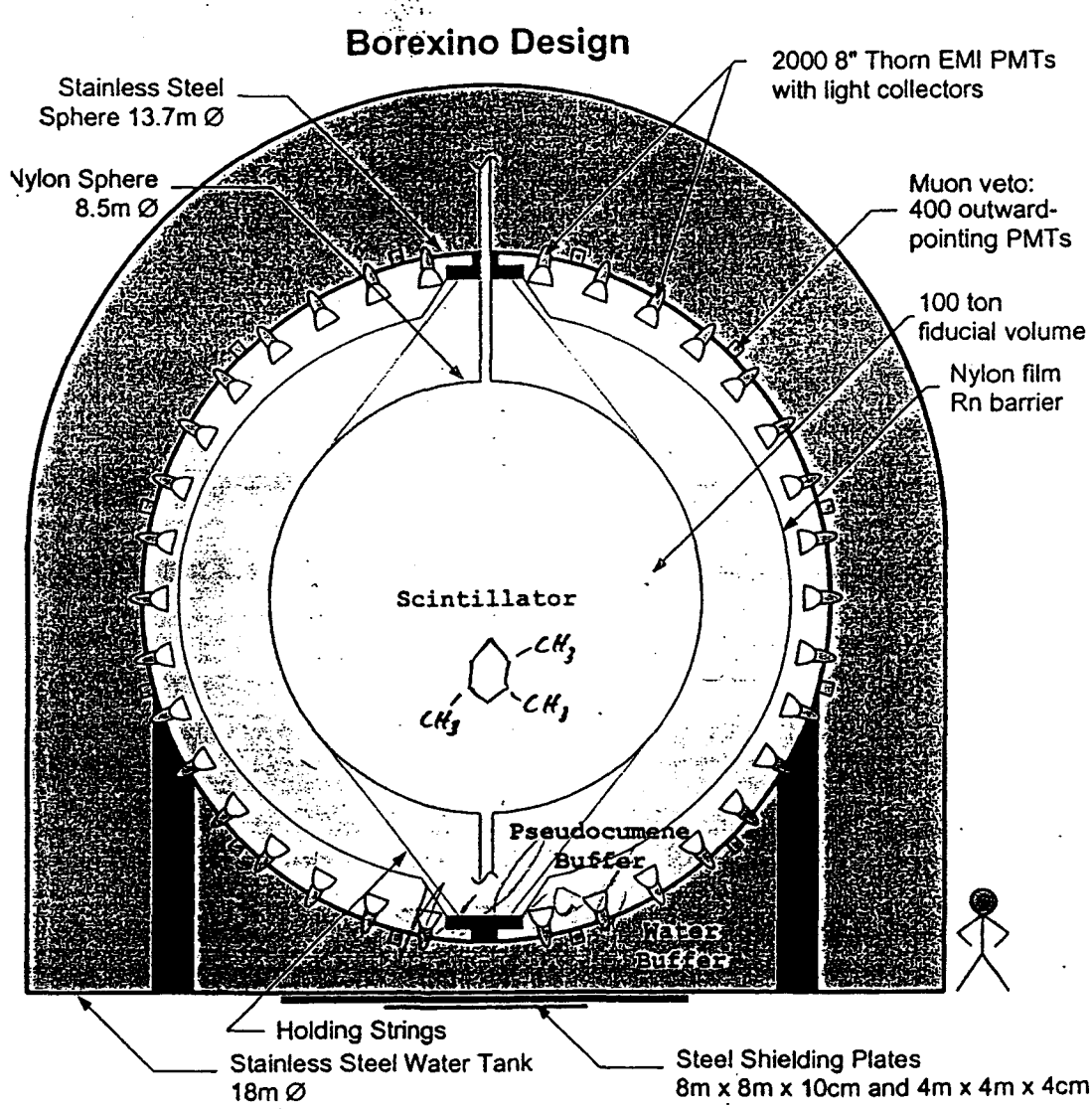
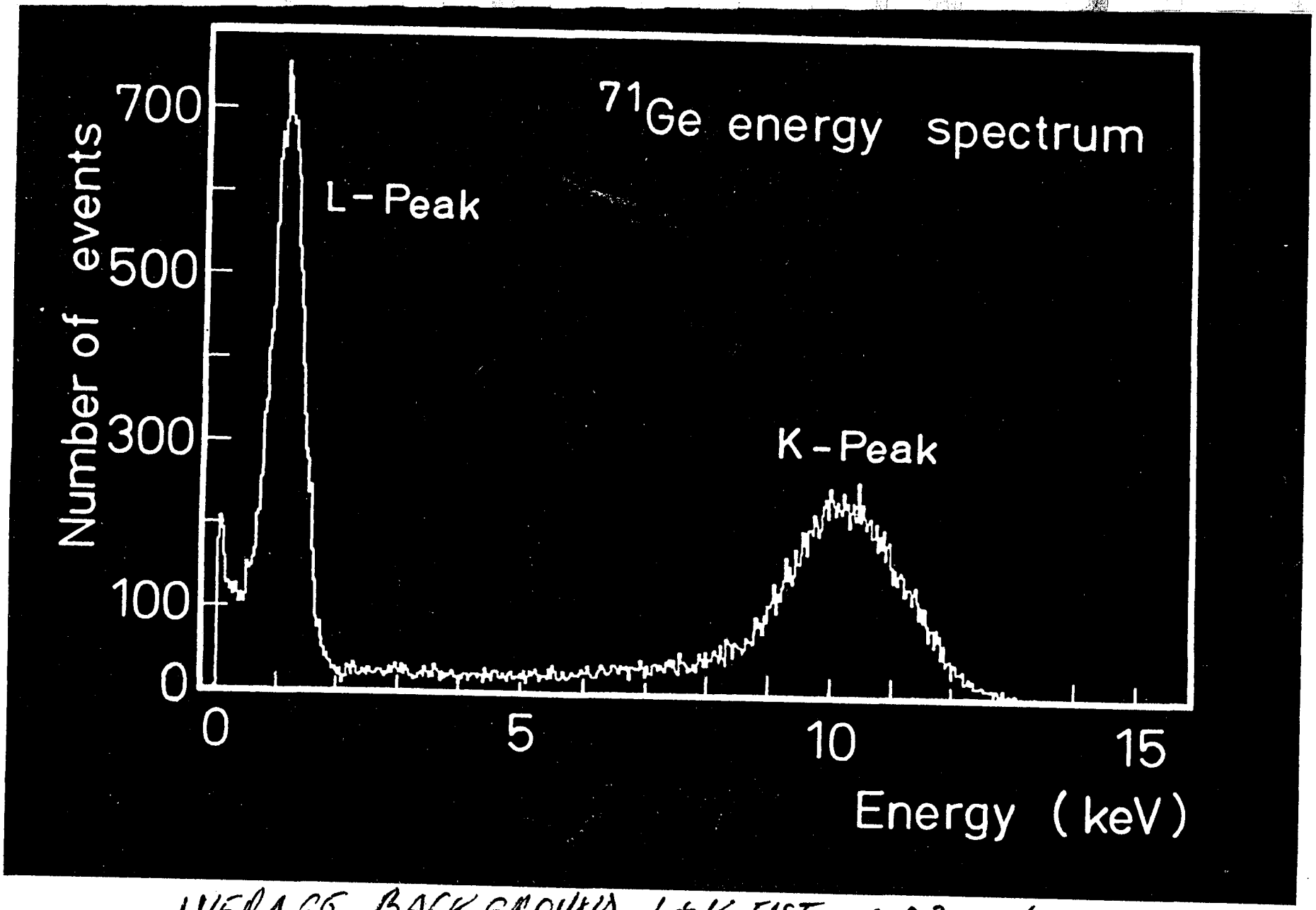


Figure 6: A schematic view of Borexino, showing the inner nylon vessel, the stainless steel sphere with photomultiplier tubes, and the outer water container.

required radiopurity of scintillator (C_9H_{12}):

$U \leq 10^{-15}$ g/g ✓✓; $Th \leq 10^{-15}$ g/g ✓✓; $^{14}C/^{12}C \leq 10^{-18}$ ✓



AVERAGE BACKGROUND L+K FAST: 0.07 cpd

NAL RN (~0.5 Bq/m³) INDUCED BACKGROUND: ~0.002 cpd active side

~0.00008 cpd passive side

Radon Background in Rare-Event Experiments
Satellite workshop of NEUTRINO 2000
Sudbury, Canada

June 14, 2000

Radon recognition and correction in ^{71}Ge proportional counting

W. Hampel

for the

GALLEX and GNO Collaborations

H. Lalla, Ph.D. thesis (GALLEX)

J. Handt, Ph.D. thesis (GNO)

Problem: In the course of counter filling a few (unwanted) ^{222}Rn-atoms are introduced into the proportional counter.

Decay of these ^{222}Rn atoms and its daughter nuclides produce events that (on the basis of single events) are not distinguishable from ^{71}Ge events

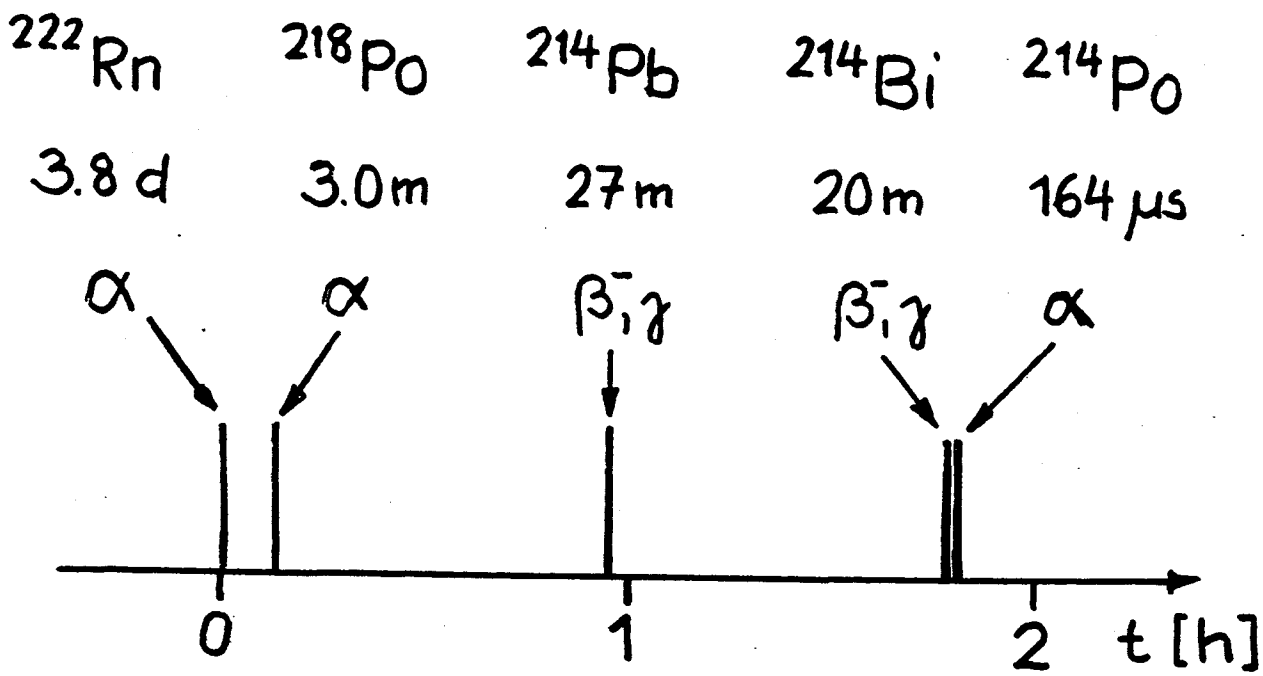
Solution: To avoid problem totally is not possible in practice

Therefore: recognize Rn events and correct their effects

→ **Radon cut**

A special cut for these Rn events has been developed in GALLEX and is applied in both GALLEX and GNO

Radon in the proportional counter



<u>Decay of one ^{222}Rn atom:</u>

~ 2.6 events total

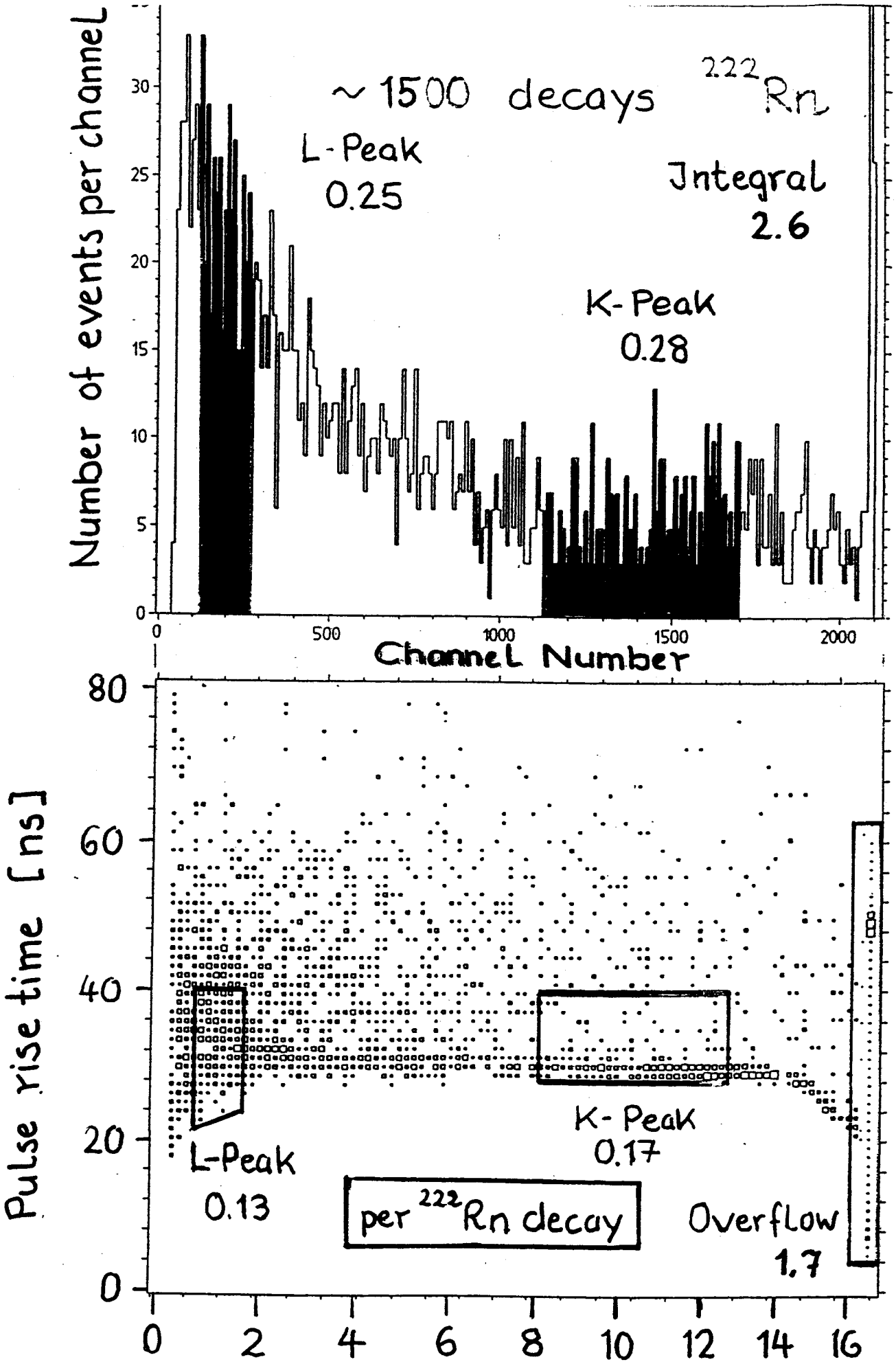
~ 1.7 overflow events (> 16 keV)

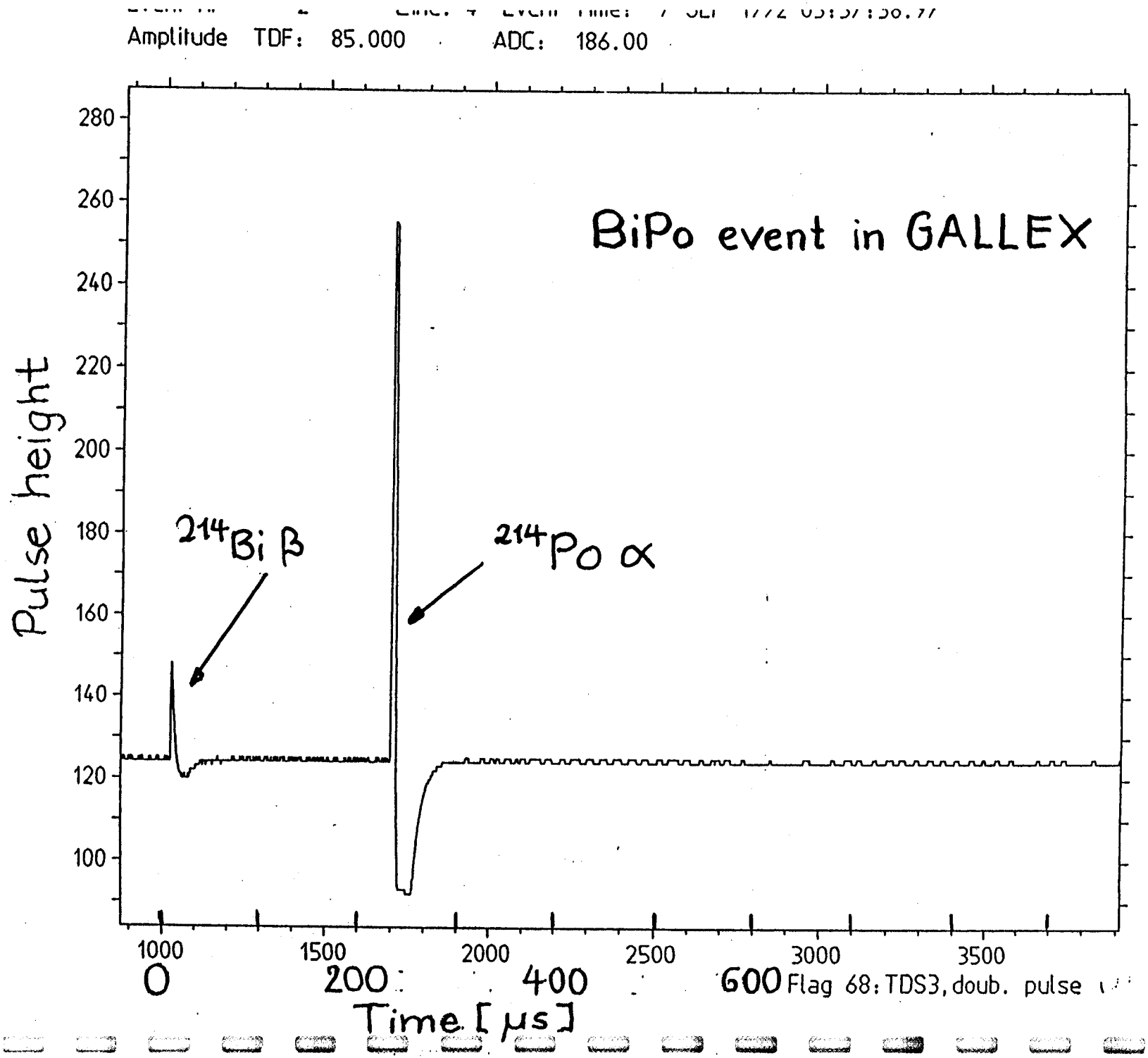
~ 0.3 events in ^{71}Ge windows (fast L or K event)

→ Radon cut

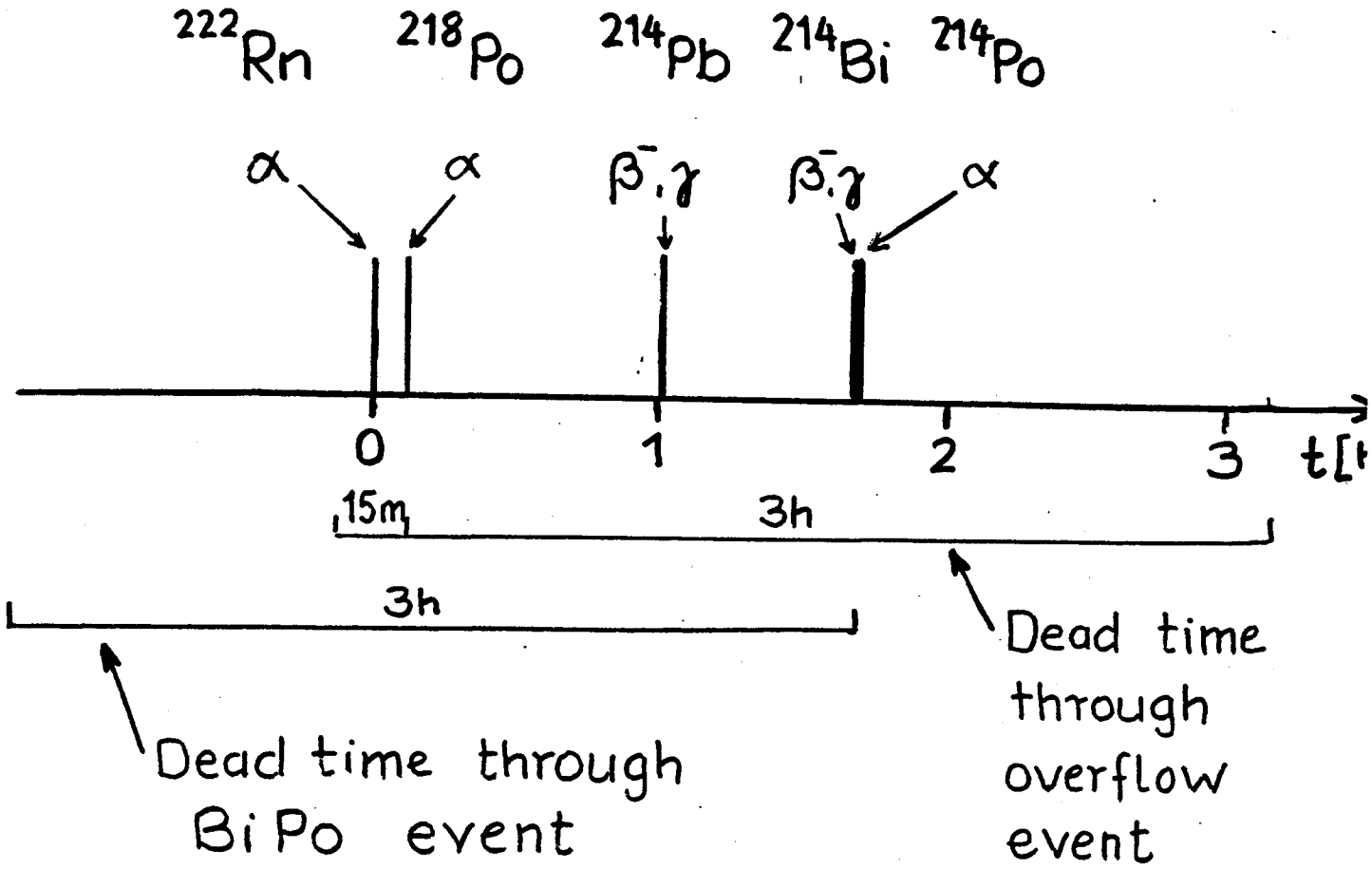
Dead time: 15 m before, 3 h after } an event with E > 16 keV

3 h before a BiPo event


~ 1500 decays
222Rn
L-Peak
0.25
Integral
2.6
K-Peak
0.28
Number of events per channel
Channel Number
Pulse rise time [ns]
L-Peak
0.13
K-Peak
0.17
per 222Rn decay
Overflow
1.7


Amplitude TDF: 85.000 ADC: 186.00
BiPo event in GALLEX
Pulse height
280
260
240
220
200
180
160
140
120
100
^{214}Bi β
^{214}Po α
1000
1500
2000
2500
3000
3500
0
200
400
600
Time [μs]
Flag 68: TDS3, doub. pulse

Radon cut in GALLEX and GNO



Efficiency of the Radon cut : (91 ± 5) %

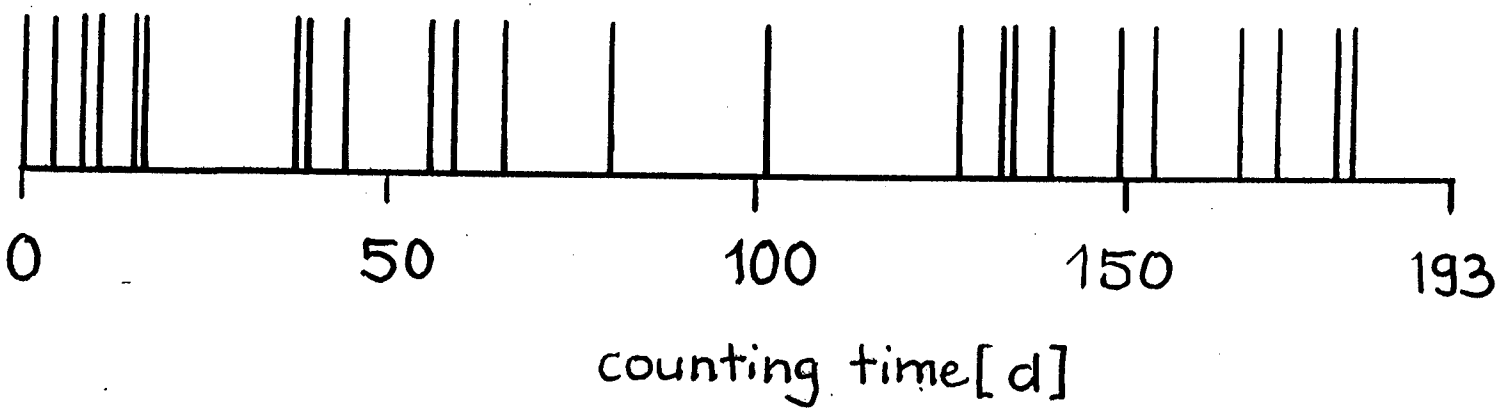
(from measurement and Monte Carlo simulations)

Radon cut inefficiency : 9 %

8 % : neither ^{222}Rn nor ^{218}Po produce an overflow event (E > 16 keV)

1 % : L or K event in ^{71}Ge window is produced later than 3 hours after the overflow event

Solar neutrino-Run A71



Measured: 24 Maximum Likelihood: 5.0 + 19.0 = 24.0

^{71}Ge Bkg

75 SNU

||

Without Radon-Cut

measured: 26 Maximum Likelihood: 7.2 + 18.8 = 26.0

103 SNU

Radon in GALLEX Counting

~ 3.8 ^{222}Rn atoms in counter at start of counting (average of 65 runs)

Efficiency of a ^{222}Rn atom to produce a count in the ^{71}Ge windows : ~ 0.3

1.15 counts due to ^{222}Rn

No Rn correction	102.4 SNU
With Rn cut	79.5 SNU
Rn cut inefficiency correction	- 2.2 SNU
Solar neutrino signal	77.3 SNU

Difference 102.4 - 77.3 = 25.1 SNU

Source of ^{222}Rn atoms

Not yet clearly identified

Clear : Rn must come from the final part in counter filling (after the GeH_4 synthesis and cleaning)

Determination of the Radon cut efficiency

1) High statistics:
   a few 1000 ^{222}Rn decays required

2) Activity:
   ~ 4-5 ^{222}Rn decays per day

3) Careful determination of counter background

GALLEX:

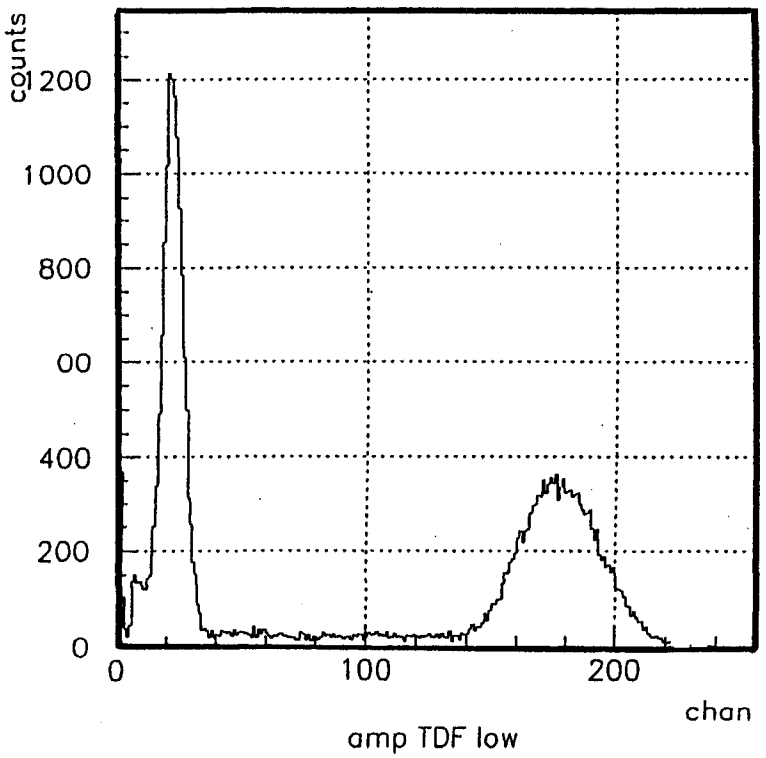
Combination of measurements at higher rate with Monte Carlo simulations

GNO:

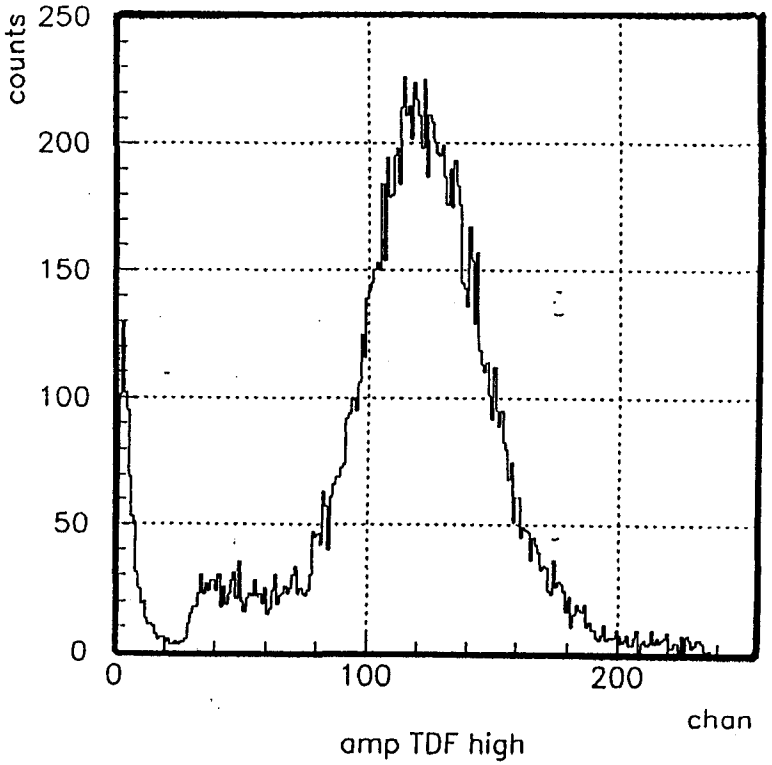
Long term measurement (> 1 year) at low rate (see above)

Gran Sasso / counter Si119 30000 events ^{71}Ge

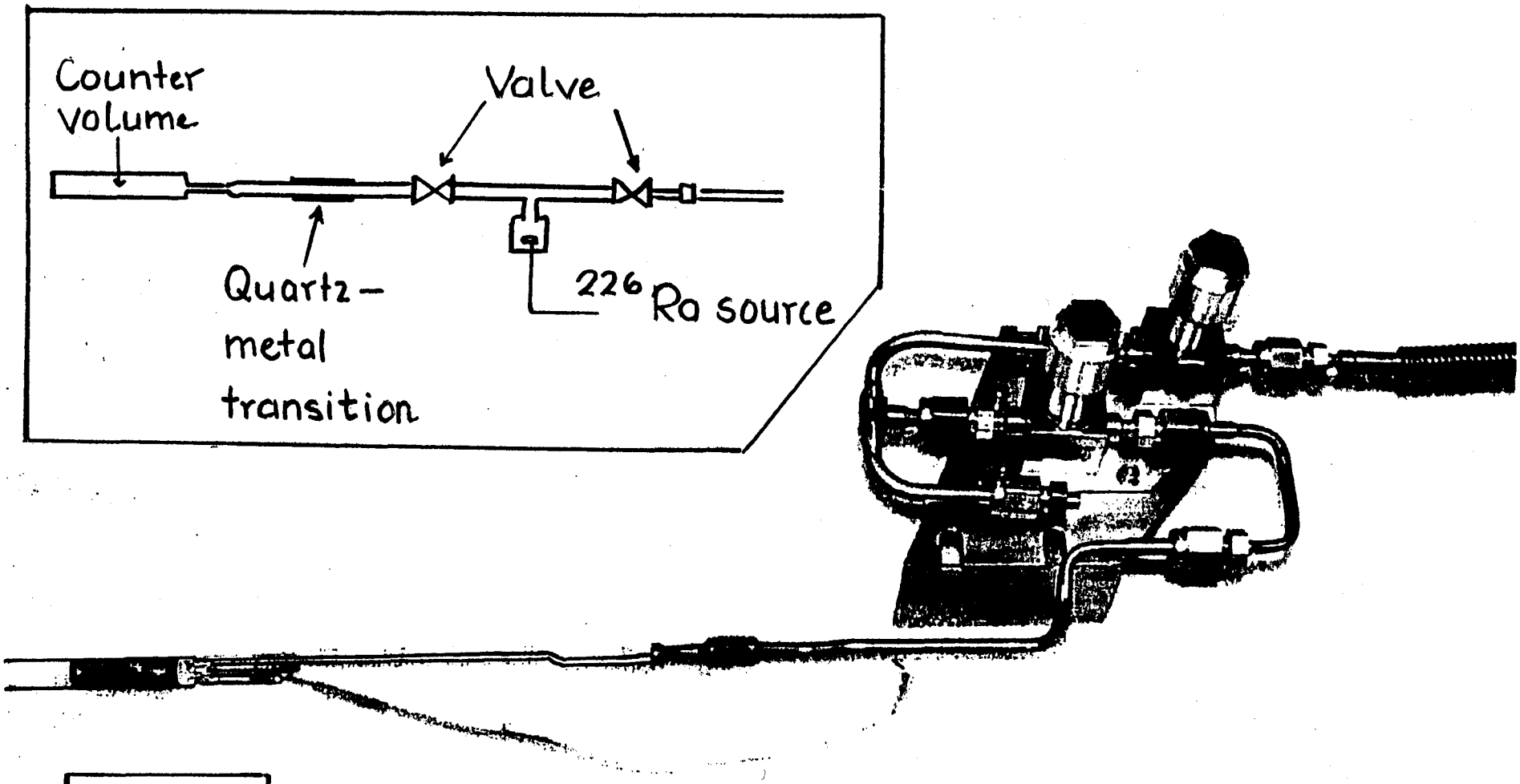
run1126.cali.0262



low amplification TDF

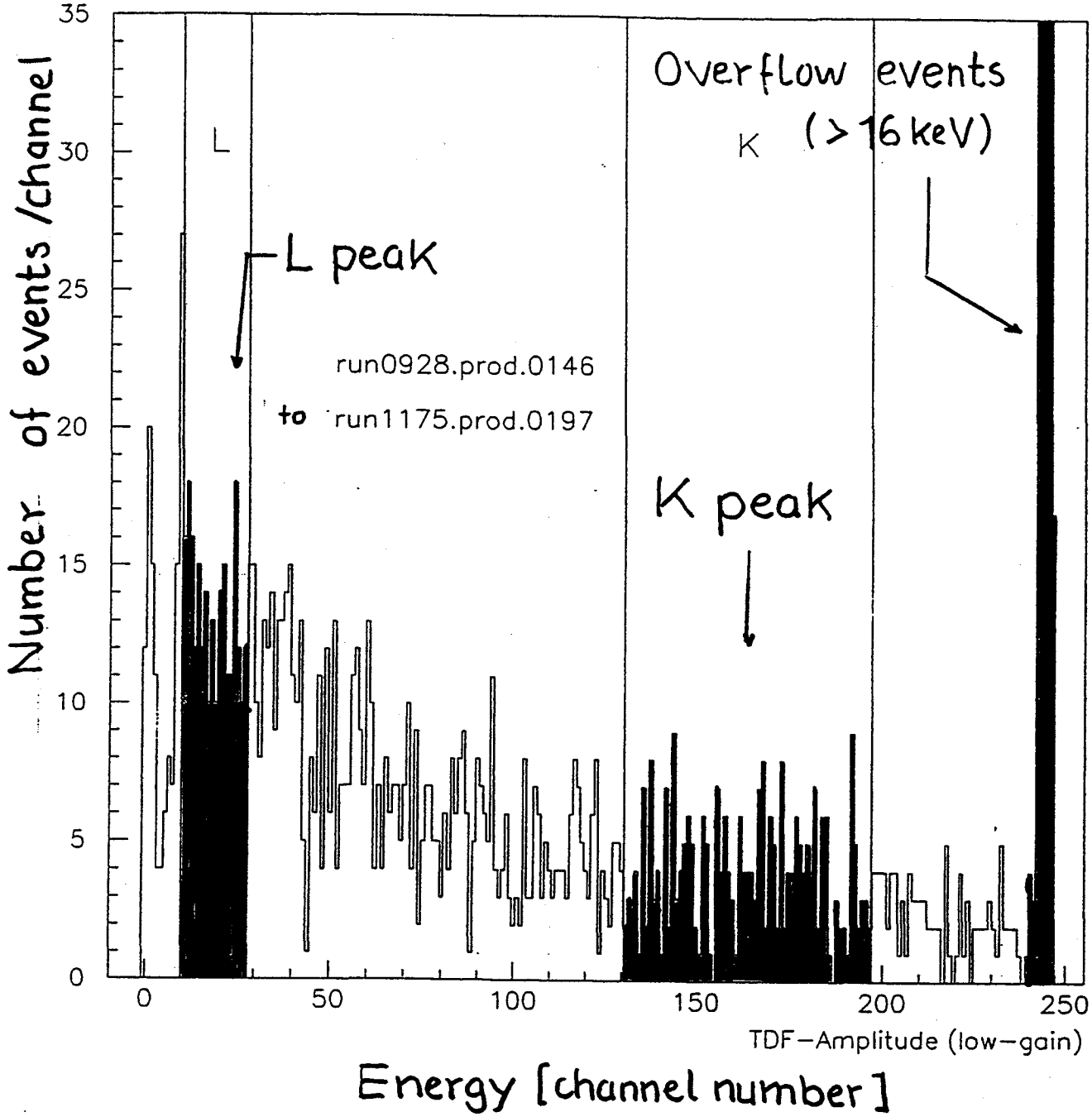


high amplification TDF


Counter volume
Valve
Quartz – metal transition
226 Ra source

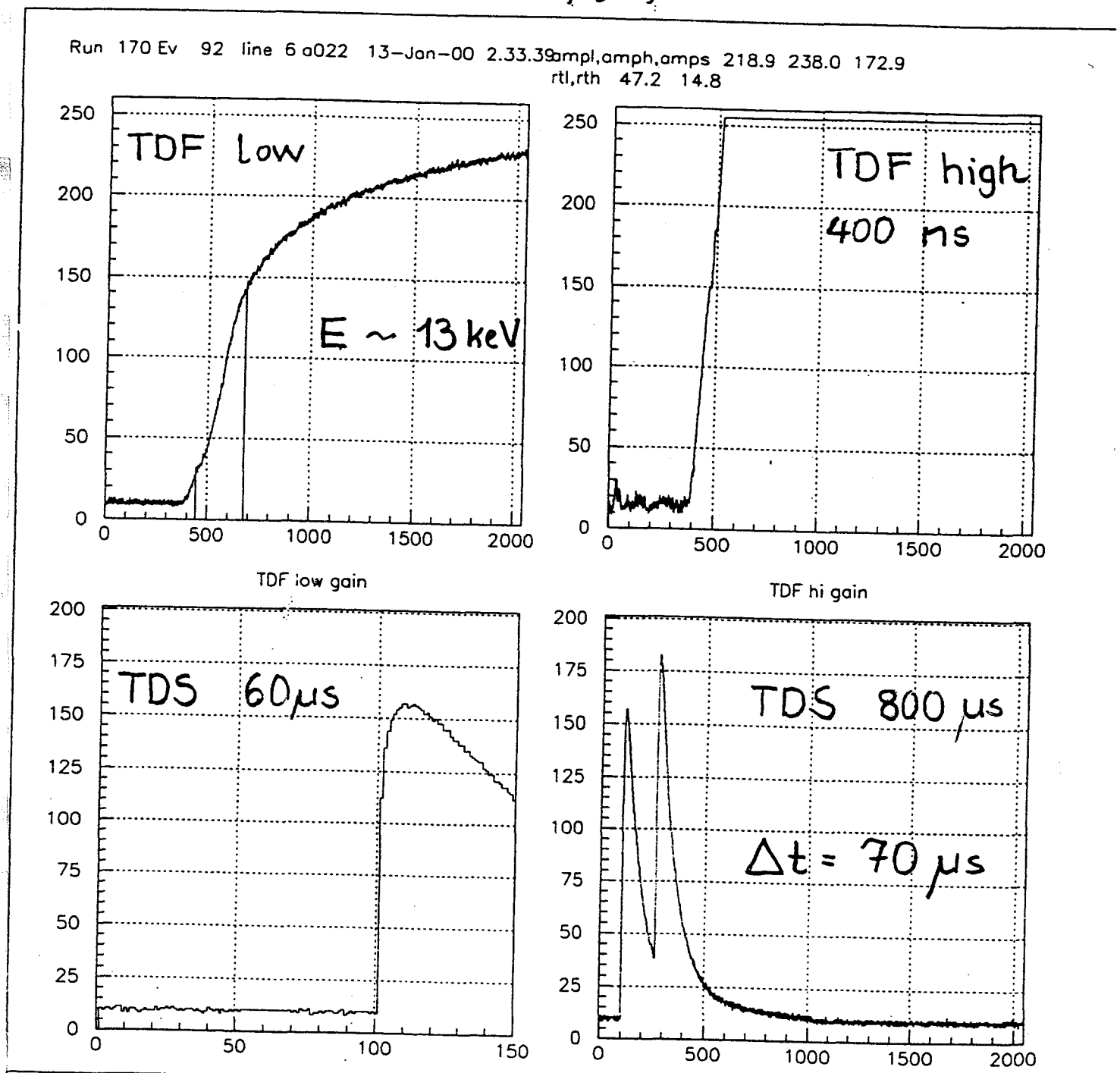
Fe 103

Radon measurement in GNO

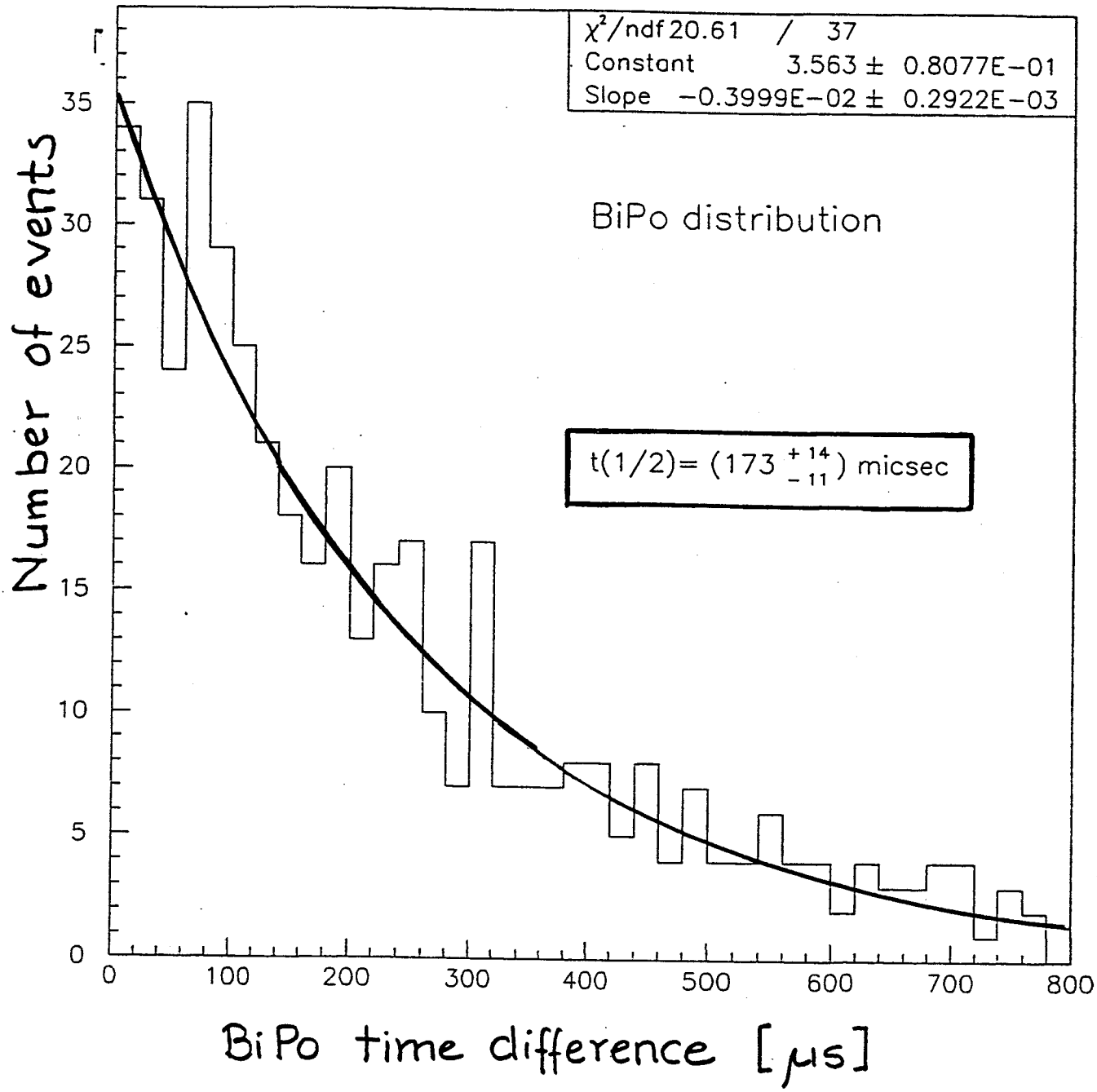


BiPo event GNO (A022)

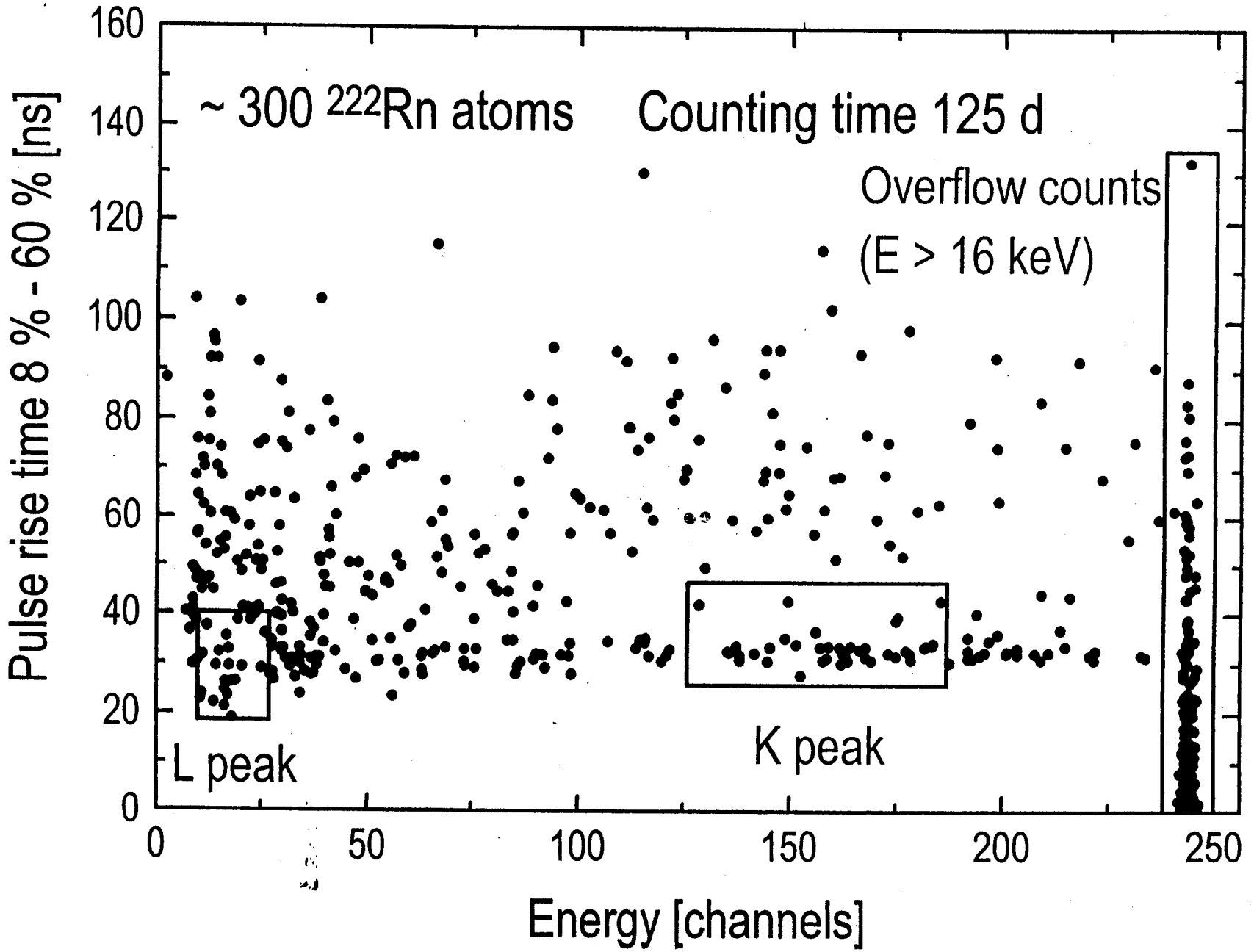
(170, 92)



Run 170 Ev 92 line 6 a022 13-Jan-00


χ²/ndf 20.61 / 37
Constant 3.563 ± 0.8077E−01
Slope −0.3999E−02 ± 0.2922E−03
BiPo distribution
t(1/2)= (173 +14 −11) micsec
Number of events
BiPo time difference [μs]
0
5
10
15
20
25
30
35
0
100
200
300
400
500
600
700
800

Radon measurement in GNO



Radon in ^{71}Ge counting of GNO

Preliminary results:

- Low activity ^{222}Rn experiment

  observed ~ 1500 ^{222}Rn decays

- Radon cut efficiency

  GALLEX (91 ± 5) %

  GNO (95.5 ± 13.5) %
  very preliminary !

- Average number of ^{222}Rn atoms in counter

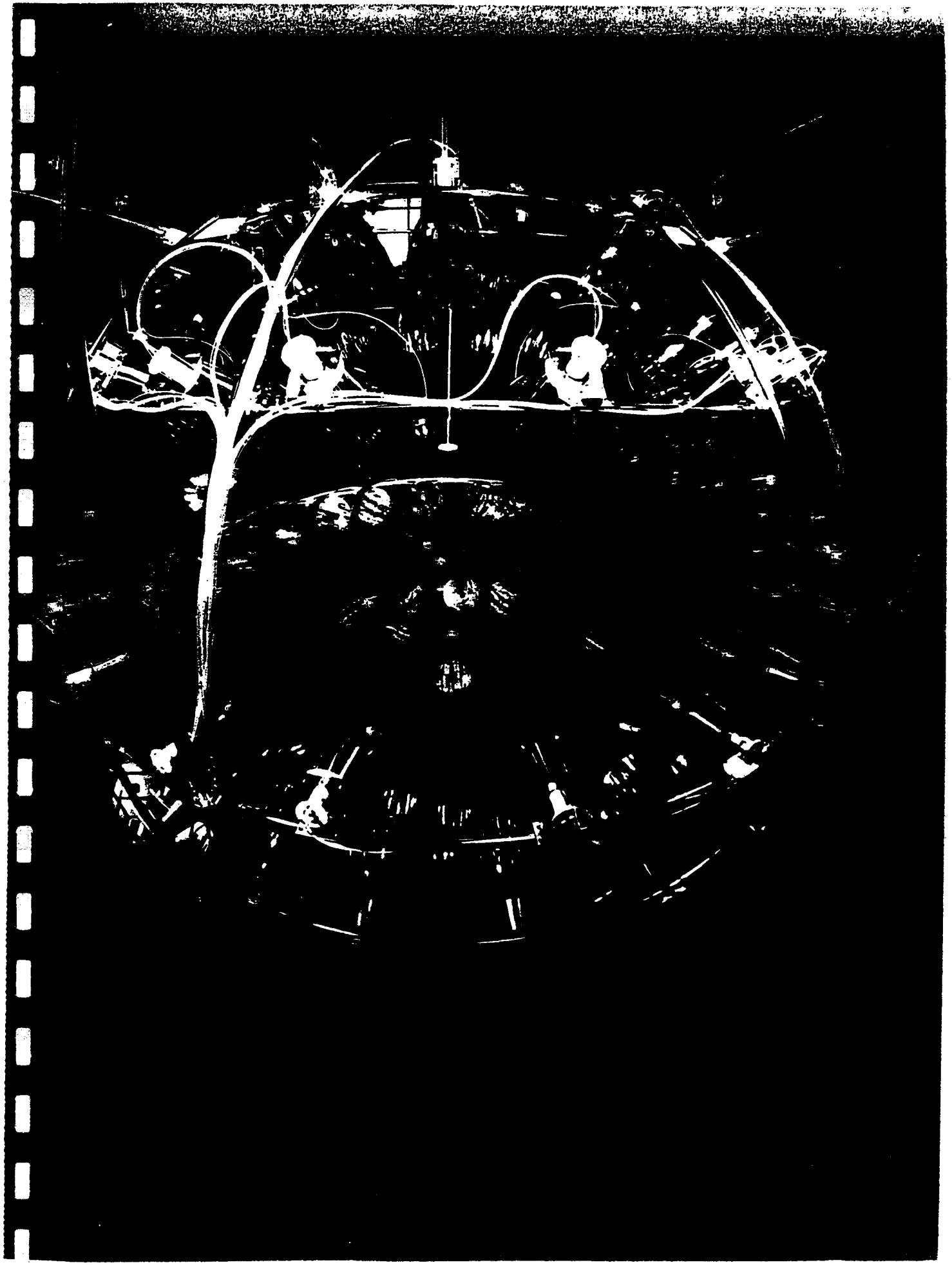
  GALLEX (65 runs) 3.8

  GNO (19 runs) 1.6

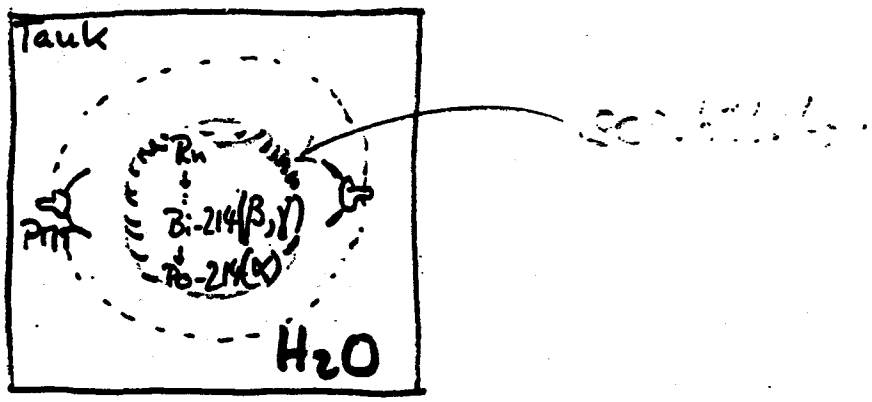
- Difference due to Radon [SNU]

  GALLEX 25.1

  GNO 10.6



Measurement of internal Rn-222 Cont.:



β-α coincidence of Bi-214 - Po-214

Bi-214 : $E_{\beta+\gamma} \lesssim 3{,}2$ MeV

Po-214 : $E_{\alpha} = 7{,}69$ MeV ($E_{vis} \sim 0{,}75$

$t_{1/2}$(Po-214) = 163 μsec

(Pulse Shape Information)

Decay of Rn-222 present in PC-scintillator directly after filling (initial contamination $2.5 \cdot 10^3/d$)

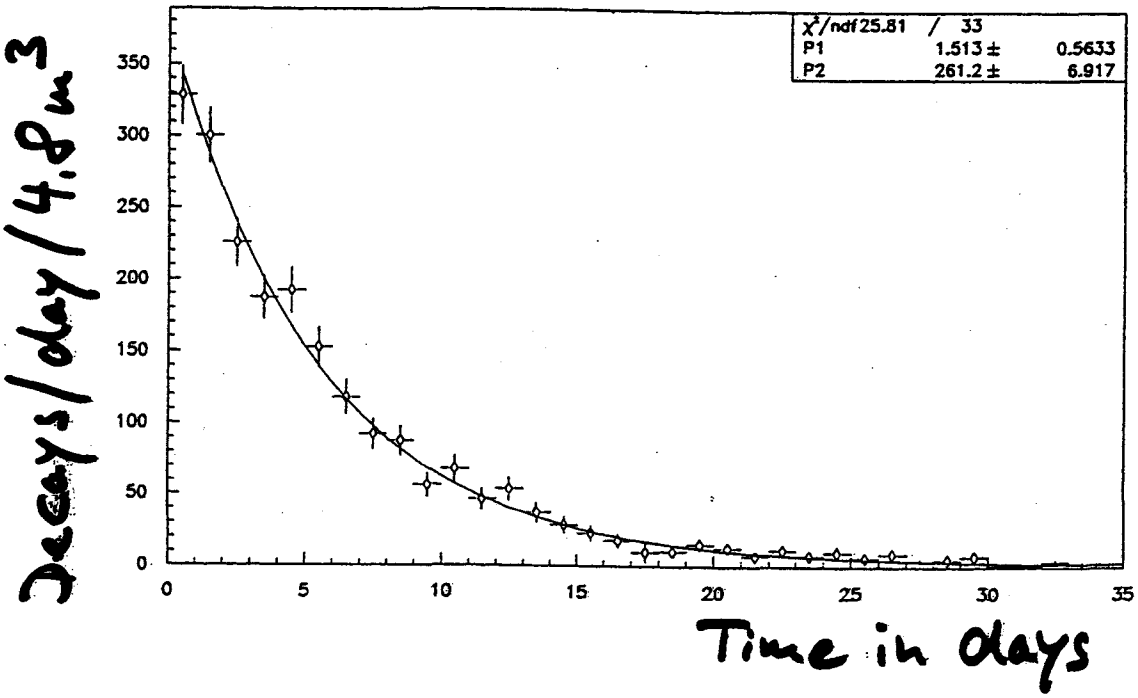
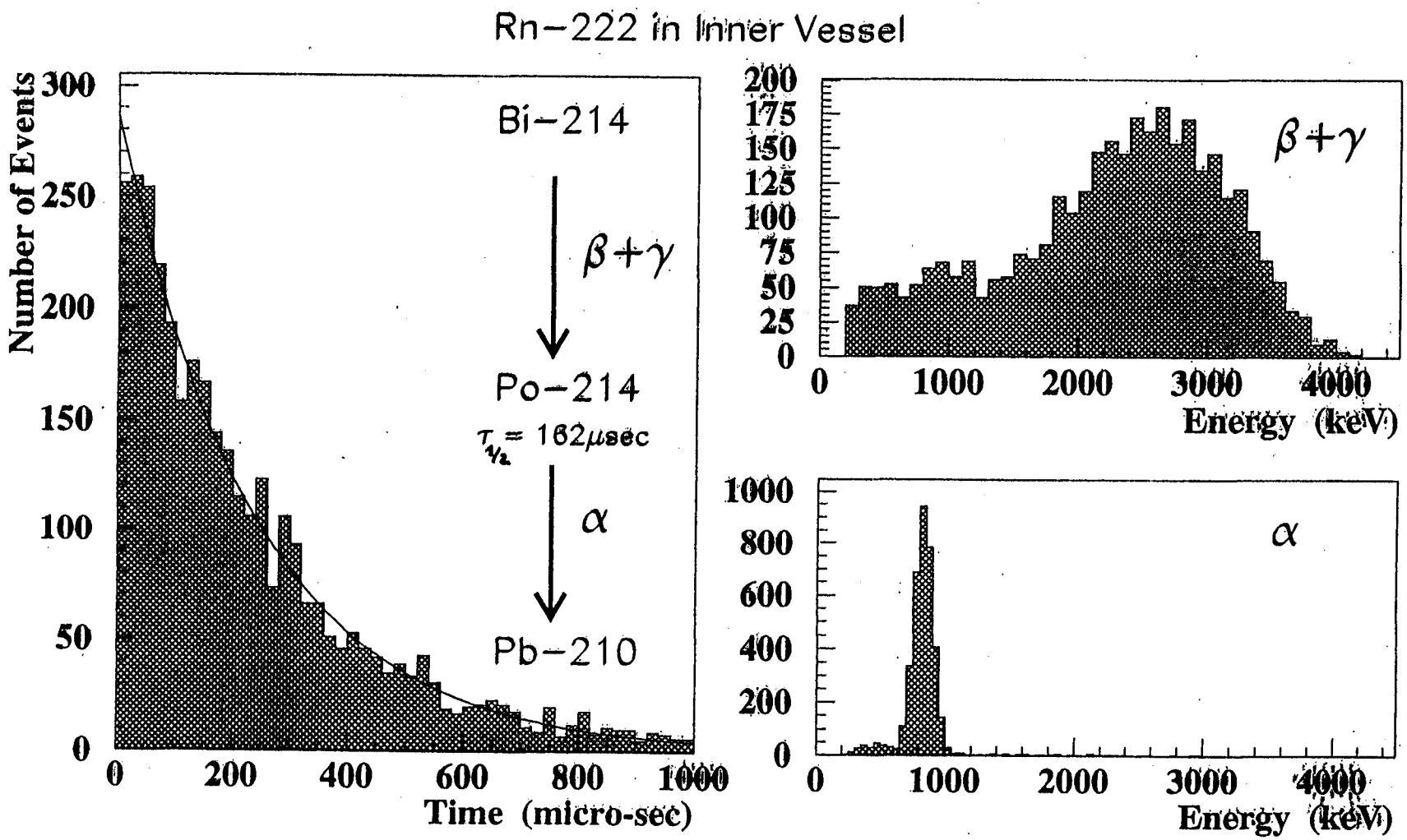


Figure IV.14: Phase Foxtrot.
Rate of mass 214 sequences selected by means of the U cuts.
The rate is corrected for dead time in data acquisition.
The distribution is fitted using an exponential with ^{222}Rn meanlife plus a constant function in order to disentangle the transient ^{222}Rn contamination from the internal intrinsic ^{238}U contamination. If allowed to vary, the meanlife is consistent with ^{222}Rn meanlife.

Residual counting rate: 1,5/d = 4 μBq

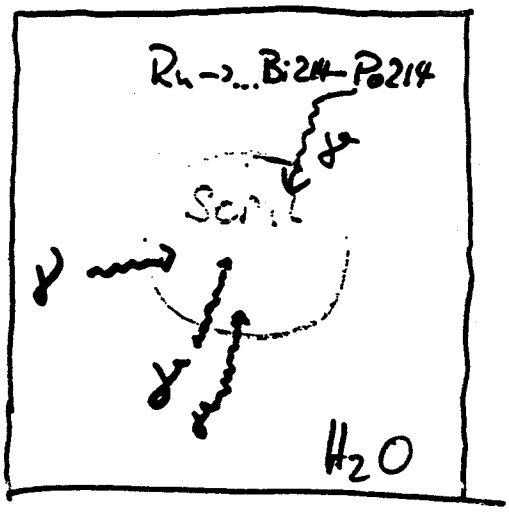
if in equil. with parents ⟹ $3{,}5 \cdot 10^{-16}$ g U/g


Rn−222 in Inner Vessel
Number of Events
Time (micro-sec)
Bi−214
β+γ
Po−214
τ1/2 = 162μsec
α
Pb−210
β+γ
Energy (keV)
α
Energy (keV)

Measurement of External Rn Cont.:

Energy spectrum dominated by:

$E_\gamma = 2{,}2$ MeV
$0{,}6$ MeV
⋮

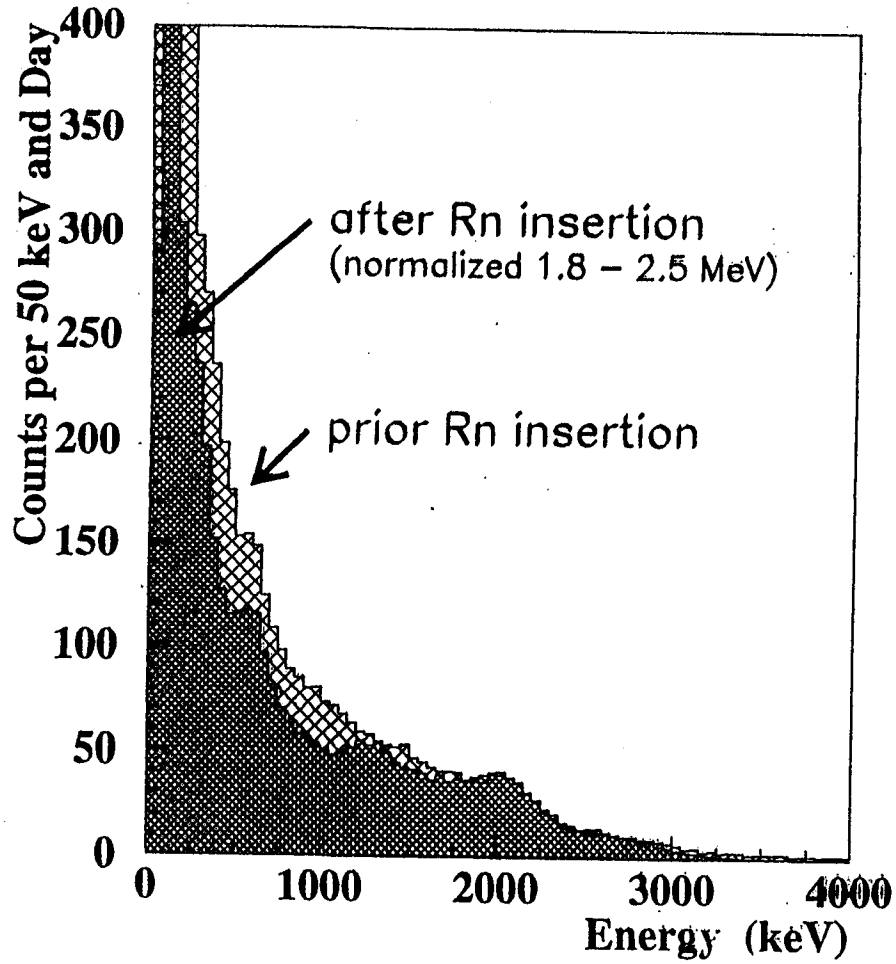


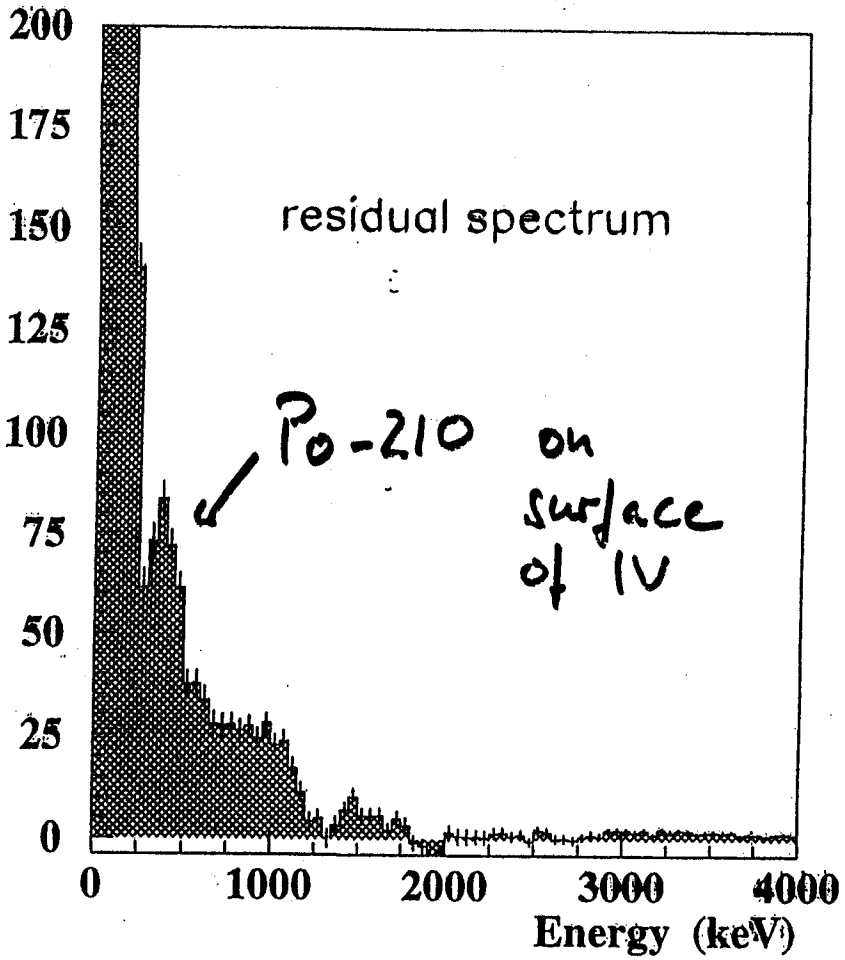
Analysis of Energy spectrum:

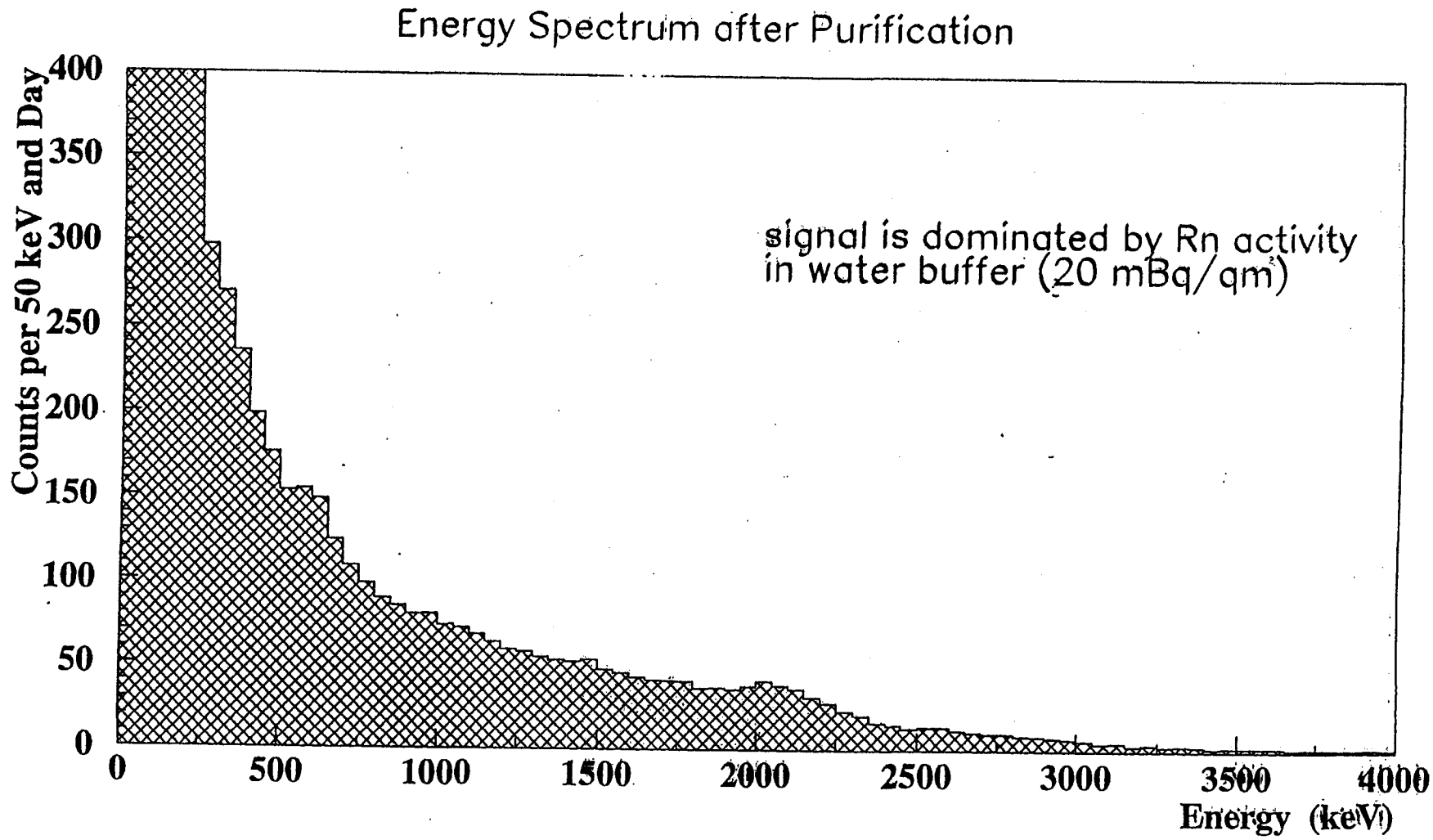
1) MC - Simulation

2) Introduction of Rn in H_2O-Shi

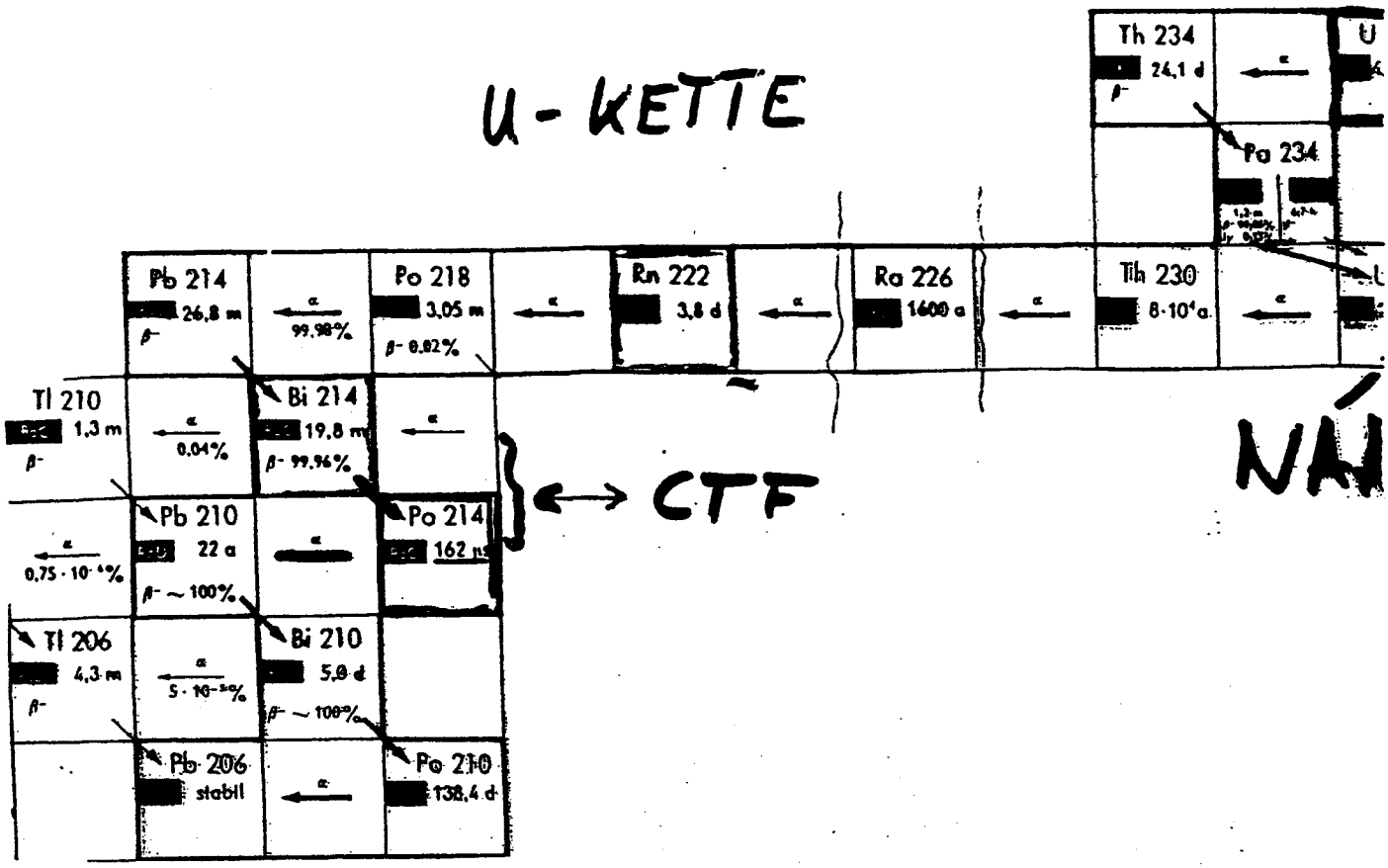
Radon Insertion into the Water Buffer



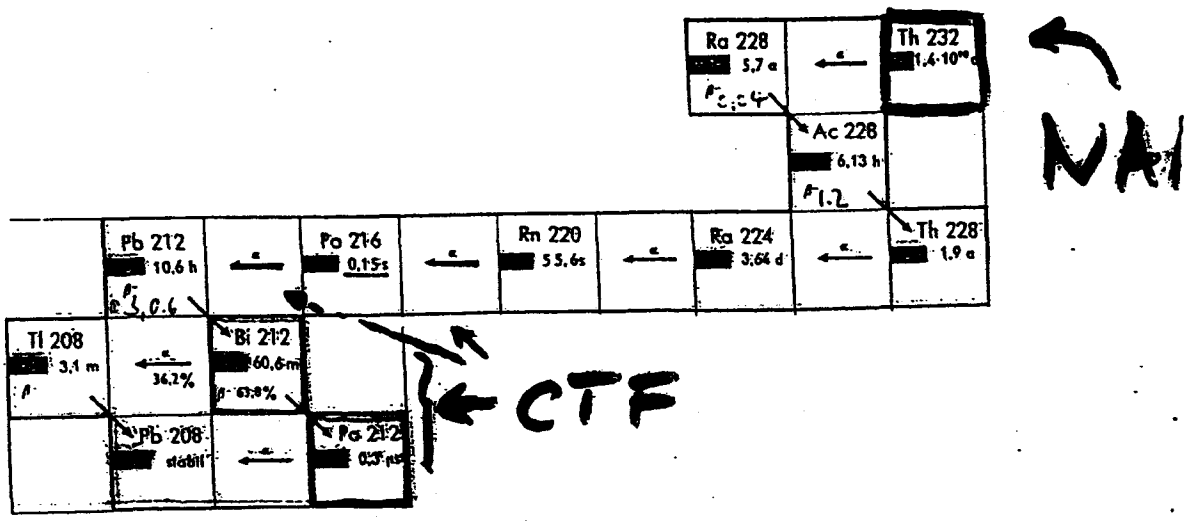



Energy Spectrum after Purification
signal is dominated by Rn activity
in water buffer (20 mBq/qm²)
Counts per 50 keV and Day
0
50
100
150
200
250
300
350
400
0
500
1000
1500
2000
2500
3000
3500
4000
Energy (keV)

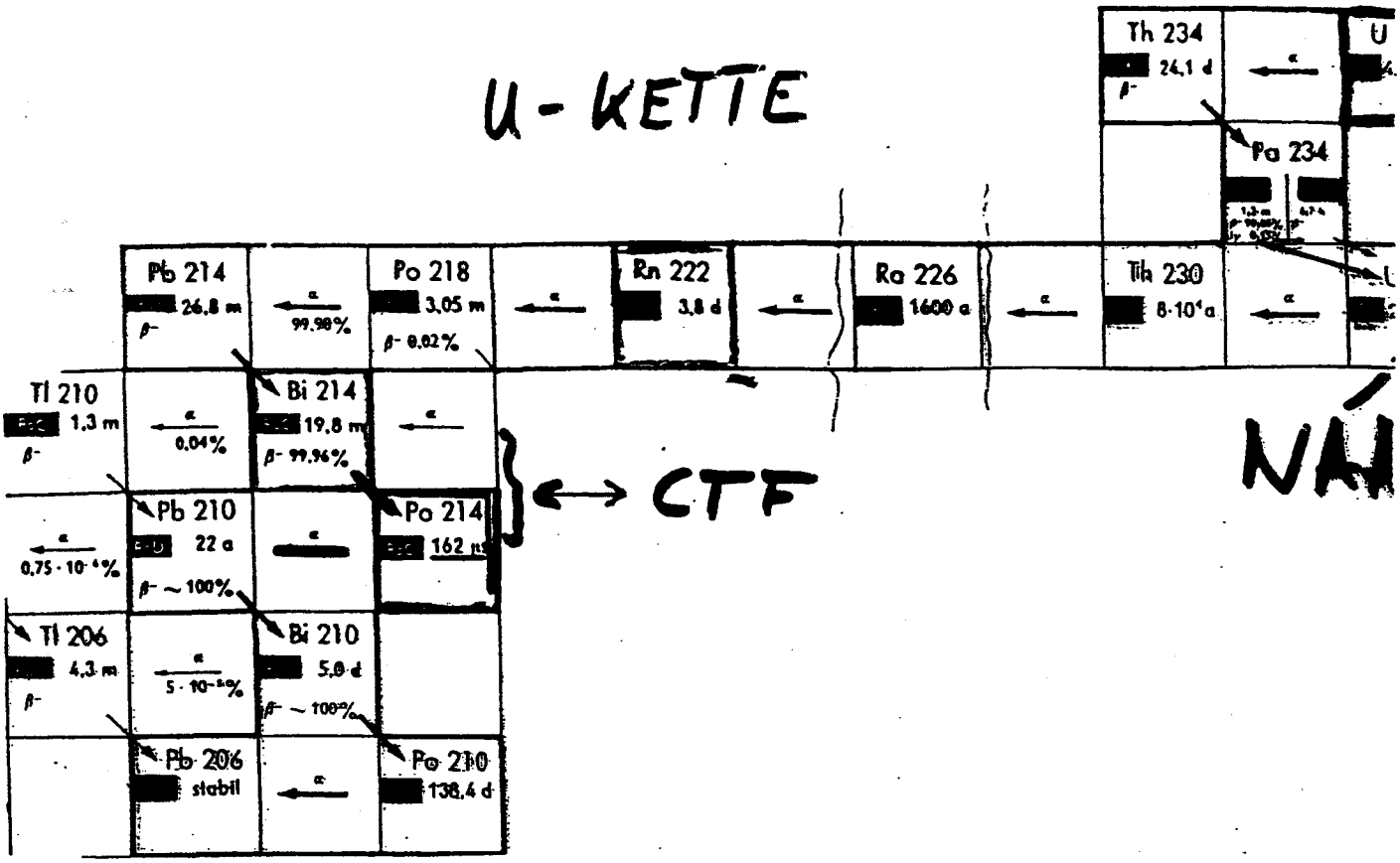
U-KETTE



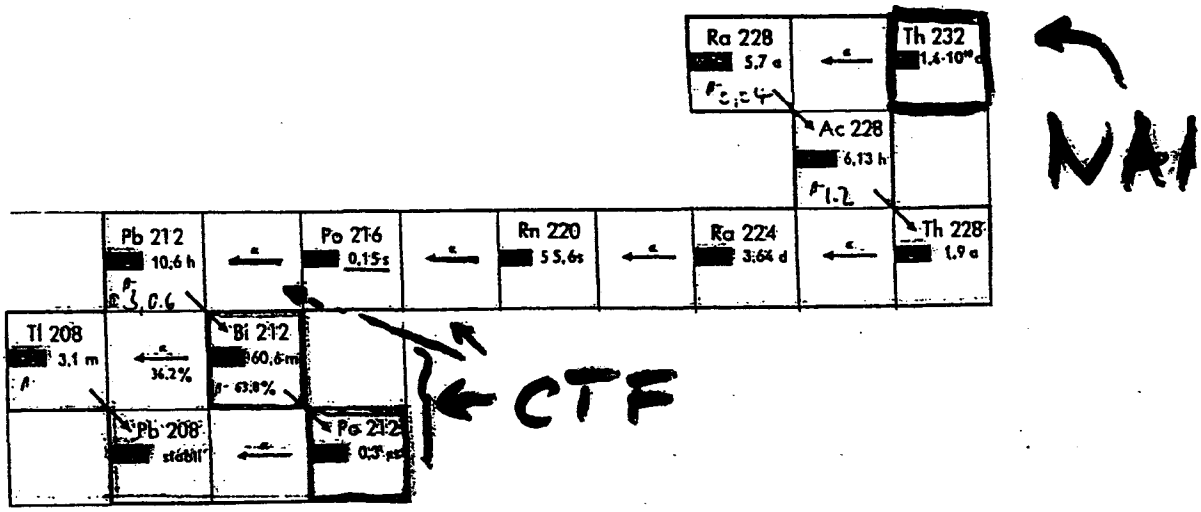
Th-KETTE



U-KETTE



Th-KETTE



CTF-II : renovation and up-grade of CTF

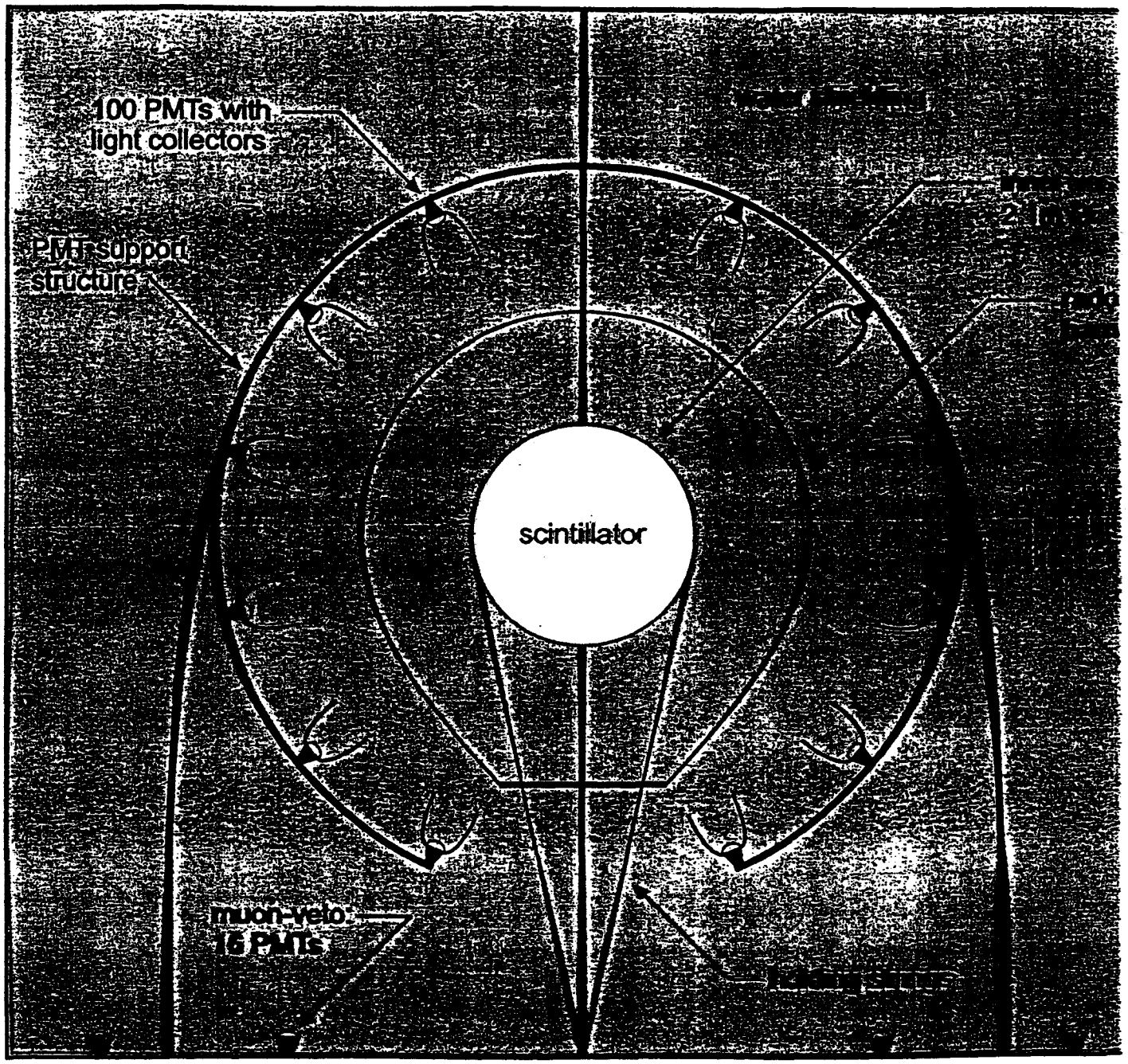
New Items:

- PMT's
- μ-veto
- Inner Vessel and shroud (Rn-barrier
- Liquid handling and purification system (sub-system of Borexino detector.

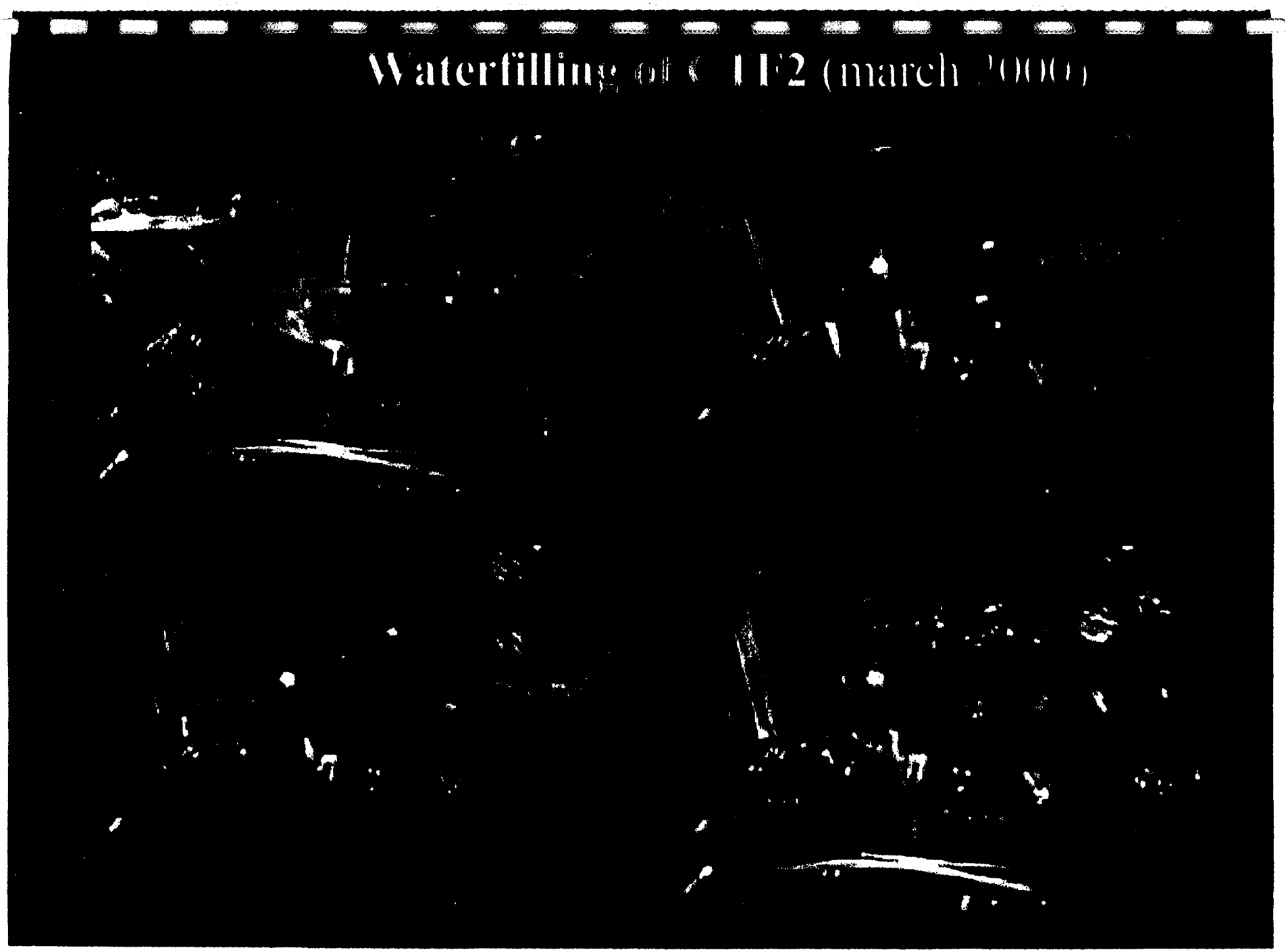
Start-up: May 2000

Goals: increased sensitivity
Reduction of Rn Bgd
Test of scint. batches for BX
Test of BX-Subsystems:
(Liq. handling: MOD*Ø, ...)

CTF II - Design

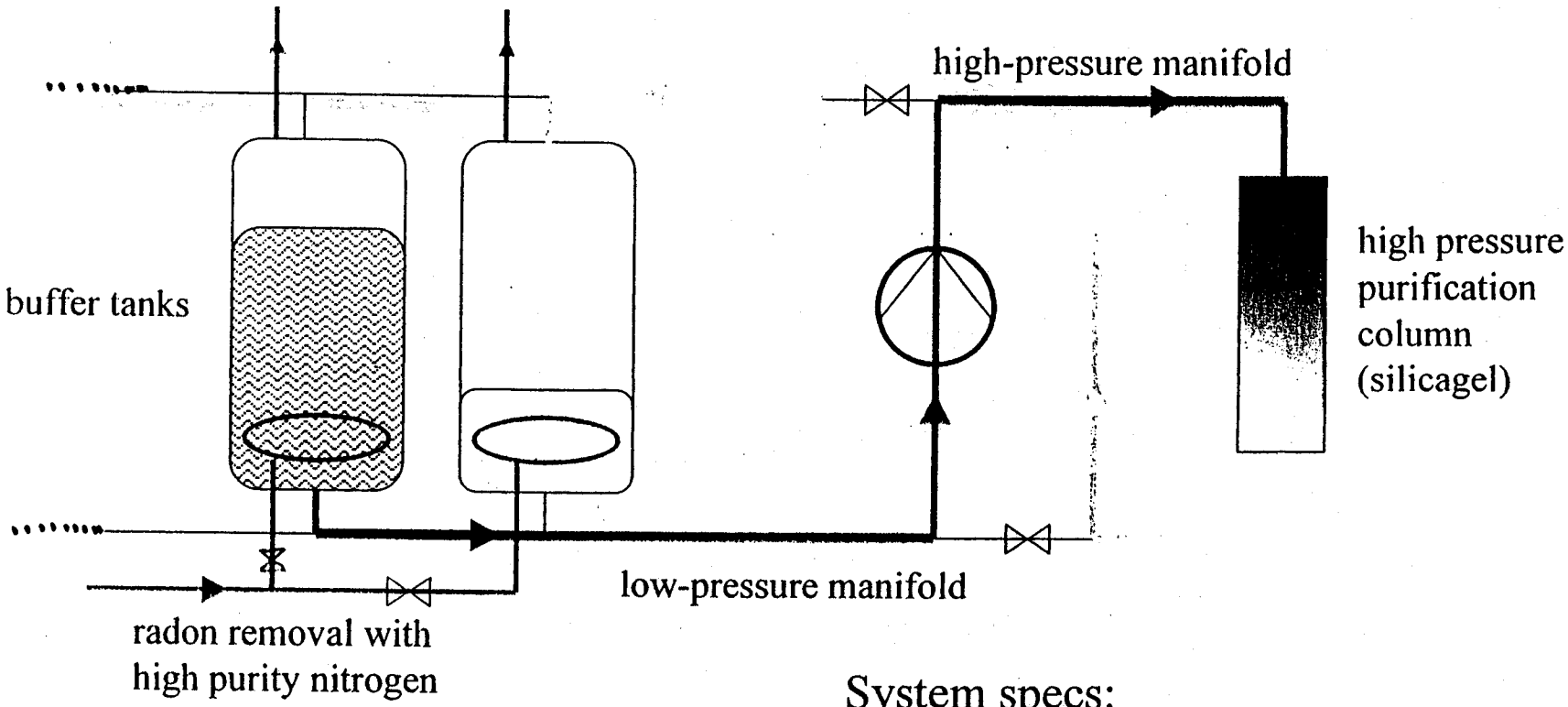


Waterfilling of C 11F2 (march 20000)



MOD*0

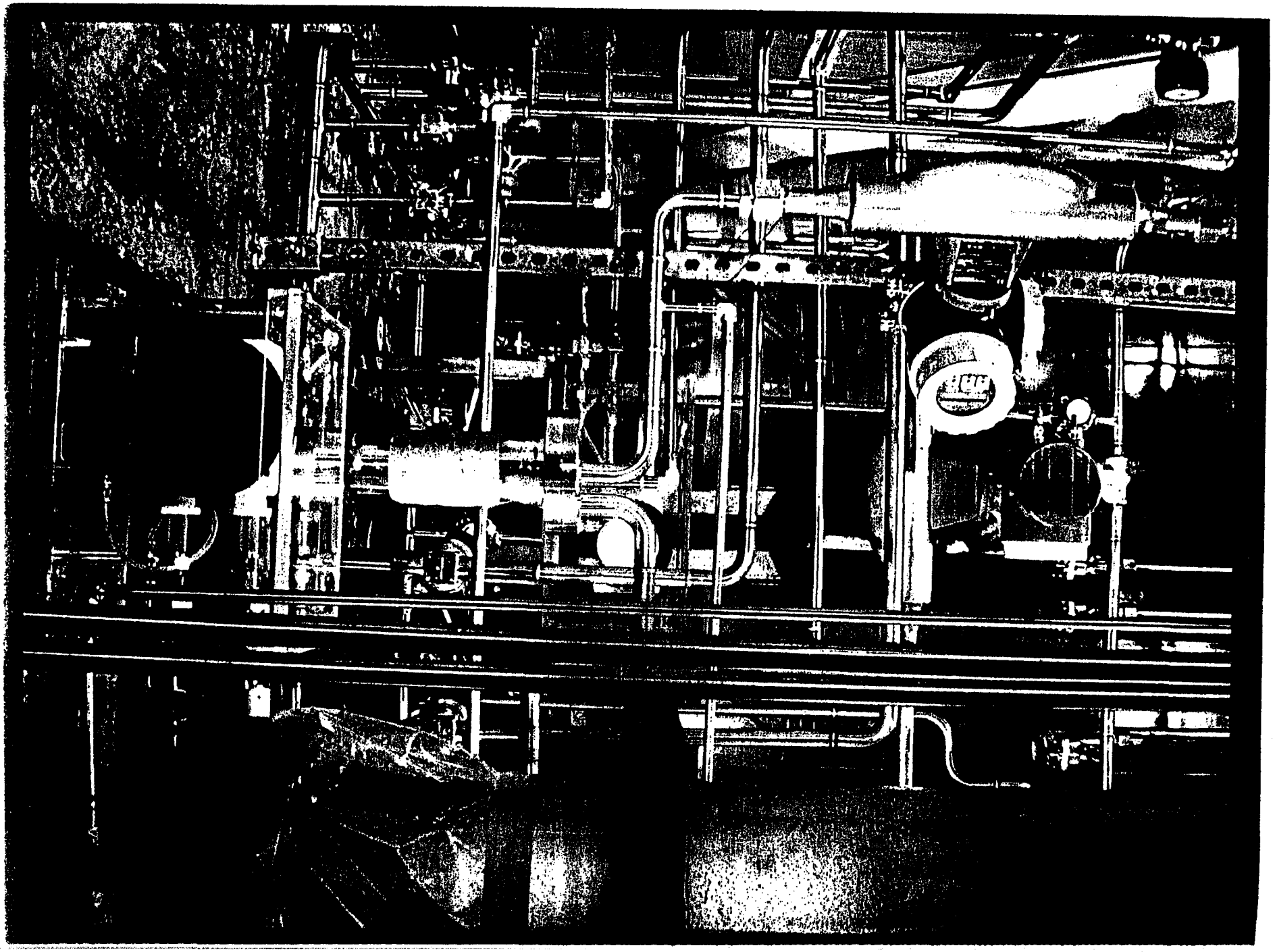
A scintillator handling ① and purification system for BOREXINO + CTF



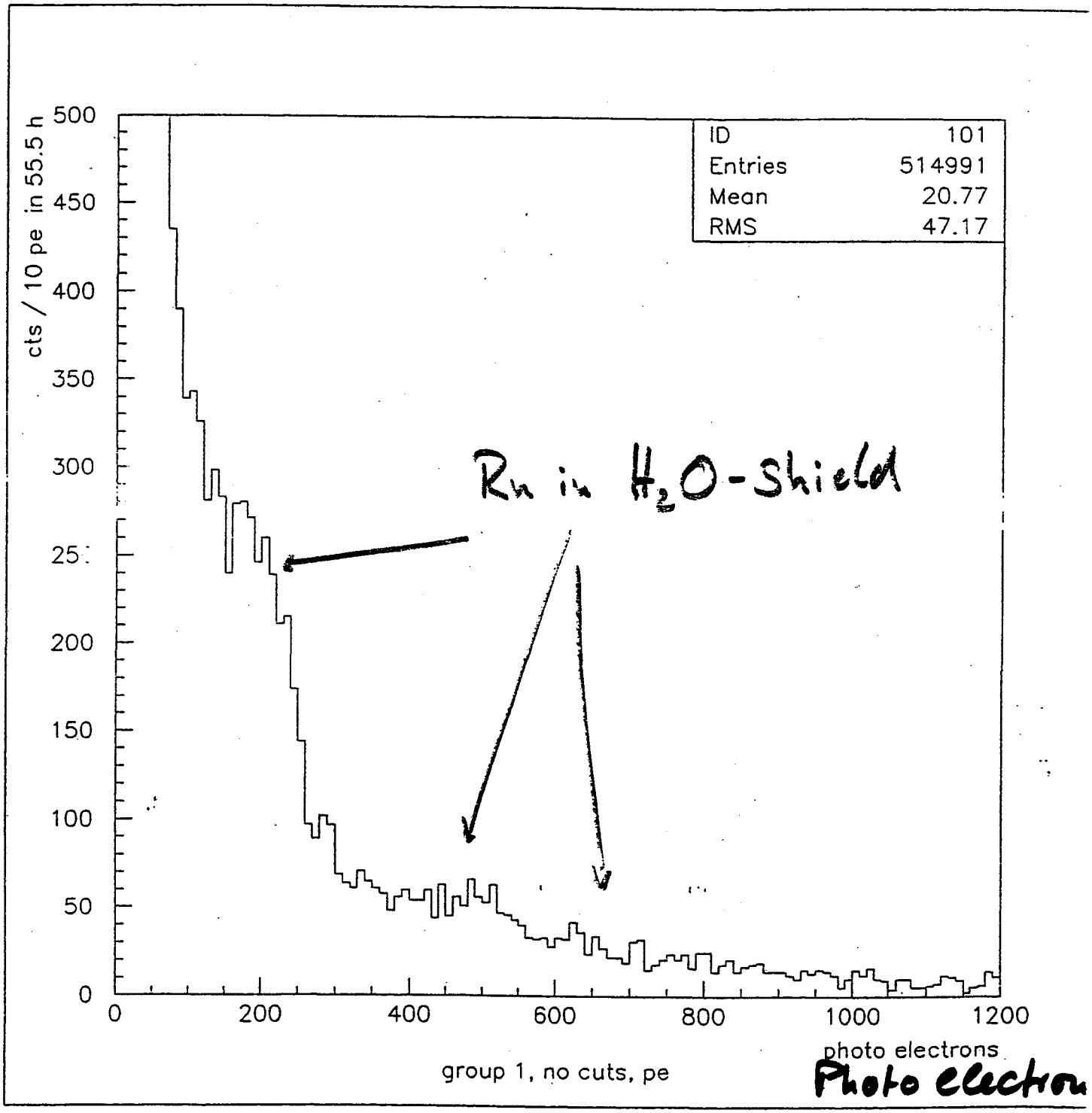
System specs:

- UHV leak tight
- electropolished stainless steel
- special cleaning

①: handling = • preparation of scint.: fluor purification + mixing with solvent
• filling of Inner Vessel and buffer volume
• ....



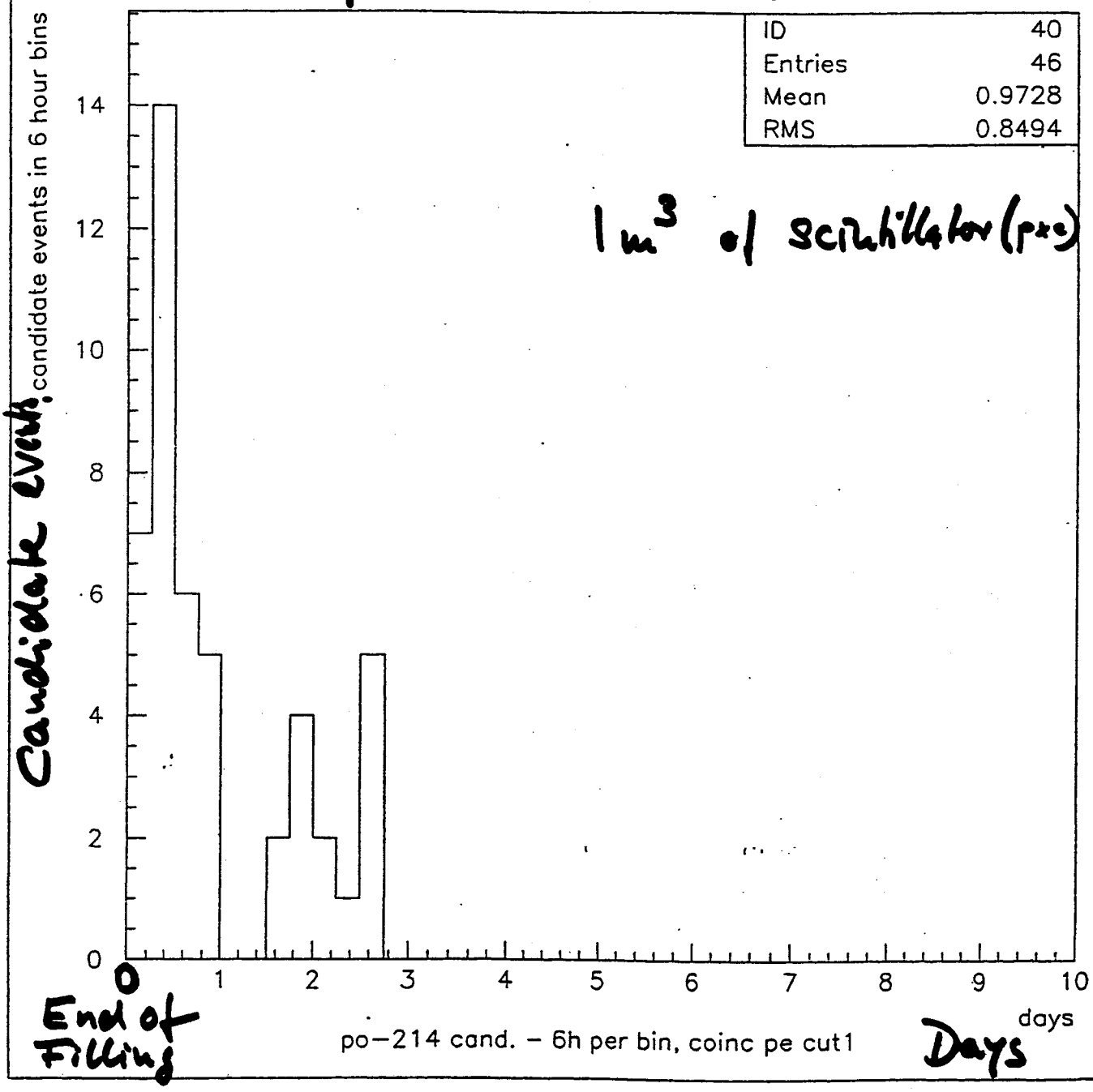
First Data of CTF-II



1 m^3 of scintillator (pxe)

Bi-214 → ~~Bi-214~~ - Po-214 (β-α coincidences)
(= Rn in PXE scintillator)

Very Preliminary!



First "Rn-free" Scintillator Purification

Summary

CTF: sensitiv instrument for ^{222}Rn (Ra/U)
sensitivity $\lesssim 1/d/4\,m^3 \cong \sim 2\,nBq/l$
(CTF II : shroud may increase sensitivity by a factor ~10)

Scint. handling + purification (test of Mod Ø):
handling and purification of scintillator at ~10/d/10^3 kg level

Shroud test : ongoing

test of all BX subsystems planned with CTF II

Radon Emanation Measurements for BOREXINO

W. Rau, G. Heusser

Neutrino 2000, pre-conference workshop:
Radon Background in Rare-Event Experiments

June 14th, 2000
Sudbury, Canada

Outline

- Principle or Rn emanation measurements
- The Emanation Assay System
- Measurement procedure
- Samples

Principle of Rn emanation measurements

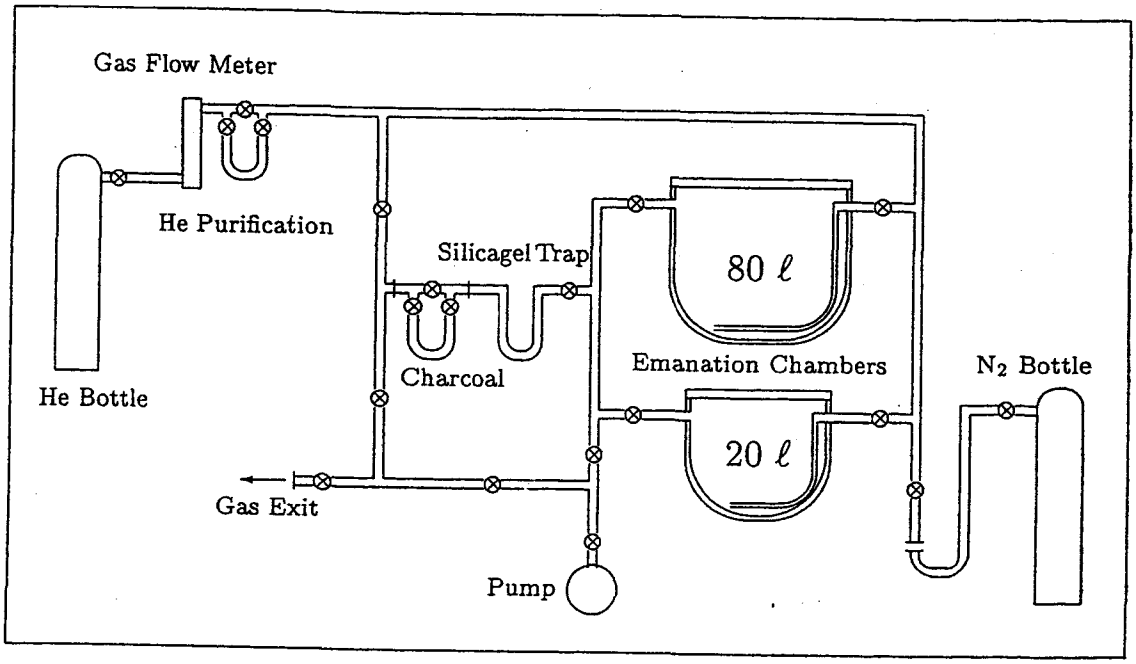
- Place sample in a sealed vessel
- Remove all trapped Rn
- Rn growth in with a half life of 3.8 days
- Extract the emanated Rn after several days
- Rn counting

Most essential prerequisits for high sensitive emanation measurements:

- Low blank activity of the complete measuring cycle (Radium contamination of the system, air leaks, memory effects)
- Highly sensitive Rn detection method

The Emanation Assay System

The Apparatus



- Material: stainless steel
- Sealing: all joints are metal sealed
- Treatment of inner surfaces: electropolishing

Blank activities of the emanation apparatus (in μBq)

charcoal (saturation)	< 36
charcoal (per sample)	< 1.1
small chamber	64 ± 15
big chamber	153 ± 25

Measurement procedure

Radon collection

- Sample loading, remove Rn (evacuation)
- Fill chamber with Rn free Helium
- Radon ingrowth
- Extract Rn together with He (evacuation)
  Rn is trapped in a charcoal trap at -196 °C

Before each operation all relevant traps are baked out under He purge

Radon sample preparation and Radon detection

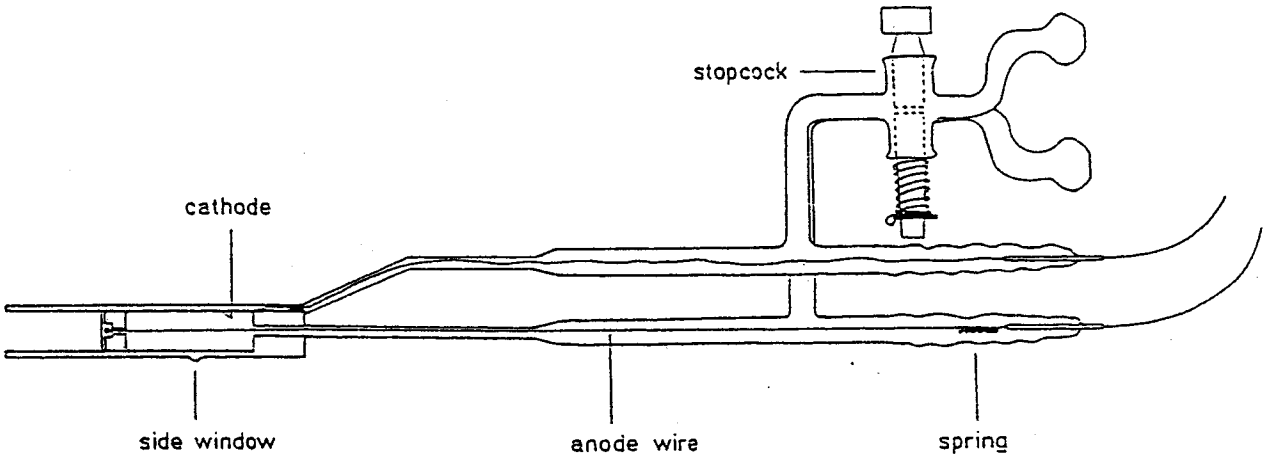
- Connect charcoal trap to a glas vacuum line
- Radon sample purification
- Transfer Rn + counting gas to a low background proportional counter
- Counting in an underground laboratory (15 mwe, additionaly ca. 10 cm lead shield); only α-decays from Rn and its progenies are counted

For small samples: glas vials available (blank consistant with 0), directly connected to glas line.

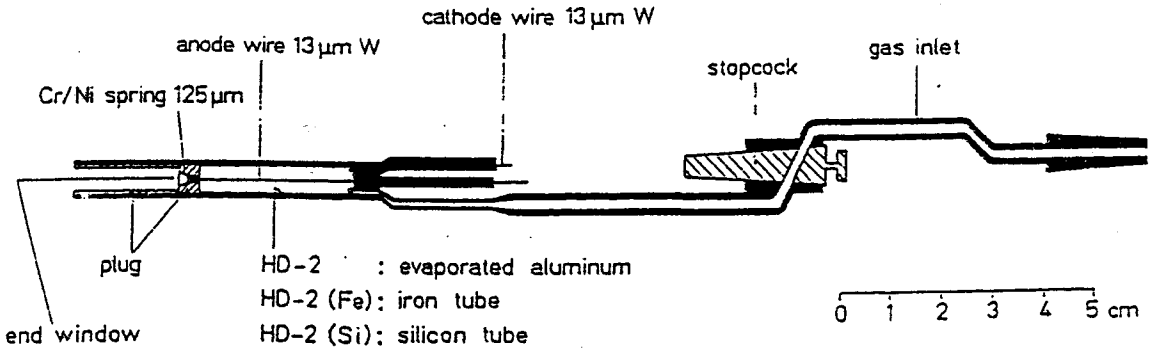
Blank activity from sample preparation line:	$10-60$ μBq
Background of counters:	$\sim$ 1 count per day
Efficiency:	$\sim$ 1.5 counts per Rn atom

Low-Level Proportional Counter

Davis-type



HD-2-type



Samples

Samples of construction material for the BOREXINO-detector, for other installations around the experiment and from the CTF are measured

Sample	Surface/ Amount	Activity [mBq]	Vessel
TYVEK 1073B	16 m^2	< 0.0039 /m^2	big chamber
Steel foil	70 m^2	(0.0100 ± 0.0007) /m^2	big chamber
PMT	1 piece	< 0.031	small chamber
Master Bond ZP45HT	340 cm^2	< 0.86 /m^2	small chamber
Butyl O-rings	0.66 m^2	(19.6 ± 0.5) /m^2	small chamber
PERMATEX	280 cm^2	(24.9 ± 4.2) /m^2	glas vial
Araldite 2012	138 cm^2	(4.8 ± 3.2) /m^2	glas vial
Nitril	96 cm^2	(187 ± 9) /m^2	glas vial
Busto	139 cm^2	(1750 ± 40) /m^2	glas vial
Kromasil	104 g	(3.6 ± 0.3) /kg	glas vial
Safety valve	1 piece	0.08 ± 0.03	direct
Carbo-Act charcoal	2.02 kg	(0.33 ± 0.05) /kg	special vessel

Rn in nitrogen: measurement and purification

W. Rau, G. Heusser, B. Freudiger, M. Laubenstein, M. Balata, T. Kirsten

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June 14th, 2000
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Outline

- Purity requirements and idea for measurement
- Radon concentration line
- Measurements I
- Nitrogen purification
- Measurements II

Purity requirements and measurement idea

Nitrogen is used for gas stripping of the scintillator to remove gaseous impurities, especialy Rn

→ required Rn purity: $< 1\ \mu\mathrm{Bq/m^3}$

→ direct measurement is impossible
(1 atom corresponds to 2 μBq!)

(State of the art: sensitivity of $1-10$ mBq/m^3 for Rn in gas (electrostatic chamber))

Idea of measurement:

- concentration of Rn from large volume of nitrogen
- remove impurities
- proportional counting

Radon concentration line

Method:

- Radon concentration on a charcoal adsorber at liquid nitrogen temperatrure

Requirements:

- hold back capacity for Rn from about 1000 m^3 nitrogen
- charcoal with low ^{226}Ra concentration
- tightness against Rn diffusion from outside
- fast concentration process to minimize emanation

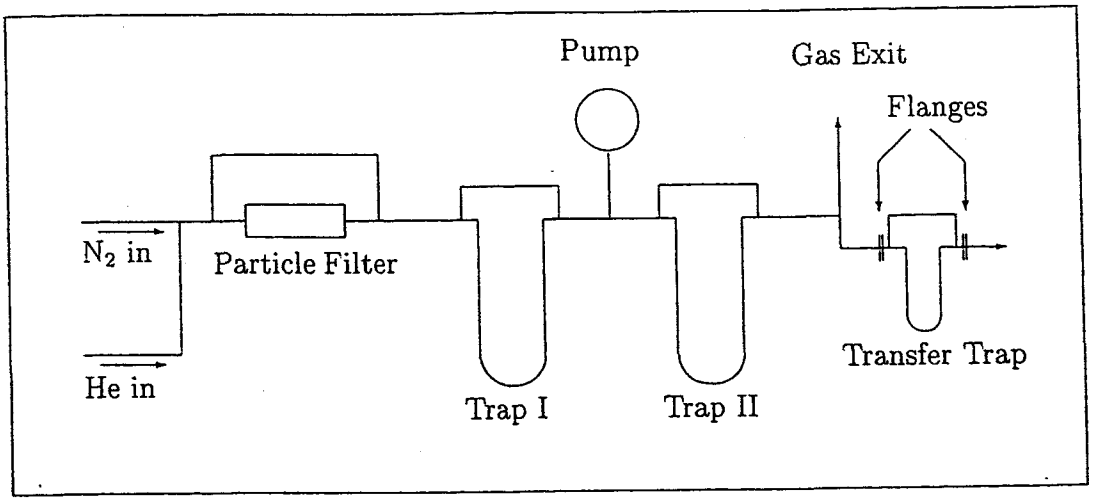
Investigations:

- selection of a charcoal (low ^{226}Ra concentration) $\Rightarrow$ choice: synthetic charcoal with (0.3 ± 0.1) mBq/kg emanation rate
- measurement of the adsorption coefficient ($\sim 2 \cdot 10^7$)

Realisation:

- high vacuum stainless steel apparatus
- all joints are metal sealed (elastomer sealings suffer from Rn emanation or high permeability for Rn)
- charcoal trap: 1.2 ℓ, filled with 156 g of charcoal
- additional parts: flow regulating valve, particle filter, flow meter (up to 20 m^3/h), pumps, pressure indicators

Scetch view of the concentration line (simplified)

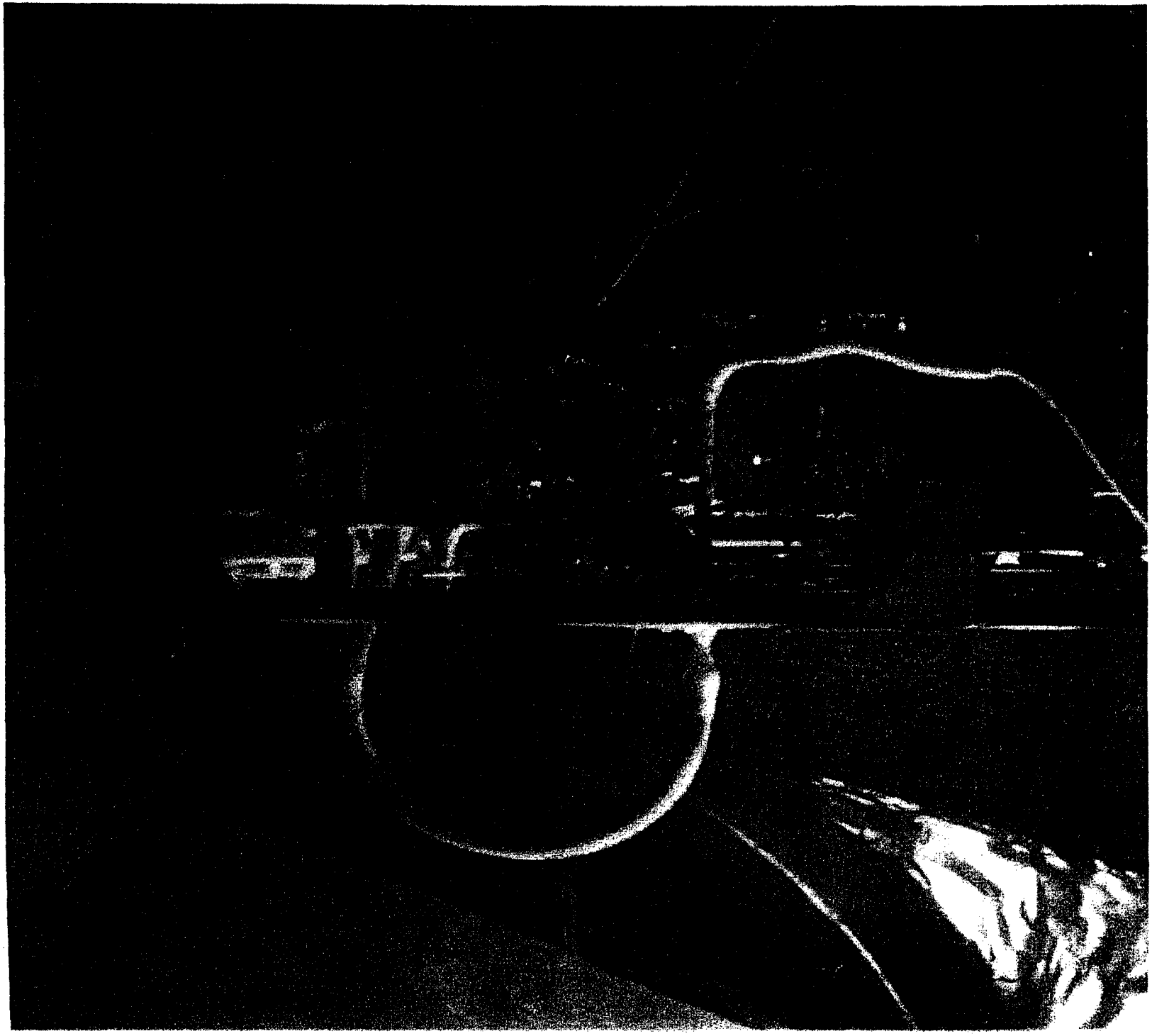


Concentration procedure:

1. evacuate line together with the charcoal trap
2. bake out trap at ~200 °C (slight purge with purified He)
3. coole trap with liquid nitrogen
4. Rn collection: N_2 flow rate ~ 20 m^3/h (STP), p ≈ 2 bar overpressure
5. evacuate trap at –196 °C, then at –100 °C to remove the nitrogen
6. heat up trap and transfer Rn to a small 'transfer trap' (~ 0.8 g charcoal, –196 °C) by He purge
7. dismount transfer trap and connect it to the counter filling line, purify sample and fill counter

Blank of the concentration line

	equilibrium activity [μBq]	106 m^3 sample [μBq]	500 m^3 sample [μBq]
charcoal	60 ± 20	3.0 ± 1.1	8.9 ± 3.3
other parts	117 ± 52	4.4 ± 2.0	17.8 ± 8.1
total		7.4 ± 2.3	26.7 ± 8.8



Measurements I

Standard purity nitrogen

sample size [m^3]	production plant	activity [$\mu Bq/m^3$]
40	Gran Sasso	51 ± 7
35	Gran Sasso	27 ± 4
36	Gran Sasso	78 ± 11
30	Gran Sasso	71 ± 9
36	Heidelberg/gas phase	63 ± 9
45	Heidelberg/gas phase	43 ± 5
50	Heidelberg/gas phase	54 ± 6
106	Heidelberg/liquid phase	144 ± 14

Rn concentration too high:

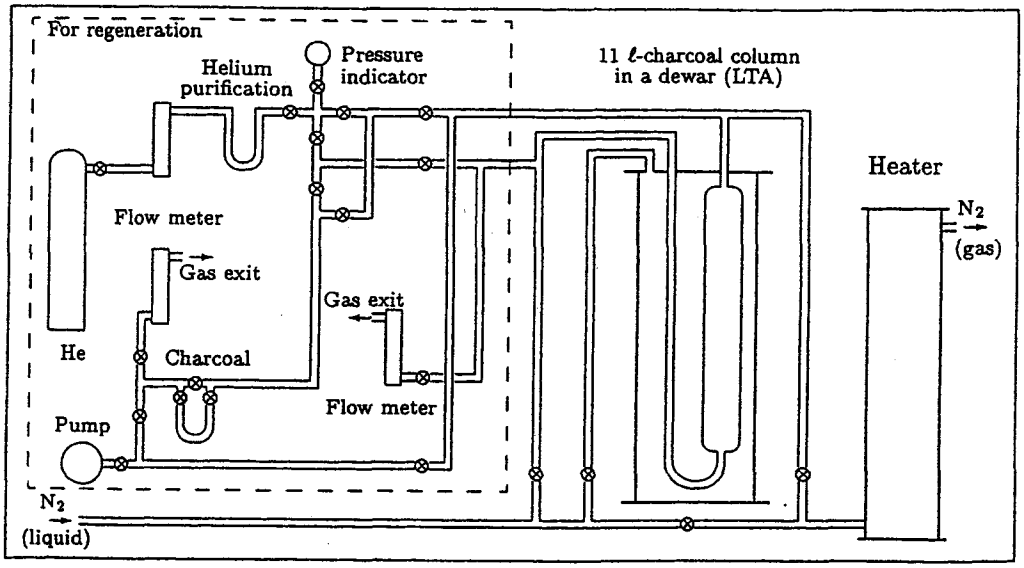
Nitrogen purification is necessary

Nitrogen purification

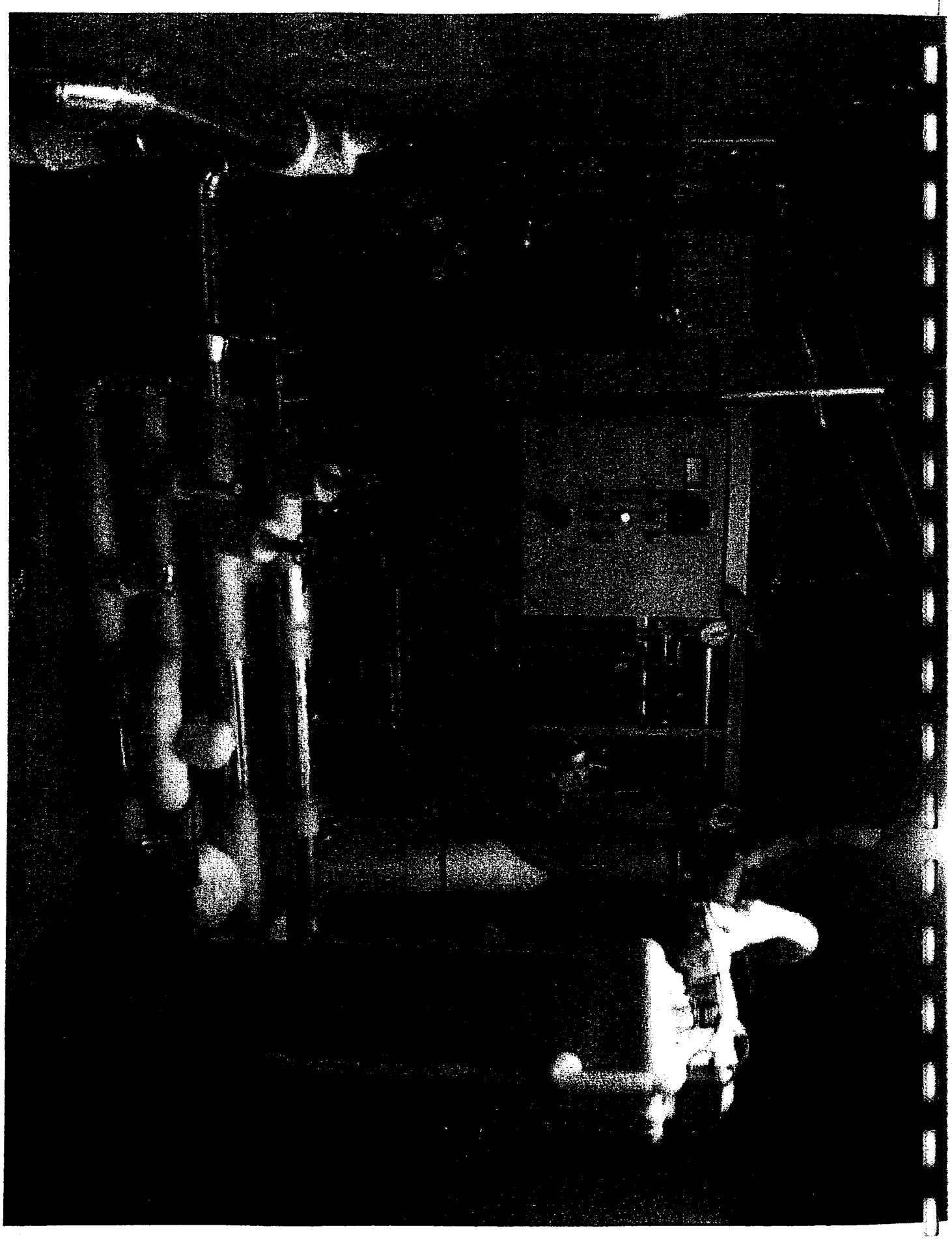
- Rn can be removed from gaseous nitrogen by adsorption on charcoal
- Adsorption at low temperature needs cooling power
- Purification at room temperature requires huge charcoal columns with ultra low ^{226}Ra concentration

New idea: charcoal purification of nitrogen in liquid phase

- Liquid nitrogen from storage passes charcoal column before evaporation
- Charcoal column is placed in a dewar to guarantee constant low temperature
- Evaporation is performed with a special small surface evaporator to reduce emanation



Scetch view of the Low Temperatur Adsorber for nitrogen purification



Measurements II

High purity nitrogen

sample size [m^3]	production line	activity [$\mu Bq/m^3$]
102	Heidelberg	< 2
103	Heidelberg	< 25
106	Gran Sasso	0.27 ± 0.31
120	Gran Sasso	0.48 ± 0.32
500	Gran Sasso	0.78 ± 0.10
500	Gran Sasso	0.30 ± 0.09

Conclusions

- Radon concentration below 1 $\mu Bq/m^3$ can be measured
- Nitrogen with the required purity (< 1 $\mu Bq/m^3$ of Rn) can be supplied in large quantities (100 m^3/h)

BOREXINO
MONITORING OF RADON IN AIR

⇒ **monitoring of Rn in air within**

a) the S.S.S. (housing of the Borexino detector)

b) the cleanrooms

⇒ **Rn concentration level:** 1 mBq/m^3

⇒ **Detection limit of the Rn monitor:** < 1 mBq/m^3

⇒ **Method:**

electrostatic collection of ^{222}Rn daughter ^{218}Po as positive ion on an α - counter

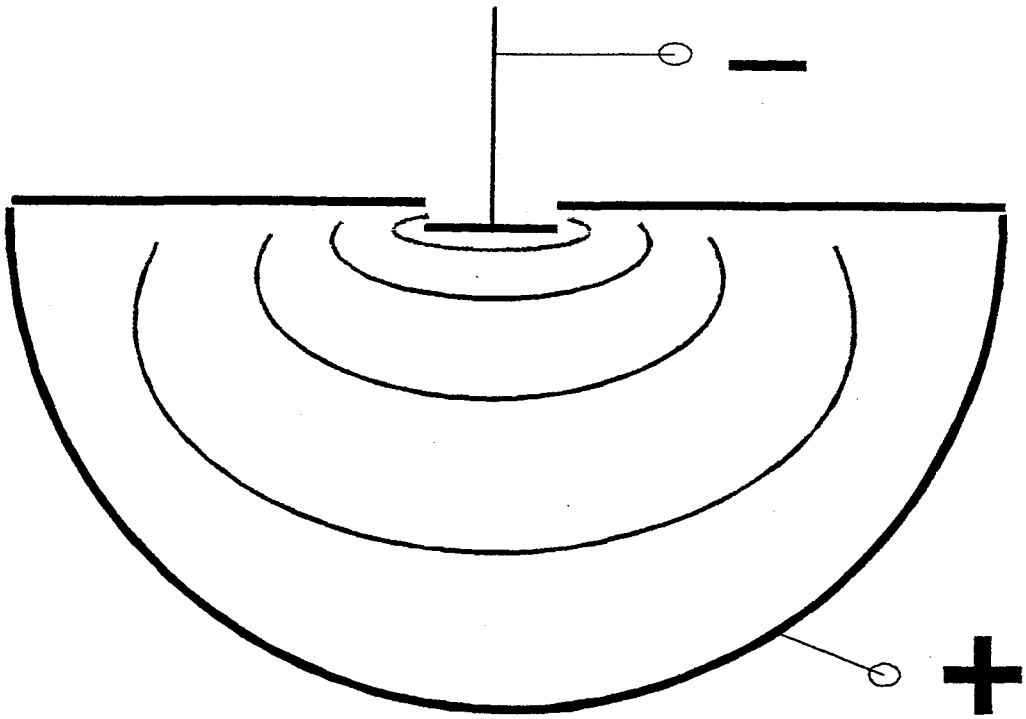
⇒ **Important parameters of Po ions in air**

mean life concerning neutralization in dry and clean air: $\geq$ 0,02 sec

mobility: 1,86 $\pm$ 0,11 cm^2/Vs

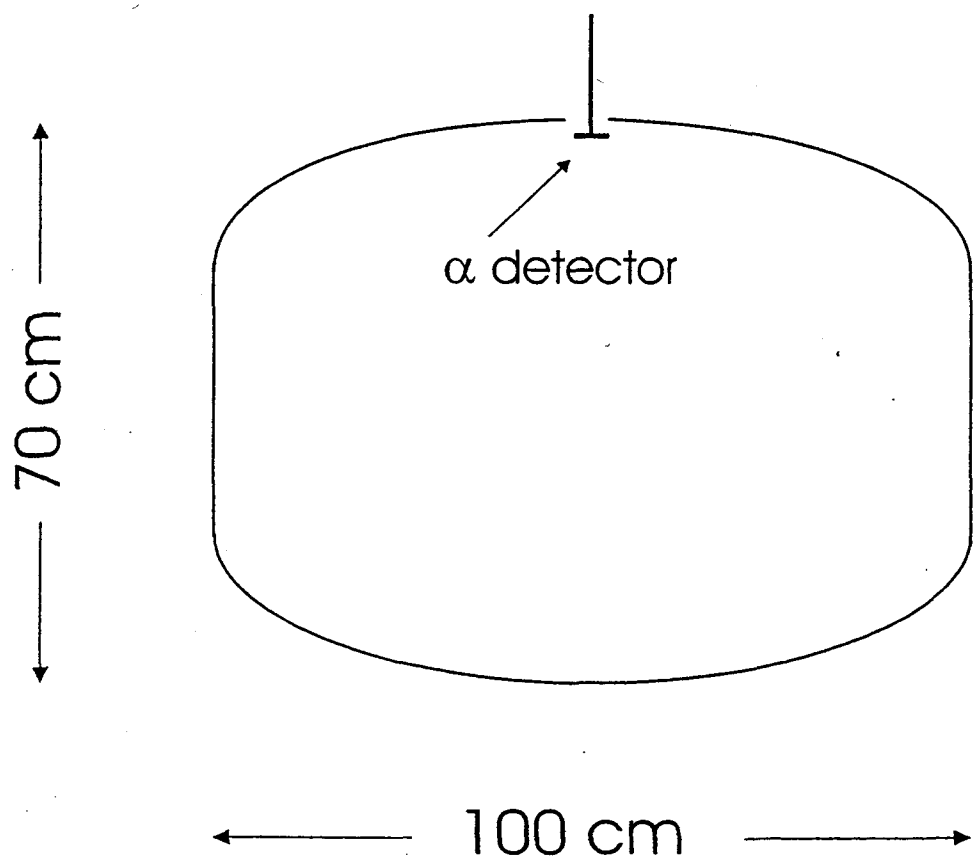
⇒ **Critical components in air:**

H_2O, NO_2, OH^-, hydrocarbons, ...

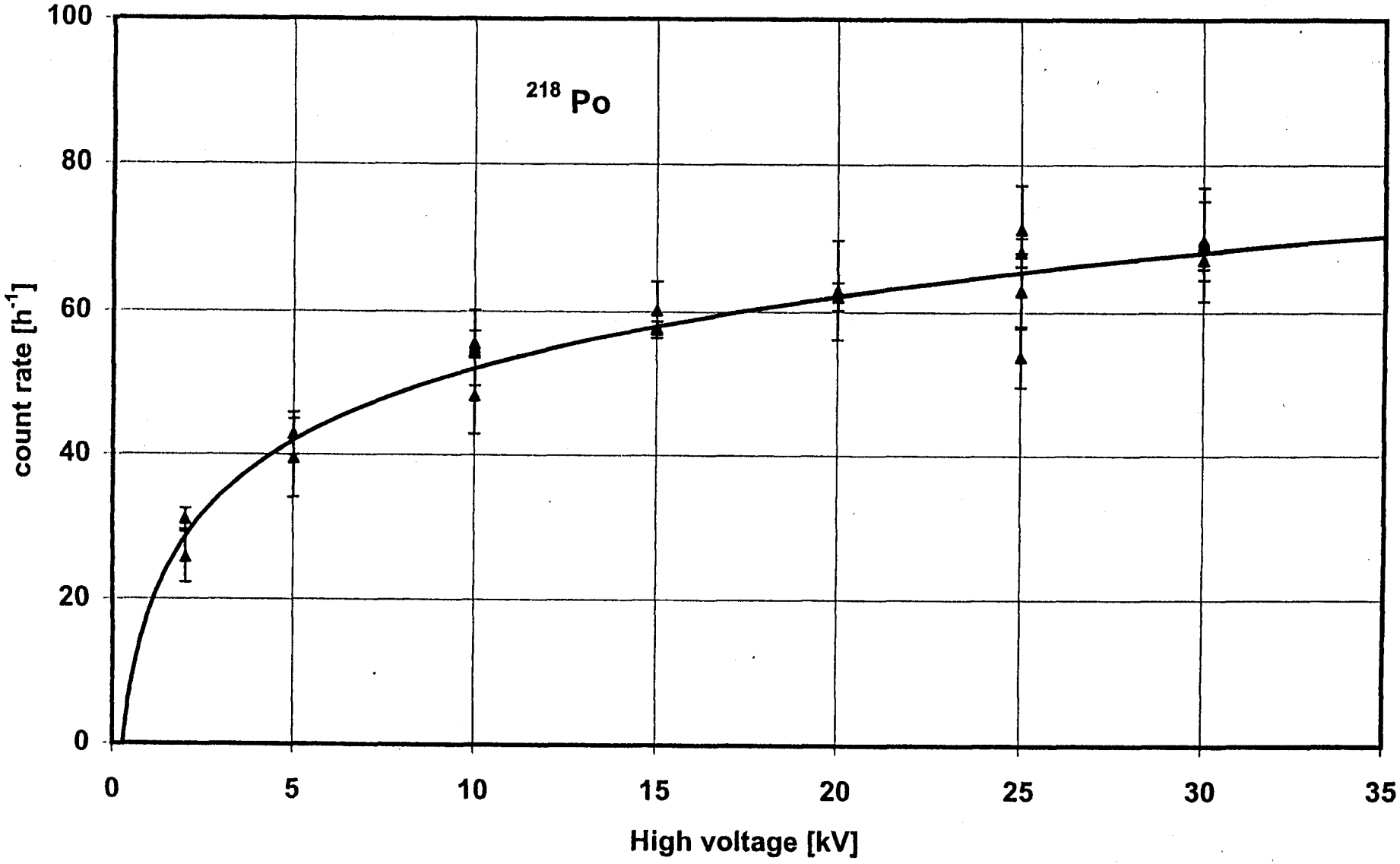


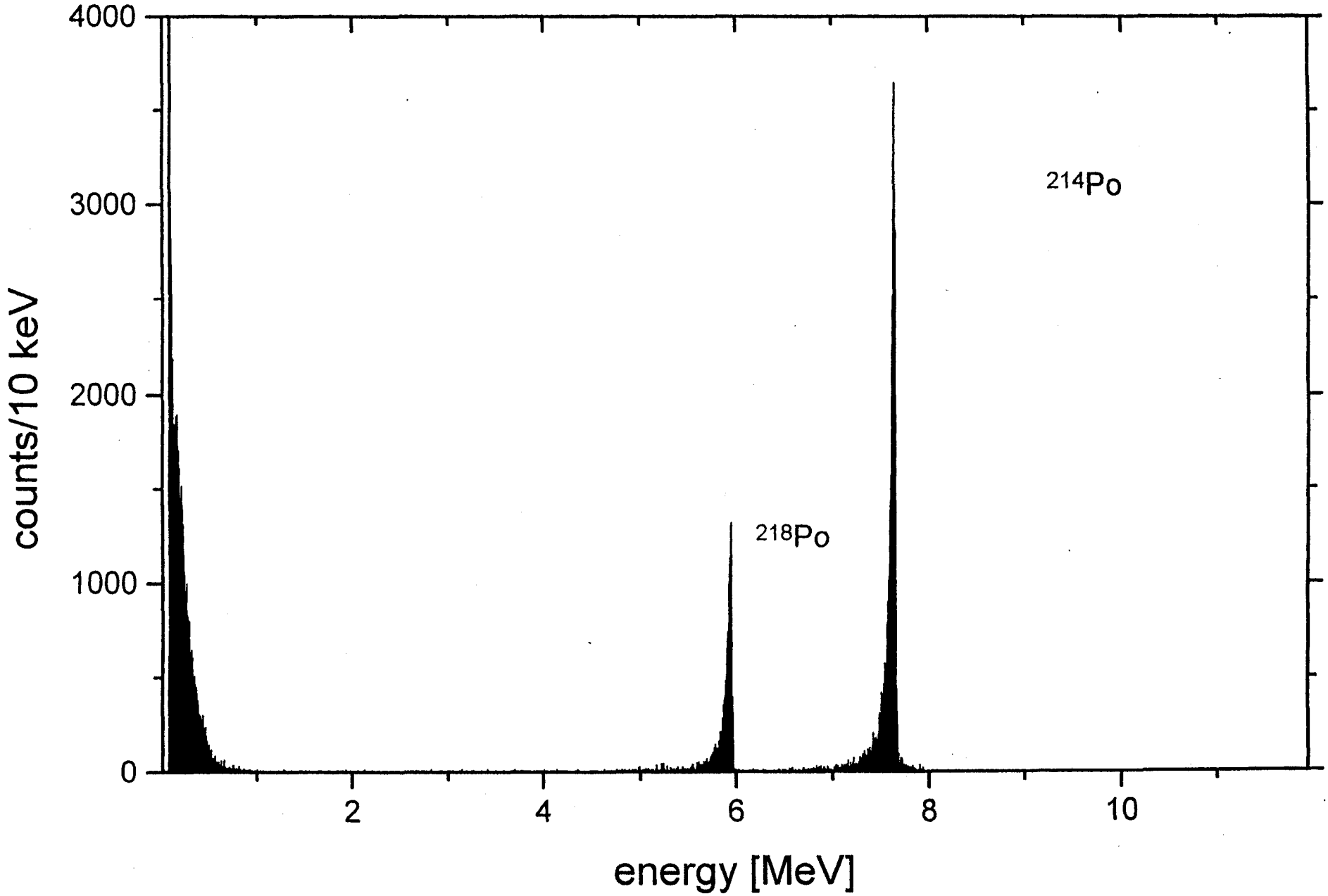
$E \cong 1/r^2$

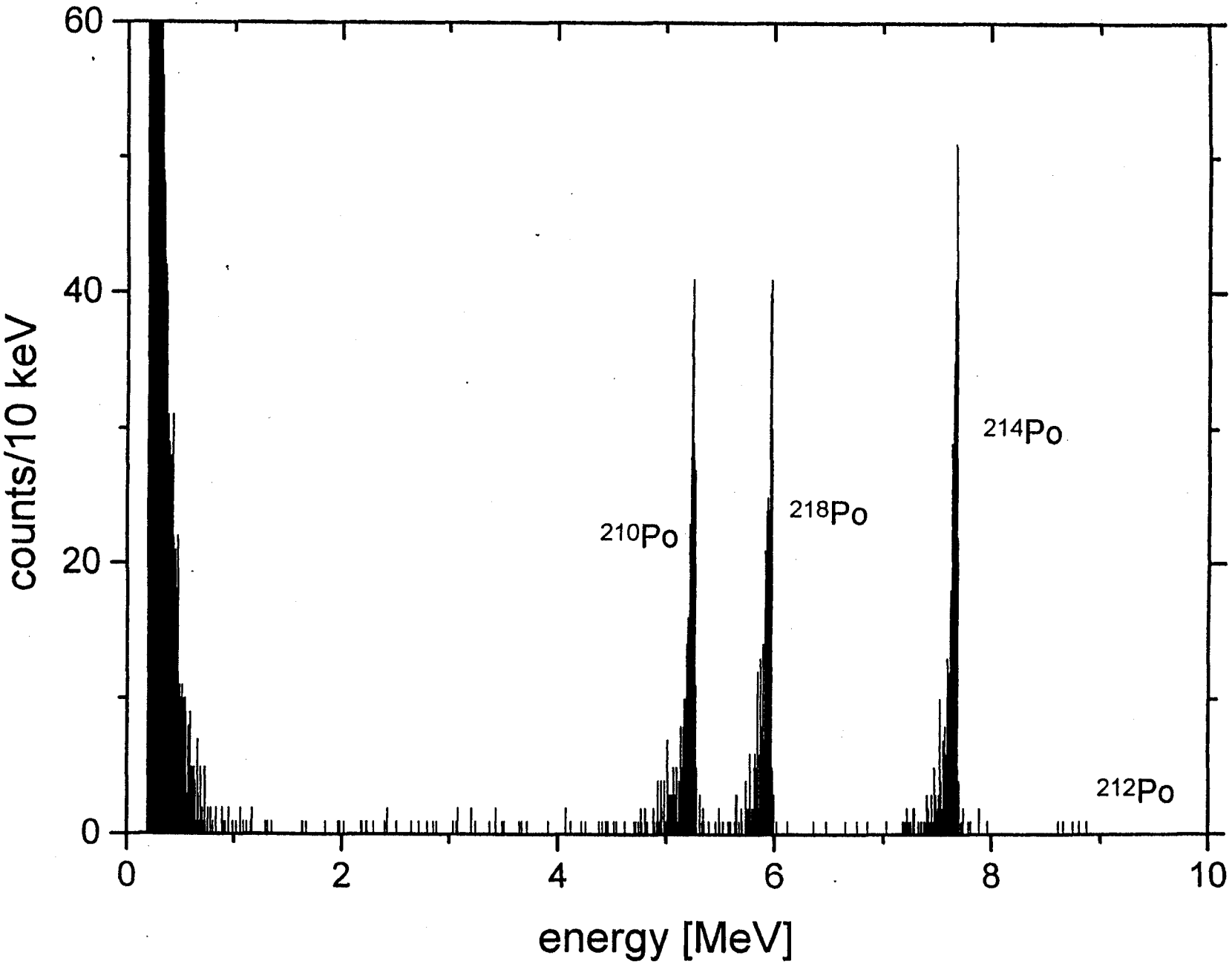
radon detector designed for BOREXINO

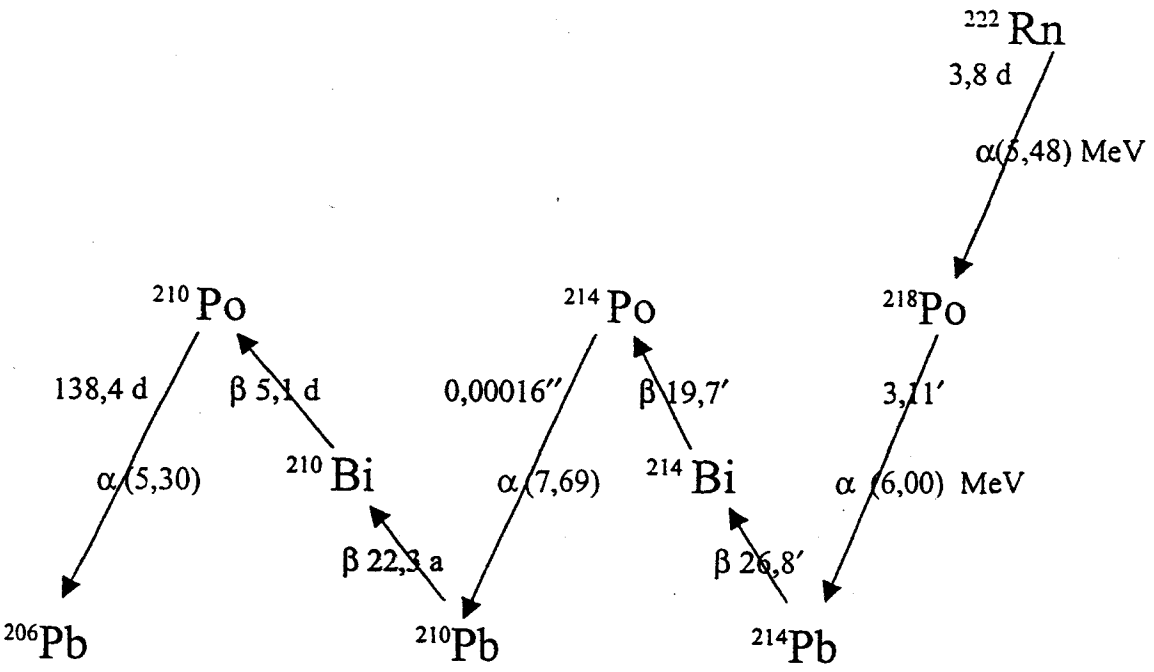


volume: 418 liters


^{218}Po
count rate [h^{-1}]
High voltage [kV]
0
20
40
60
80
100
0
5
10
15
20
25
30
35


4000
3000
2000
1000
0
counts/10 keV
2
4
6
8
10
energy [MeV]
^{214}Po
^{218}Po


60
40
20
0
counts/10 keV
0
2
4
6
8
10
energy [MeV]
210Po
218Po
214Po
212Po


222 Rn
3,8 d
α(5,48) MeV
210 Po
214 Po
218 Po
138,4 d
β 5,1 d
0,00016″
β 19,7′
3,11′
α (5,30)
210 Bi
α (7,69)
214 Bi
α (6,00) MeV
β 22,3 a
β 26,8′
206 Pb
210 Pb
214 Pb

The measured **count rate Z_{218} for ^{218}Po** in counts/sec is given by

$$Z_{218} = \varepsilon_N \, \varepsilon_{C218} \, A \, V \qquad (1)$$

with

ε_N= detection probability of the detector for α decays on the surface. Due to a simple 2π - geometry $\varepsilon_N \cong 0.5$,

ε_{C218}= collection efficiency of ^{218}Po ions on the α detector,

V= active volume of the detector, here V = 0,418 m^3,

A= ^{222}Rn activity in Bq/m^3.

The **count rate Z_{214} for ^{214}Po** is given by

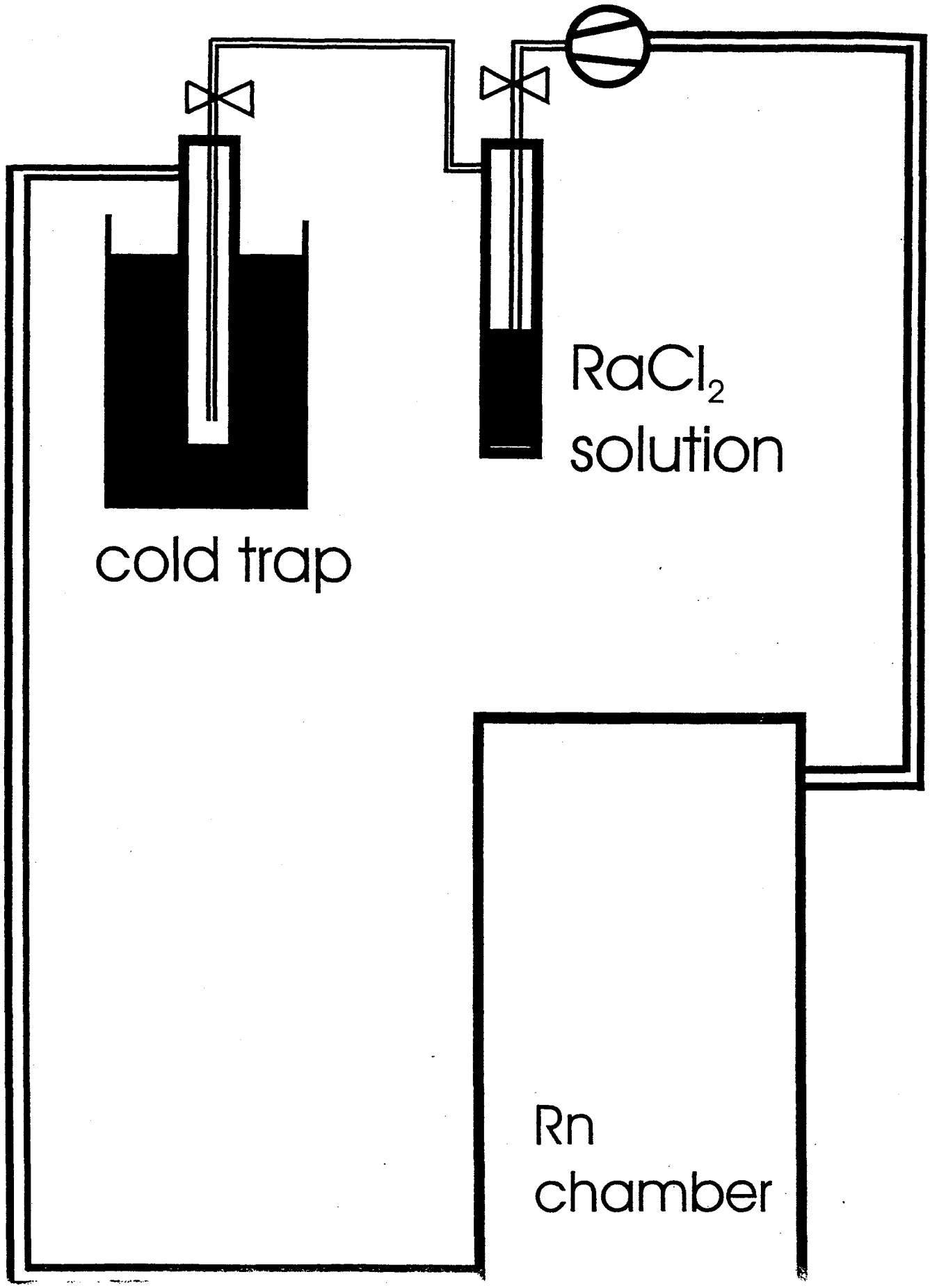
$$Z_{214} = Z_{218} + \varepsilon_N \, \varepsilon_{C214} \, A \, (1 - \varepsilon_{C218}) \, V \qquad (2)$$

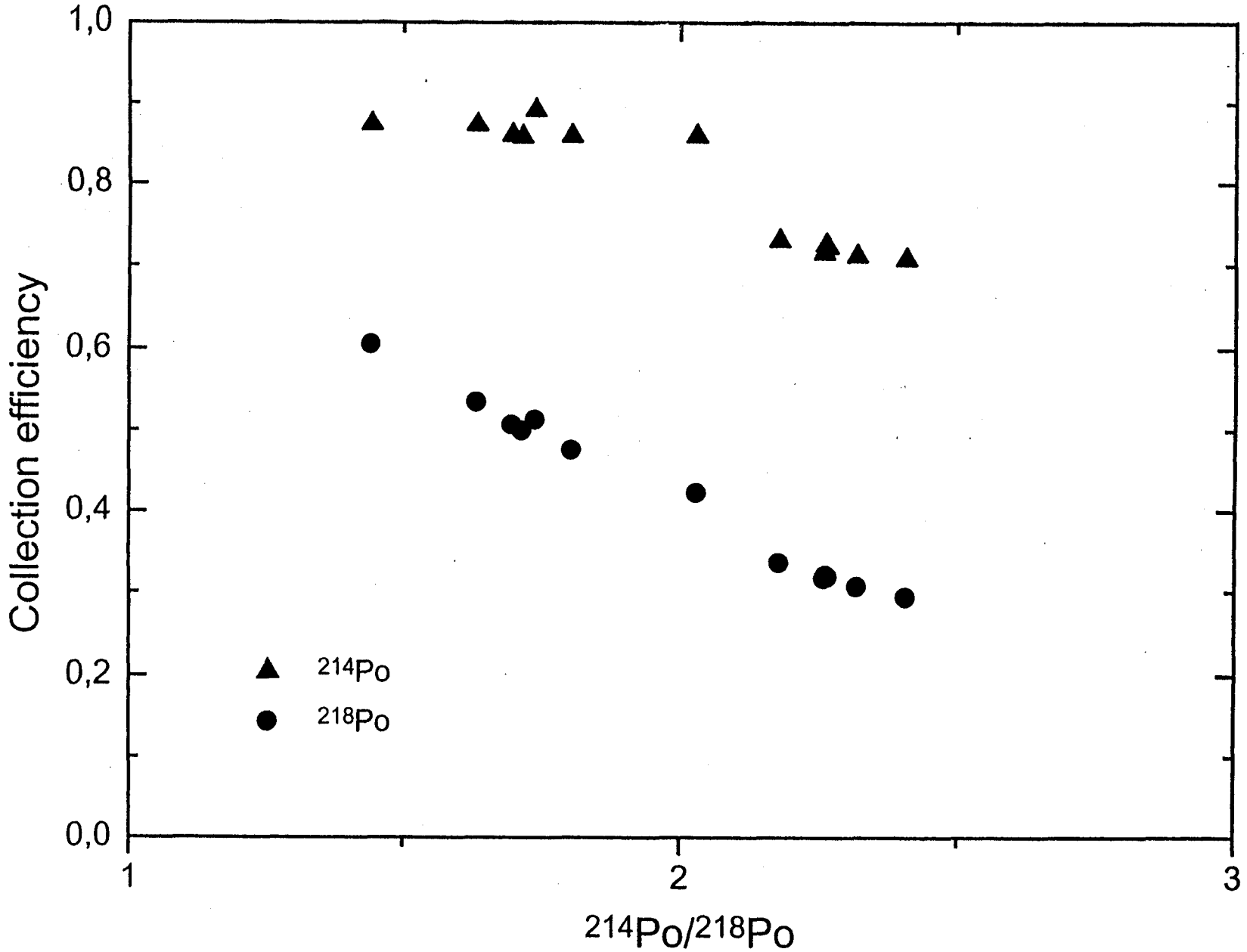
where

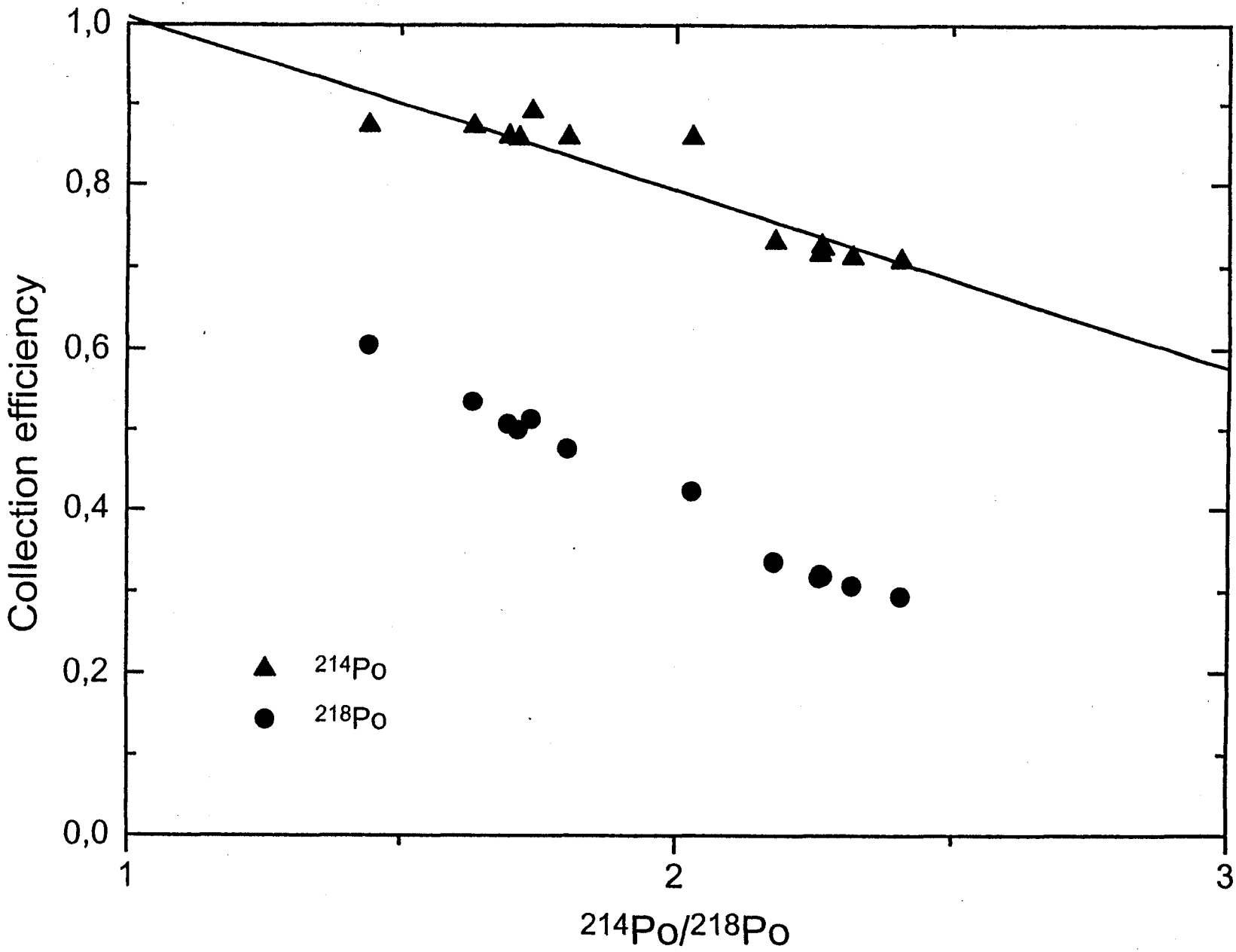
ε_{C214}= overall collection efficiency of ^{214}Pb through ^{214}Po ions on the α detector.

Combining equation (1) and (2) one gets

$$R = (\varepsilon_{C218} + \varepsilon_{C214} - \varepsilon_{C218} \, \varepsilon_{C214}) / \varepsilon_{C218} \qquad (3)$$


$RaCl_2$
solution
cold trap
Rn
chamber


1,0
0,8
0,6
0,4
0,2
0,0
Collection efficiency
1
2
3
214Po/218Po
214Po
218Po


1,0
0,8
0,6
0,4
0,2
0,0
Collection efficiency
1
2
3
214Po/218Po
214Po
218Po

The measured **count rate Z_{218} for ^{218}Po** in counts/sec is given by

$$Z_{218} = \varepsilon_N \, \varepsilon_{C218} \, A \, V \qquad (1)$$

with

ε_N= detection probability of the detector for α decays on the surface. Due to a simple 2π - geometry $\varepsilon_N \cong 0.5$,

ε_{C218}= collection efficiency of ^{218}Po ions on the α detector,

V= active volume of the detector, here V = 0,418 m^3,

A= ^{222}Rn activity in Bq/m^3.

The **count rate Z_{214} for ^{214}Po** is given by

$$Z_{214} = Z_{218} + \varepsilon_N \, \varepsilon_{C214} \, A \, (1 - \varepsilon_{C218}) \, V \qquad (2)$$

where

ε_{C214}= overall collection efficiency of ^{214}Pb through ^{214}Po ions on the α detector.

Combining equation (1) and (2) one gets

$$R = (\varepsilon_{C218} + \varepsilon_{C214} - \varepsilon_{C218} \, \varepsilon_{C214}) / \varepsilon_{C218} \qquad (3)$$

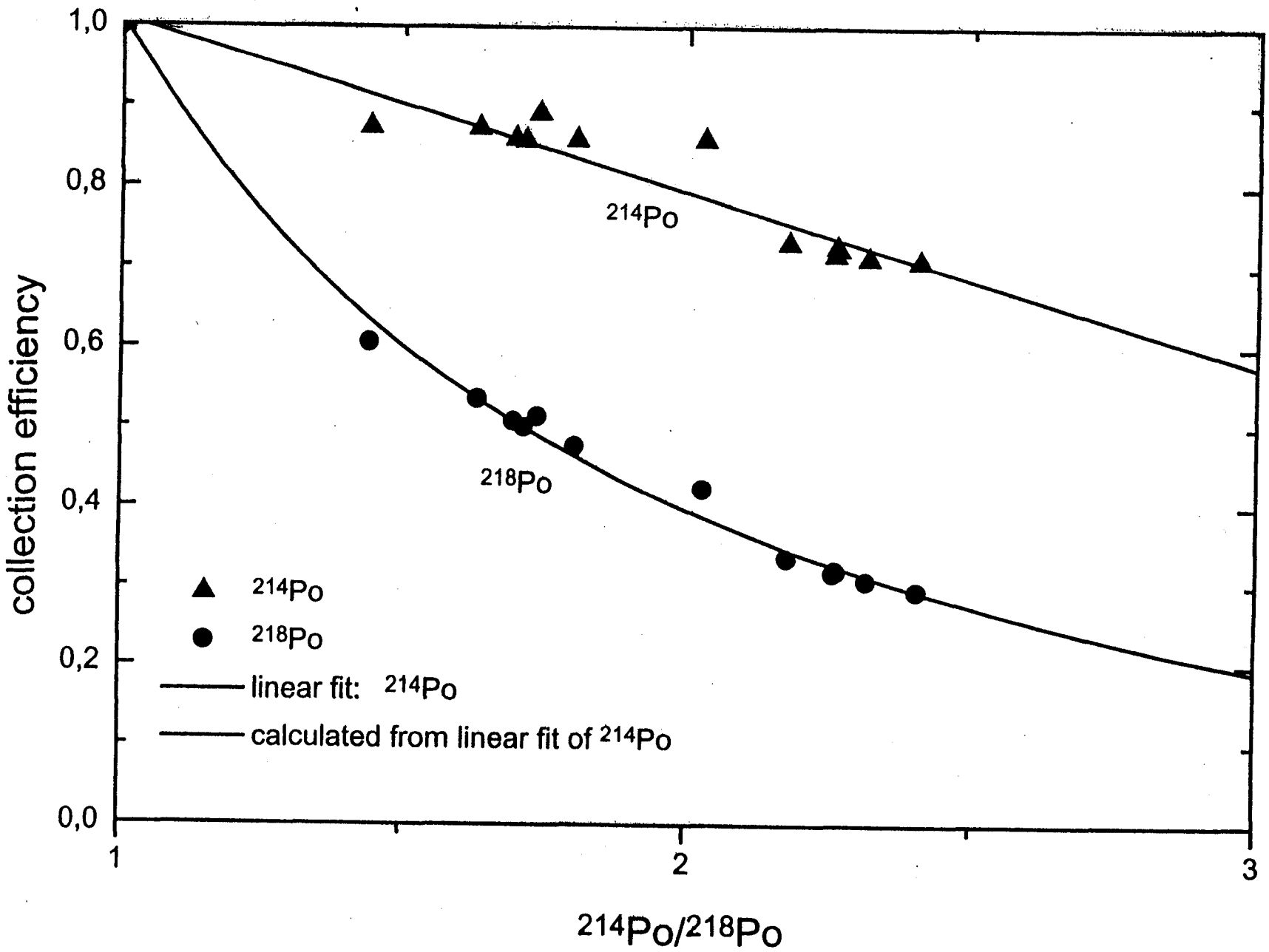
$$\varepsilon_{C218} + \varepsilon_{C214} - \varepsilon_{C218} \, \varepsilon_{C214} = a - b \, R \qquad (4)$$

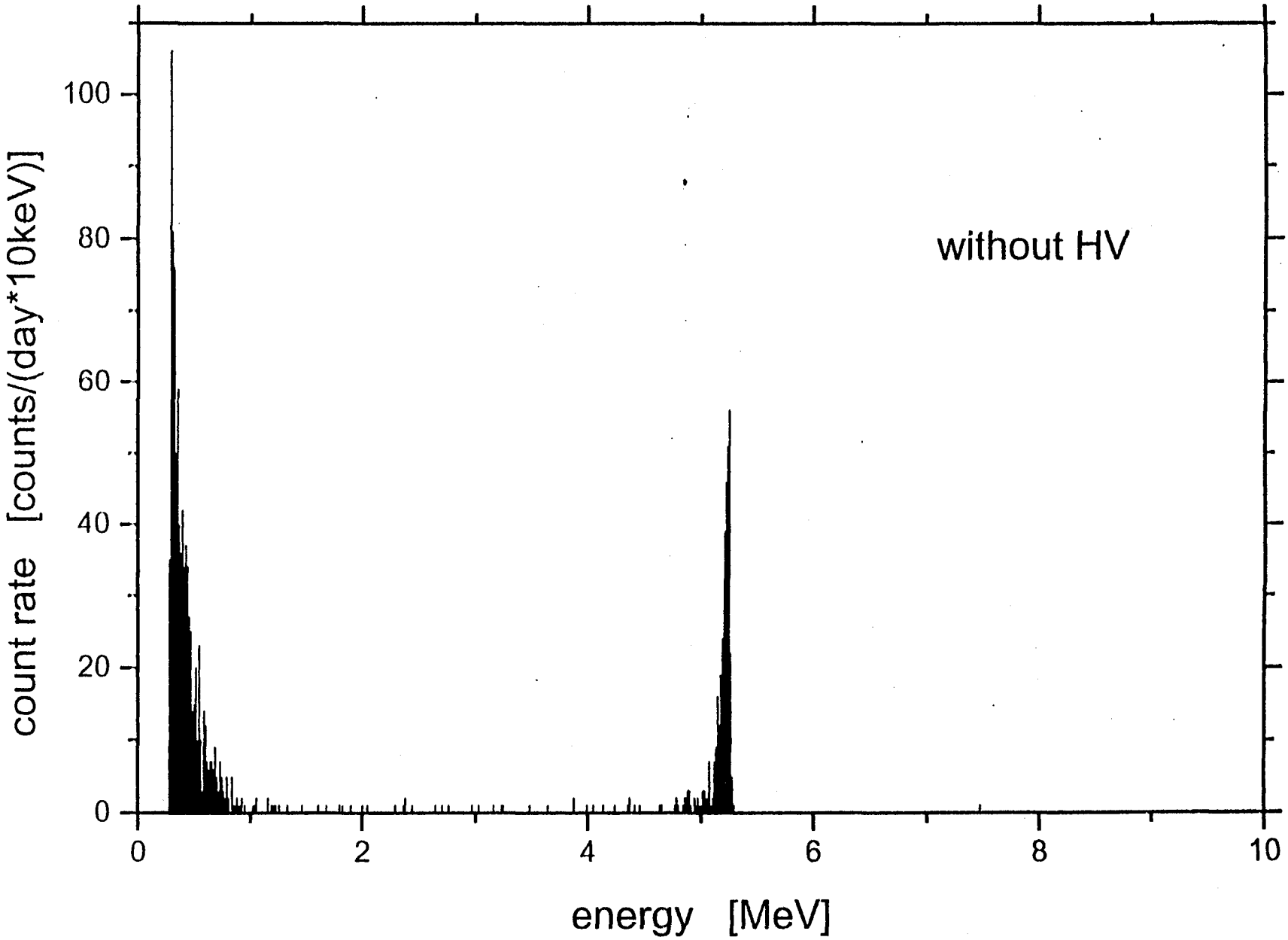
with

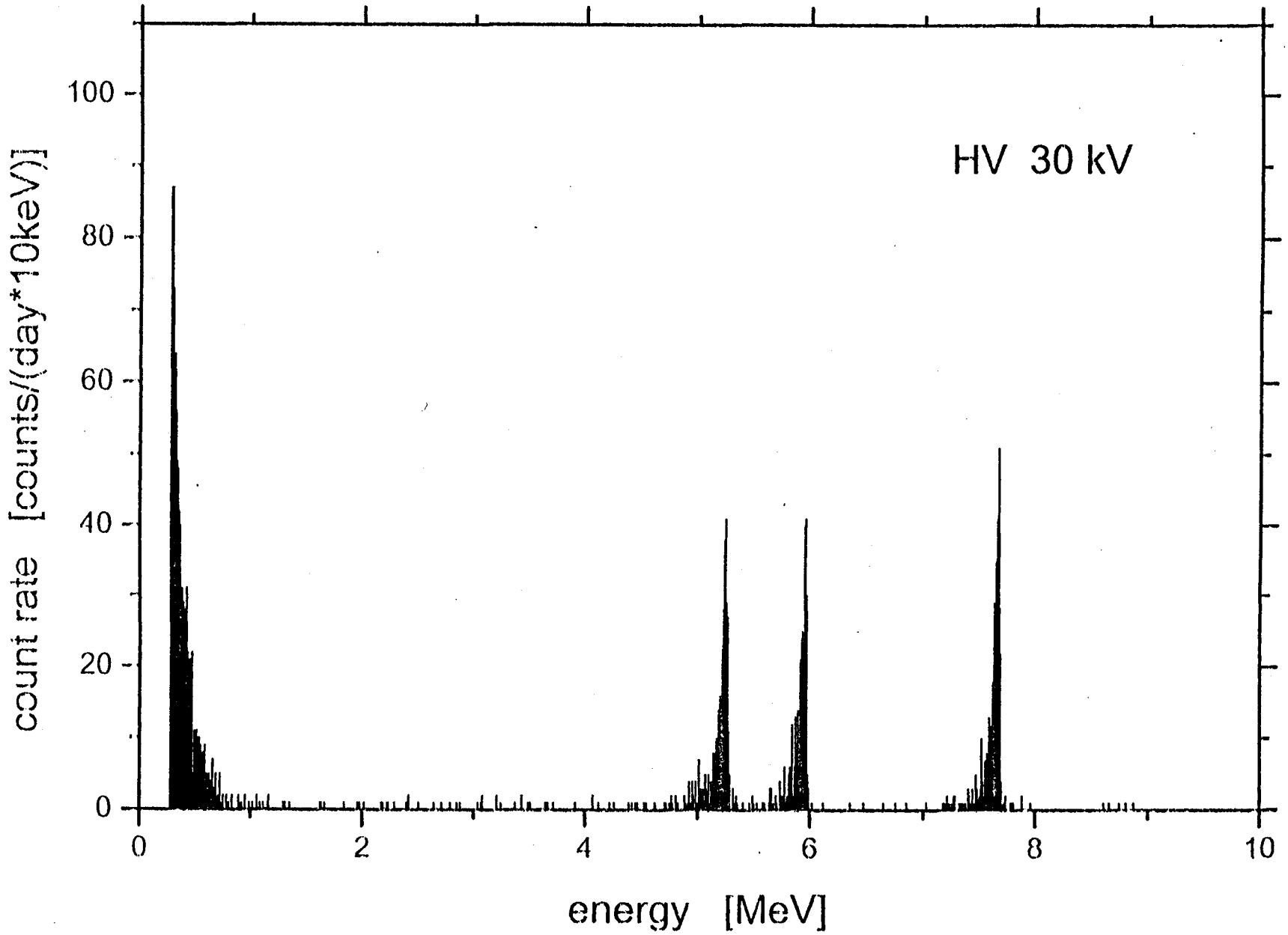
a = 1.225 ± 0.039 and b = 0.216 ± 0.020 .

And with equation (3) and (4) one gets

$$\varepsilon_{C218} = a/R - b \qquad (5)$$


1,0
0,8
0,6
0,4
0,2
0,0
collection efficiency
1
2
3
214Po/218Po
214Po
218Po
214Po
218Po
linear fit: 214Po
calculated from linear fit of 214Po


without HV
count rate [counts/(day*10keV)]
energy [MeV]
0
20
40
60
80
100
0
2
4
6
8
10


HV 30 kV
count rate [counts/(day*10keV)]
100
80
60
40
20
0
0
2
4
6
8
10
energy [MeV]

Background

Noise from detector and amplifier	**small compared to ionisations signal**
Noise from high voltage	**small compared to ionisations signal**
Electromagnetic pickup and ionization background within the region of interest	**depends on environment** **in Heidelberg ~ 1 event/day**
Intrinsic Rn - emanation within the detector volume (equilibrium)	**1,7 mBq**
At normal operation condition the detector vessel is flushed with 200 l/h of air sample	**< 15 μBq ^{222}Rn activity**

Total background: **< 70 μBq/m^3 Rn equivalent**

This is the detection limit if one accept the definition signal/ background =1

Conclusion

- **The described Rn – Monitor allows ^{222}Rn measurements in air at concentration levels of mBq/m^3.**
  The statistical error is ≈20 % for 1 day data taking at 1 mBq/m^3 ^{222}Rn.

- **The ratio $^{214}Po/^{218}Po$ indicates the degree of air contamination with ion neutralizing chemical agents and**
  can be used for correction of basic calibration.

- **The collection efficiency of ions for this detector is rather high.**
  Higher sensitivity for this kind of detectors can be achieved by increasing the detector volume.
  A detector with ≈2 m^3 active volume seems to be feasible.

Radon-Backgrounds in Rare-event Experiments

Pre-Conference Workshop
NEUTRINO 2000, Sudbury / Canada

Measurements of Rn/Ra in Water for BOREXINO

H. Simgen[1)], M. Balata[2)], M.G. Giammarchi[3)], G. Heusser[1)], T. Kirsten[1)], M. Laubenstein[2)], S. Nisi[2)], W. Rau[1)]

1) Max-Planck-Institut für Kernphysik
Heidelberg / Germany
2) Laboratori Nazionali del Gran Sasso (LNGS)
Assergi / Italy
3) Physics Dept. of the University and INFN
Milano / Italy

- Sensitivity Requirements
- Method of Measurement
- The Radon-Concentrator
- Detection Limit
- First Measurements at LNGS

Sensitivity Requirements

Need for ultrapure water in BOREXINO:

- Scintillator purification (water extraction)
- Shielding against external gamma background

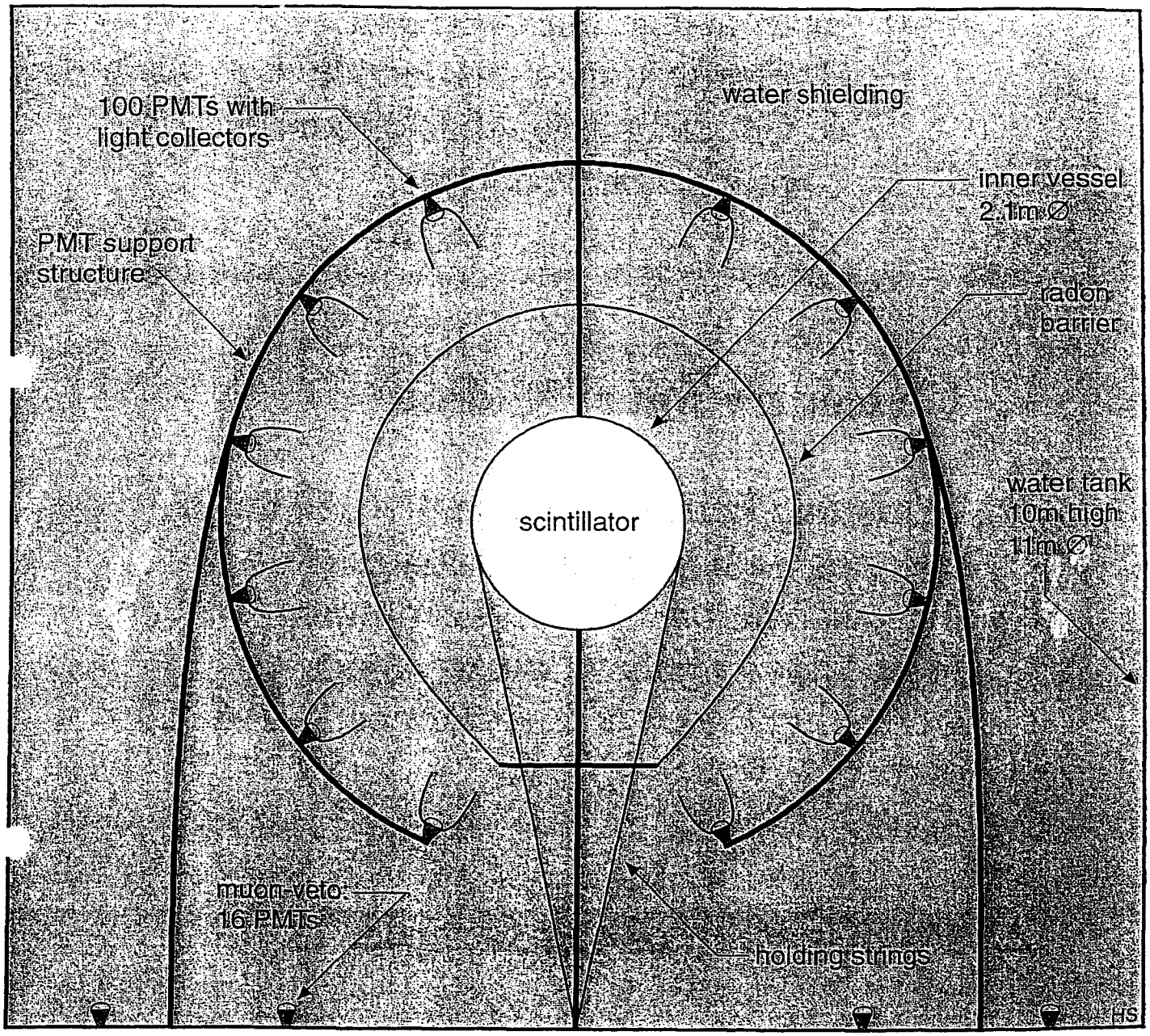
  ⇒ More critical for CTF (= Counting Test Facility) than for BOREXINO, because water is in direct contact with Inner Vessel

Limits for Rn/Ra-concentrations: ~ 10^{-3} Bq/m^3

(Compare with Rn in LNGS-water: ~ 10^4 Bq/m^3)

Therefore: BOREXINO needs a method to measure Radon and Radium in water at mBq/m^3-level!

CTF II - Design



Method of Measurement

3 Steps:
1. Concentration of Radon
2. Purification of gas sample
3. Counting in low-background proportional counters

1. Concentration of Radon

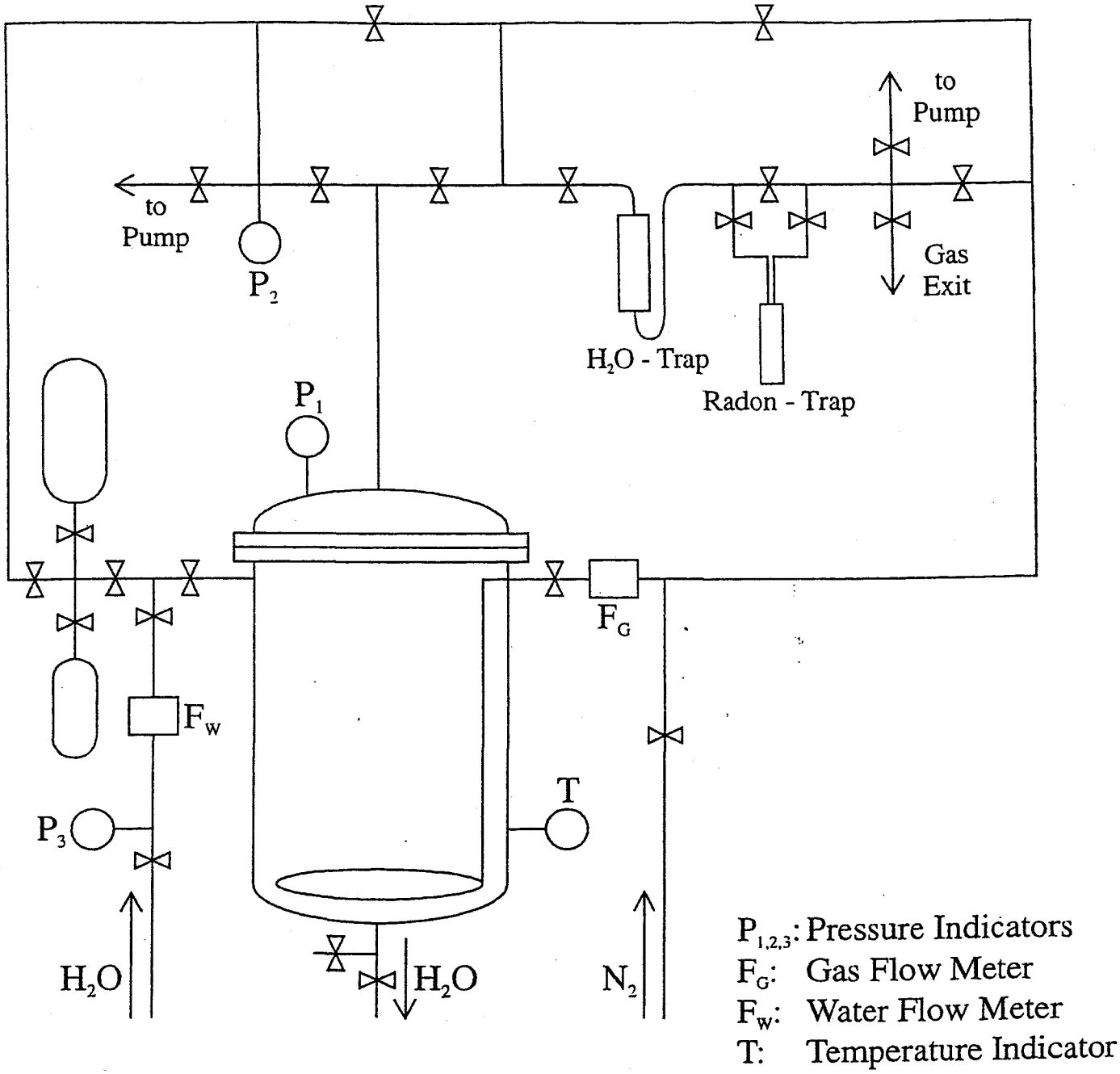
a) Extracting Radon from water (~ 350 lit.) by sparging with high-purity nitrogen (~ 5 m^3)
b) Pre-purification during concentration (especially to remove water-vapor)
c) Collecting Radon at cold charcoal (– 196 °C)

2. Purification of gas sample

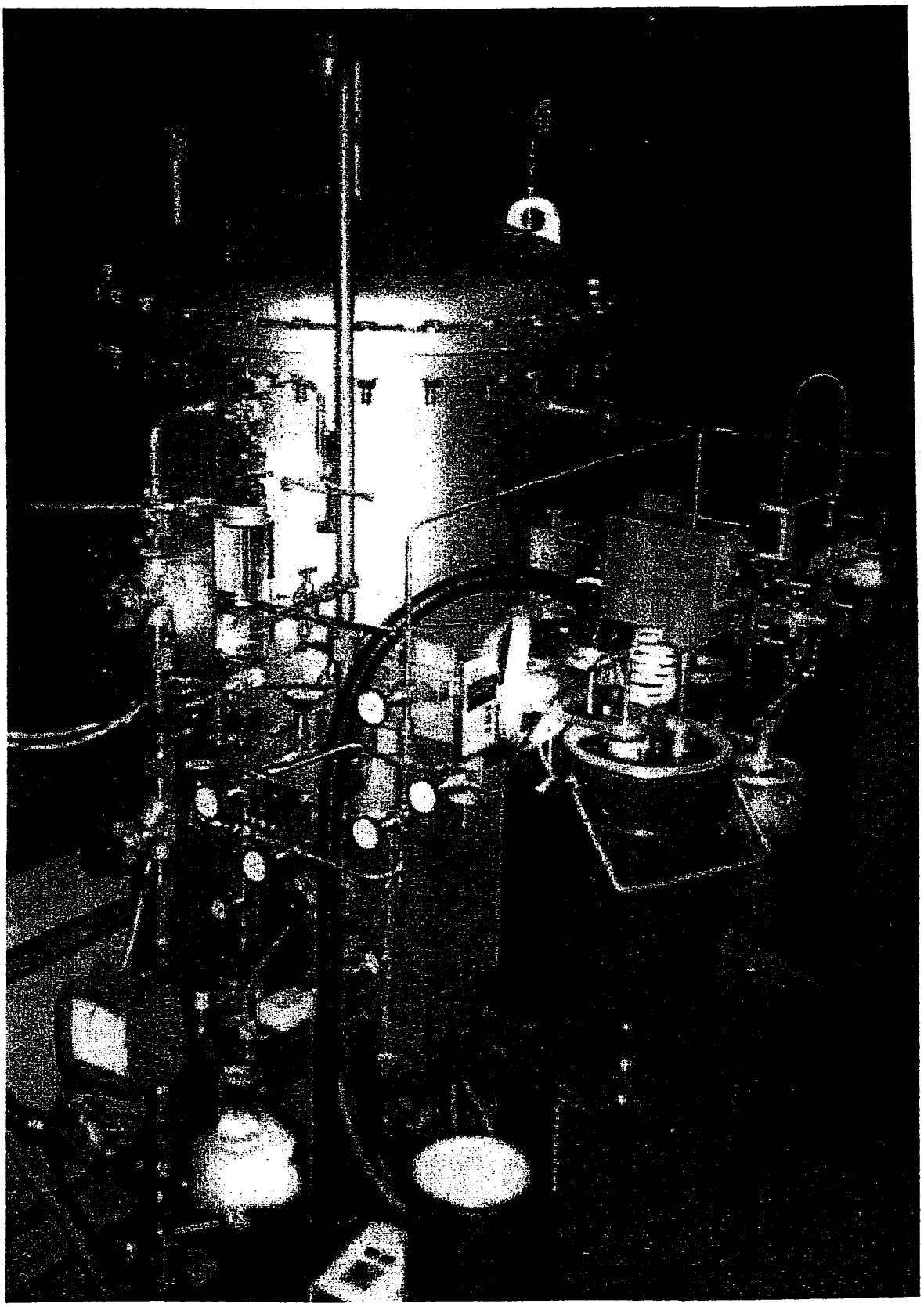
3. Counting

→ talks of G. Heusser, W. Rau

The Radon-Concentrator



Sketch of the Radon-Concentrator



Characteristics of the Radon-Concentrator

- Complete piping is made of stainless steel.
- All flanges are metal sealed.
- Water-tank and traps are electropolished.

Water-tank

- Volume: 480 liters
- Maximum overpressure: 2 bar
  ⇒ fast extraction becomes possible
  (~ 2.5 h limited by size of charcoal-trap).
- Maximum temperature: 80 °C
- Big top flange is sealed with polyurethane-O-ring and additional indium sealing.

Water-trap

- Contains ~ 100 g copper wool as adsorber
  ⇒ practically no contamination
  ⇒ good heat conductivity

- Volume: ~ 310 ml

- Adsorption of Water (and other contaminations) at – 60 °C (cooled with ethanol)

Charcoal-trap

- Contains ~ 10 g of charcoal with extremely low Radium content (~ 0.3 mBq/kg)

- Volume: ~ 80 ml

- Adsorption of Radon at –196 °C (liquid N_2)

Detection Limit

Contributions to background:

Counters:	0.1 – 2 counts per day
Sample preparation:	10 – 60 μBq
Water-tank:	1.5 ± 0.1 mBq in saturation

Residence-time of water-sample in water-tank:

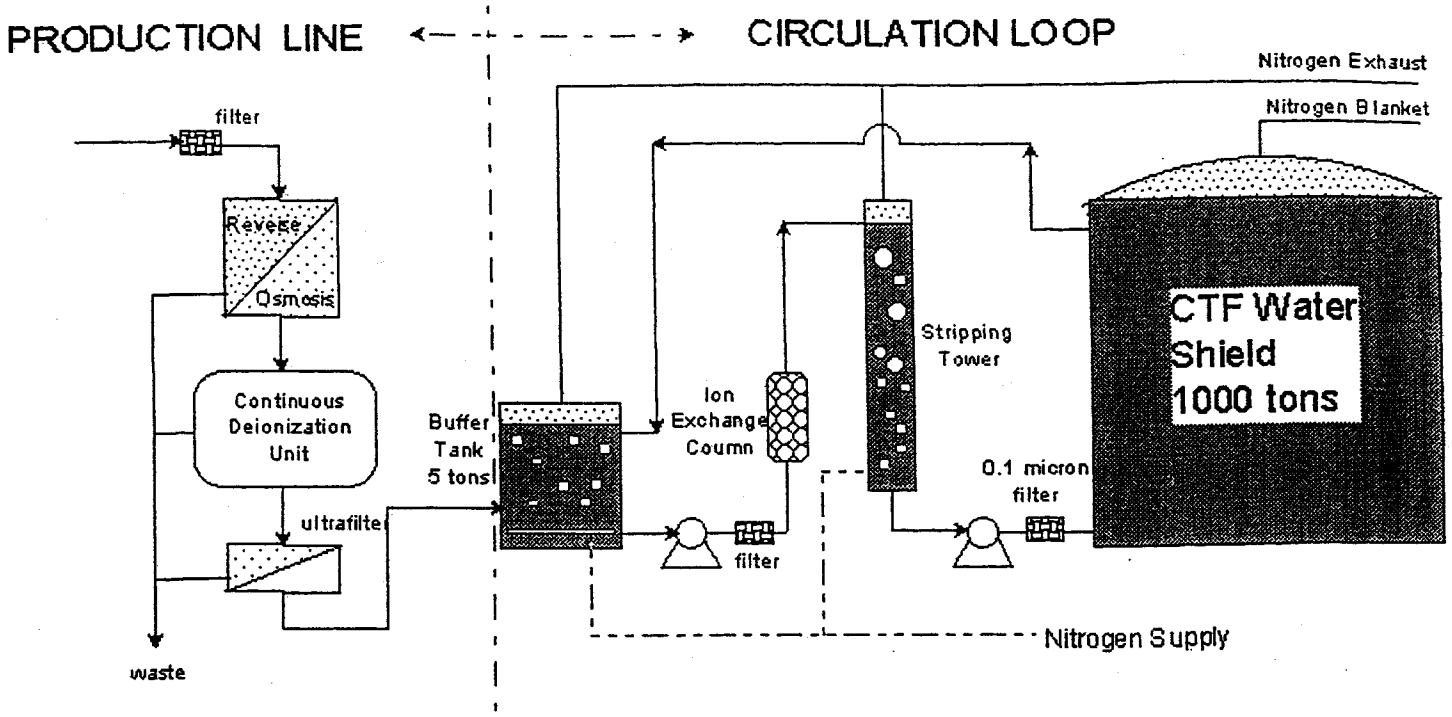
for Radon:	only extraction time (~2.5 h)
for Radium:	several days

⇒ detection limit for Radium ~ 1 mBq/m^3
⇒ detection limit for Radon ~ 0.1 mBq/m^3

The sensitivity requirements have been fulfilled!

First Measurements at LNGS

- Rn/Ra assay system was tested in Heidelberg and installed at LNGS in October 1999.
- Tests were done to prove the performance of the system (extraction-efficiency is (97 ± 4) %).
- Water purification plant for CTF at LNGS was restarted in autumn 1999.
- Delivered water quality was tested with the system before re-filling of CTF in March 2000.
- Tests to optimize working conditions of water purification plant have been done.



The water purification plant at LNGS

LNGS water-sample	H_2O-flow [m^3/h]	Radon [mBq/m^3]	Radium [mBq/m^3]
raw-water	---	$\sim 10^7$	$\sim 3 \cdot 10^5$
after some circulations	?	3.0 ± 0.4	1.2 ± 0.9
after single passage	1	187 ± 5	1.8 ± 0.6

Selected water-measurements

Summary

- BOREXINO needs a method to measure Rn/Ra in water at the mBq/m^3–level.
- An assay system was developed and tested in Heidelberg (improved version of a former system).
- The assay system fulfills the requirements.
- First measurements indicate, that the achieved water quality is sufficient for CTF/BOREXINO.

Future plans:

- Regular controls of water-quality inside CTF
- Radon emanation measurements of large samples in humid environment (e.g. nylon)

H. Simgen

Tests of O-Rings

- **Measurements of emanation-rate**
  → direct (emanation-chambers)
- **Measurements of diffusion**
  → indirect (influence on blank-activity of water-tank)

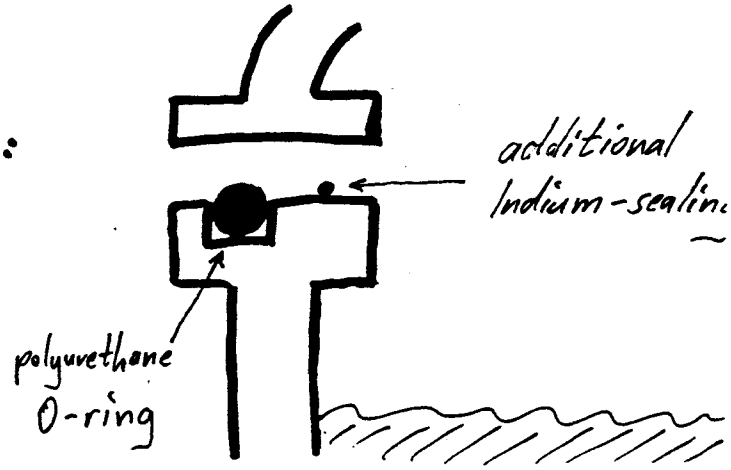
	emanation-rate [mBq/m²]	blank-activity of water-tank [mBq]	diffusion-const. [cm²/s]
viton	322 ± 8	10,9 ± 0,5*	—
butyl	13,2 ± 0,6	2,5 ± 0,2	$4{,}9 \cdot 10^{-9}$
polyurethane	<0,42 (90% CL)	2,0 ± 0,1	$8{,}8 \cdot 10^{-9}$

*leak in tank

→ blank-activity higher than expected from steel emanation-rate!

⇒ **best solution: (so far)**

→ **1,5 ± 0,1 mBq**
blank-activity



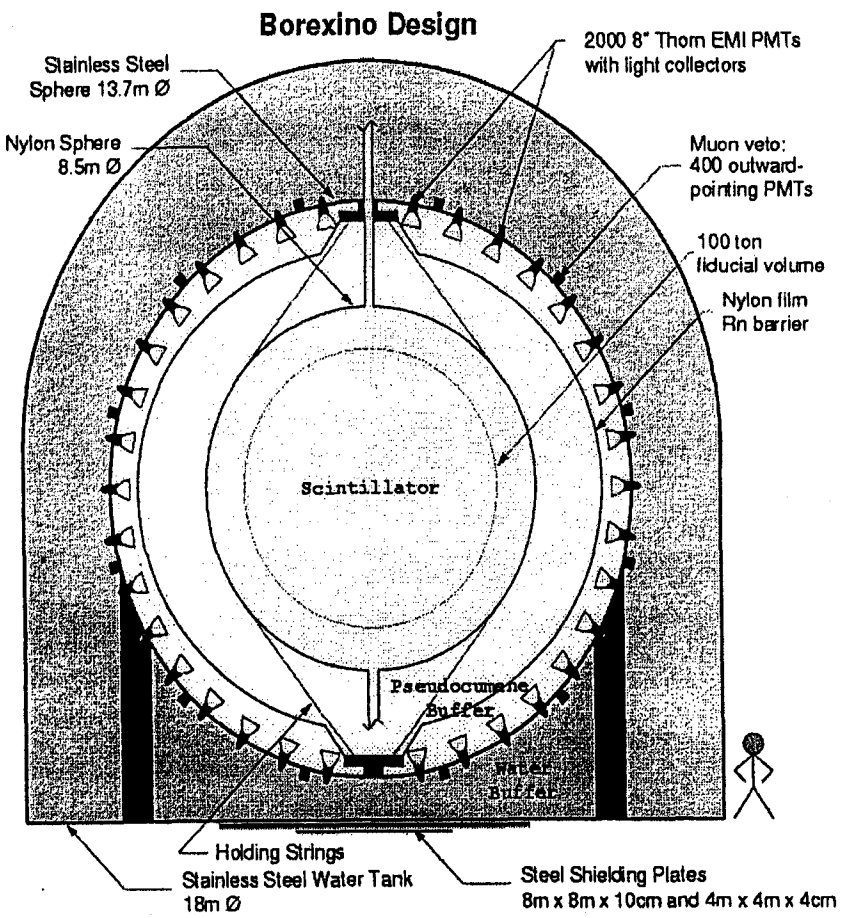
The Radon Filtering VSA System

A. Pocar, J. Benziger, F. Calaprice, E. deHaas, E. Harding, T. Shutt

Princeton University

Radon Backgrounds in Rare–Event Experiments, Neutrino 2000
Sudbury, 14th June 2000

Radioactive Contamination Requirements for the Borexino Inner Vessel



Limits on ^{210}Pb contamination:

< 1 count/day

in the 300 tons of scintillator
(from 1.17 MeV β–decay of ^{210}Bi)

→ minimize exposure time of the nylon

→ low radon levels in the air of the assembly site

Inner Vessel Assembly: the Princeton Clean Room

Clean room main characteristics: class 100, volume=700 m^3
needs ~170 m^3/h makeup air

Contamination mechanism: ^{222}Rn in the clean room air
↓
plateout onto nylon surface and buildup of ^{210}Pb
(depends on the exposure time)
↓
washoff into the scintillator (amount not known)

Worst case scenario (100% plateout + 100% washoff):

~ 1000 counts /day/hour of exposure
(for a typical value of indoor ambient air of 20 Bq/m^3)

Radon reduction: $\geqslant$ 1000

Air Filtering Technique I: Adsorption on Activated Carbon

Adsorption: phenomenon by which particles in a fluid experience a drop in potential energy associated with their interactions (van der Waals, dipole, quadrupole) with a solid surface. This allows them to concentrate and "stick" to the surface, no longer being in the gas phase; any gas is adsorbed on any solid at low enough temperature and high enough partial pressure (similar to condensation of liquids)

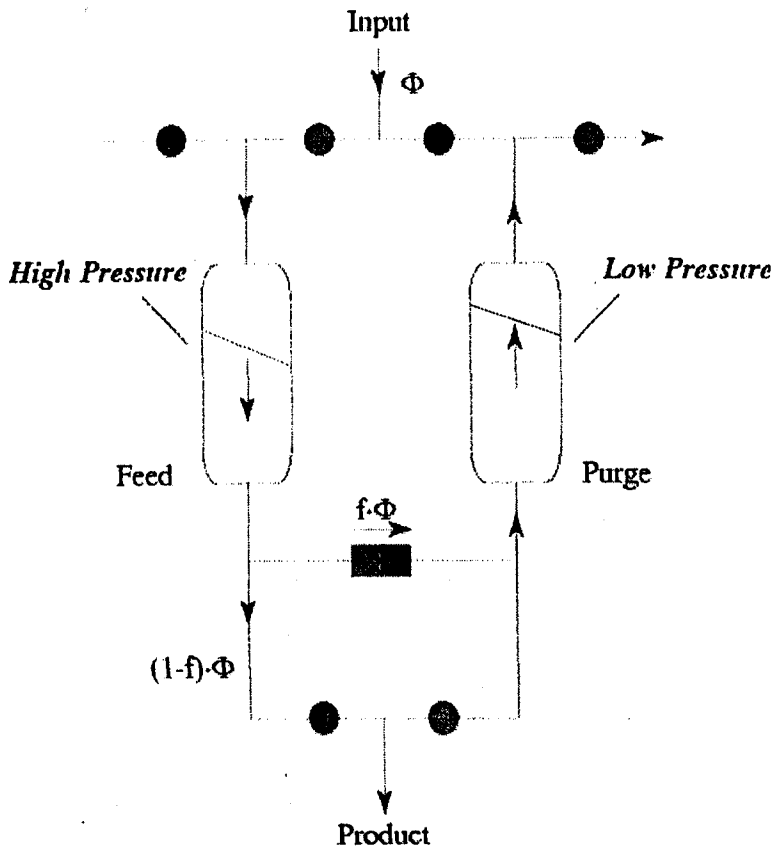
Activated carbon: graphite specifically treated to obtain a substance with an enormous surface to volume ratio ($50 \div 1000\ m^2/g$, pore diametre of a couple of molecular radii). Preferably adsorbs radon than the other components of air (N_2, O_2, Ar, ...)

→ cheap, good mechanical properties

→ Need regeneration of carbon to avoid saturation

Air Filtering Technique II

Activated Carbon–Based Pressure Swing Adsorption (PSA) System



Principle

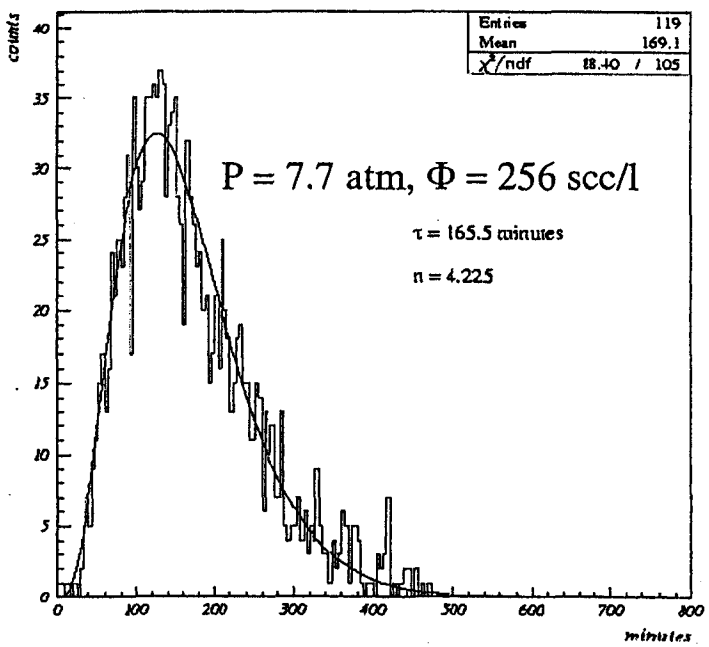
use the pressure dependent adsorption properties of activated carbon, namely its bigger adsorption capacity at higher pressures, in a two–filter system which undergoes a feed/purge cycle (widely used in air separation processes)

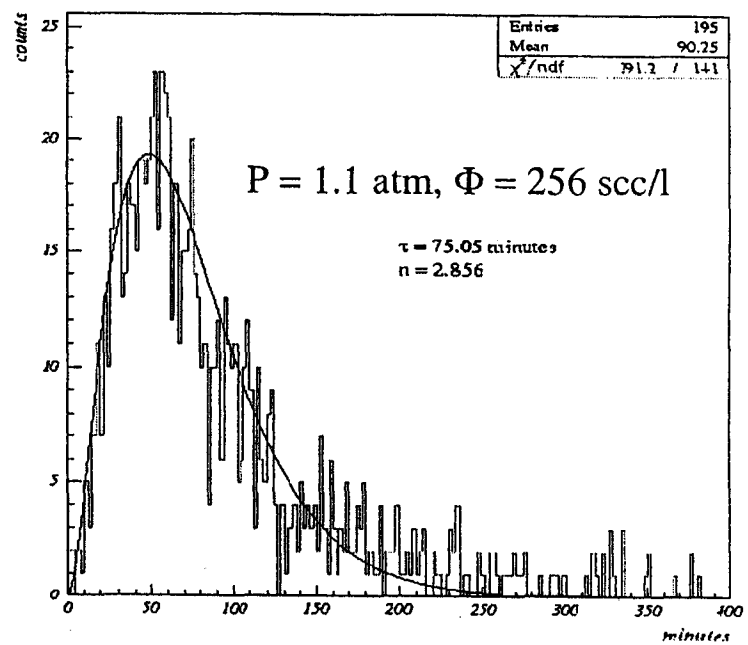
Gain

$$\frac{\text{volume flowrate (pressure) ratio}}{\text{adsorption constant ratio}}$$

Single Column Elution Curves

Studies of the charcoal properties were performed using a single flow–through column and the basic parameters, τ (mean breakthrough time) and n (number of equivalent theoretical stages), were measured at different pressures and at different flow rates via elution curves





Output for an input spike (δ–function): $y(t) = \frac{n^n}{(n-1)!} \left(\frac{t}{\tau}\right)^{n-1} e^{-n\frac{t}{\tau}}$

Plate Model for Adsorption

The column length is divided into n equal equilibrium stages (plates); equilibrium at every stage is expressed by:

$$s_j = K \cdot y_j \qquad j = 1, \ldots, n$$

y = concentration of Rn in air (volume^{-1})
s = concentration of Rn on the carbon (area^{-1})

Evolution equation:

$$\Phi\,(y_j - y_{j-1}) = -a\,(\partial_t s_j)$$

a = surface area per stage

The characteristic breakthrough time τ is given by ($k_a = \sigma K$, σ=surface area per unit mass) :

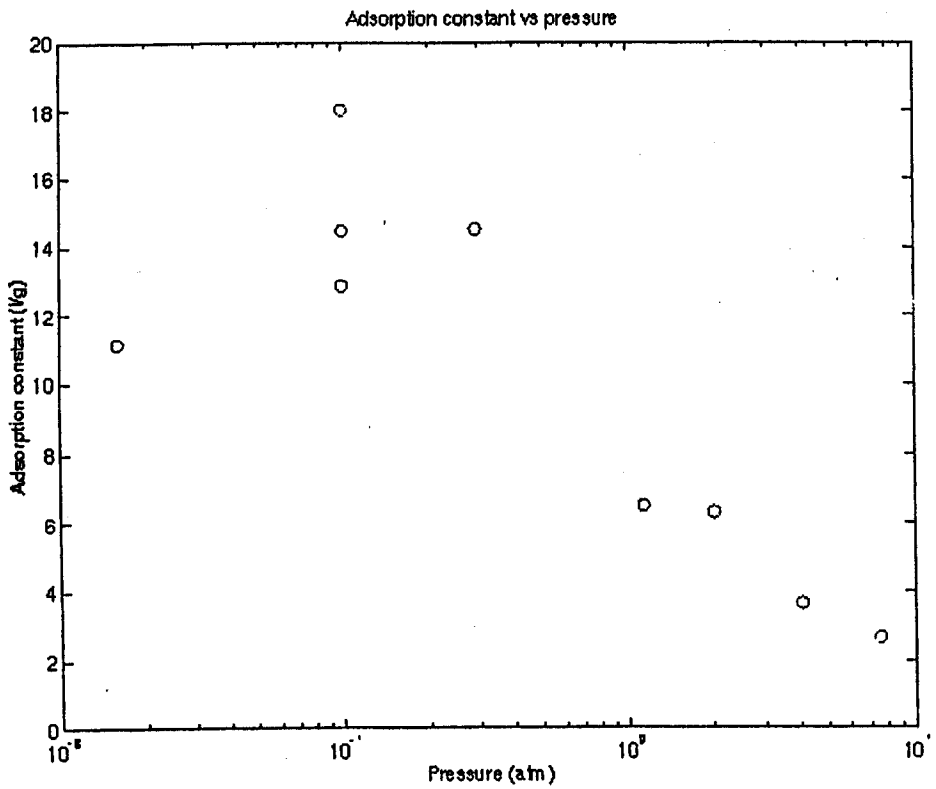
$$\tau = k_a \cdot m/\Phi$$

Φ = volume flow (l/s)
m = mass of carbon (g)

k_a = adsorption constant (l/g)

Adsorption "Constant"

The adsorption constant shows a non negligeable saturation effect with pressure, which significantly lowers the expected adsorption efficiency at high pressure (a factor of ~3 is lost at 8 atm); there seems to be a pressure–dependent competition between Rn and N_2



A PSA system operated between 1 and 8 atmospheres and at 0.3 m^3/h flow rate has been shown to be somewhat inefficient, requiring a 75% purge fraction to achieve 99% radon removal (N. Darnton, 1997), proving it to be very impractical to scale it up to the dimensions needed for the clean room.

Vacuum Swing Adsorption

Physical principles upon which it relies are conceptually identical to those of a Pressure Swing Adsorption system

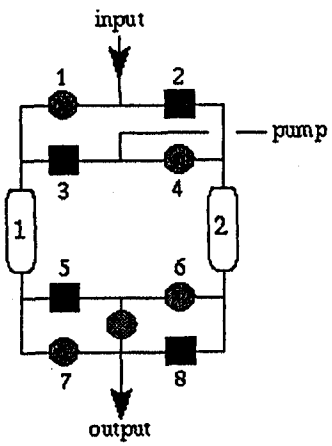
Using subatmospheric pressures in the cycle is advantageous because of the large volume flowrate gains available for the purge with respect to the feed (up to ~100)

A small scale Vacuum Swing Adsorption (VSA) system with the following specifications has been operated:

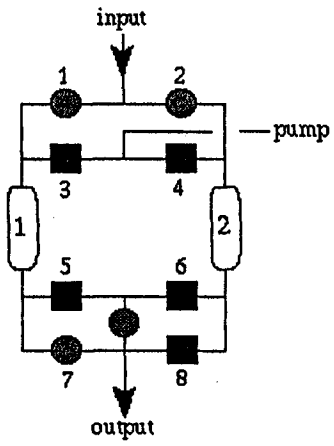
→ Flow ~ 1.7 m^3/ hour (1% of the final system)
→ Mass of carbon = 1350 g/column
→ Volume of each column ~ 4 litres
→ Continuous radon input from a 252 Bq/min flow–through source

The Vacuum Swing Cycle

Feed cycle 1 (t1)

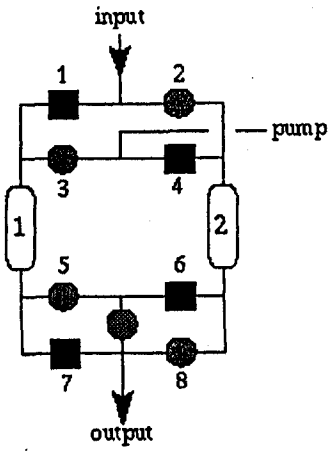


Repressurization 1 (t2)

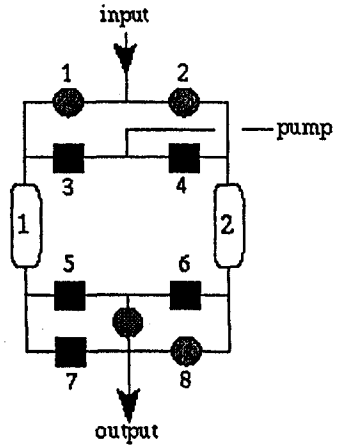


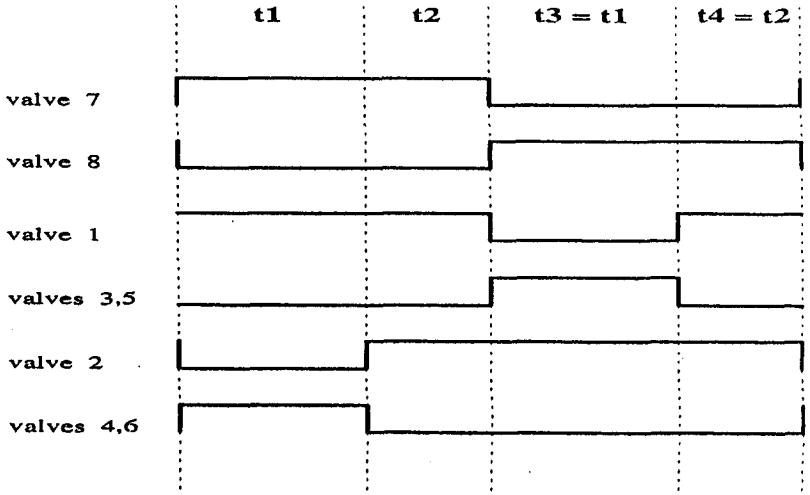
The cycle the system undergoes is made up of four steps. While one column is being fed, a fraction of its output is pushed back into the other column, thus purging it. Before the fed and purged columns exchange their roles, time is given for the evacuated column to repressurize, in order to avoid backflow of gas from the output.

Feed cycle 2 (t3 = t1)



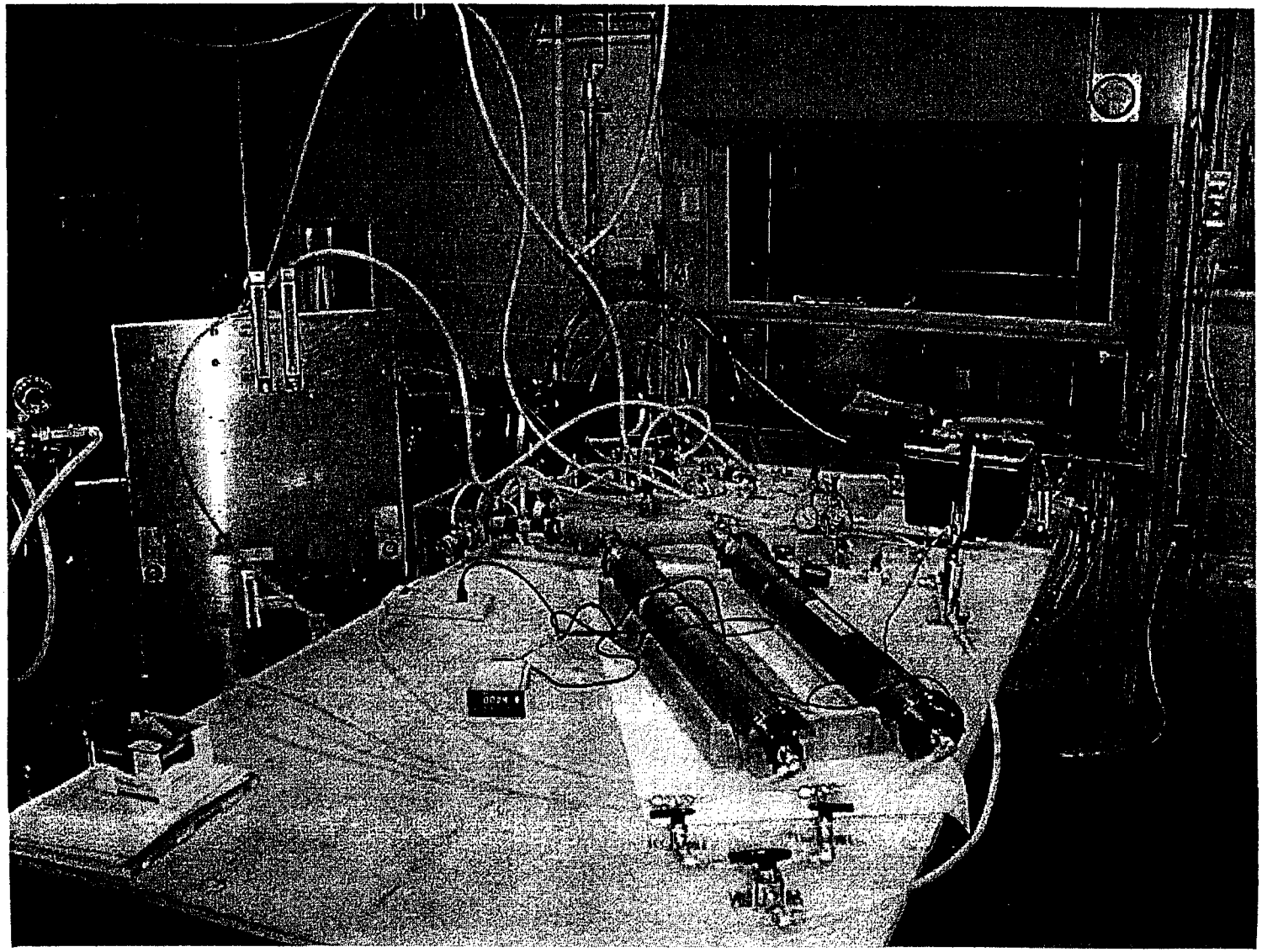
Repressurization 2 (t4 = t2)



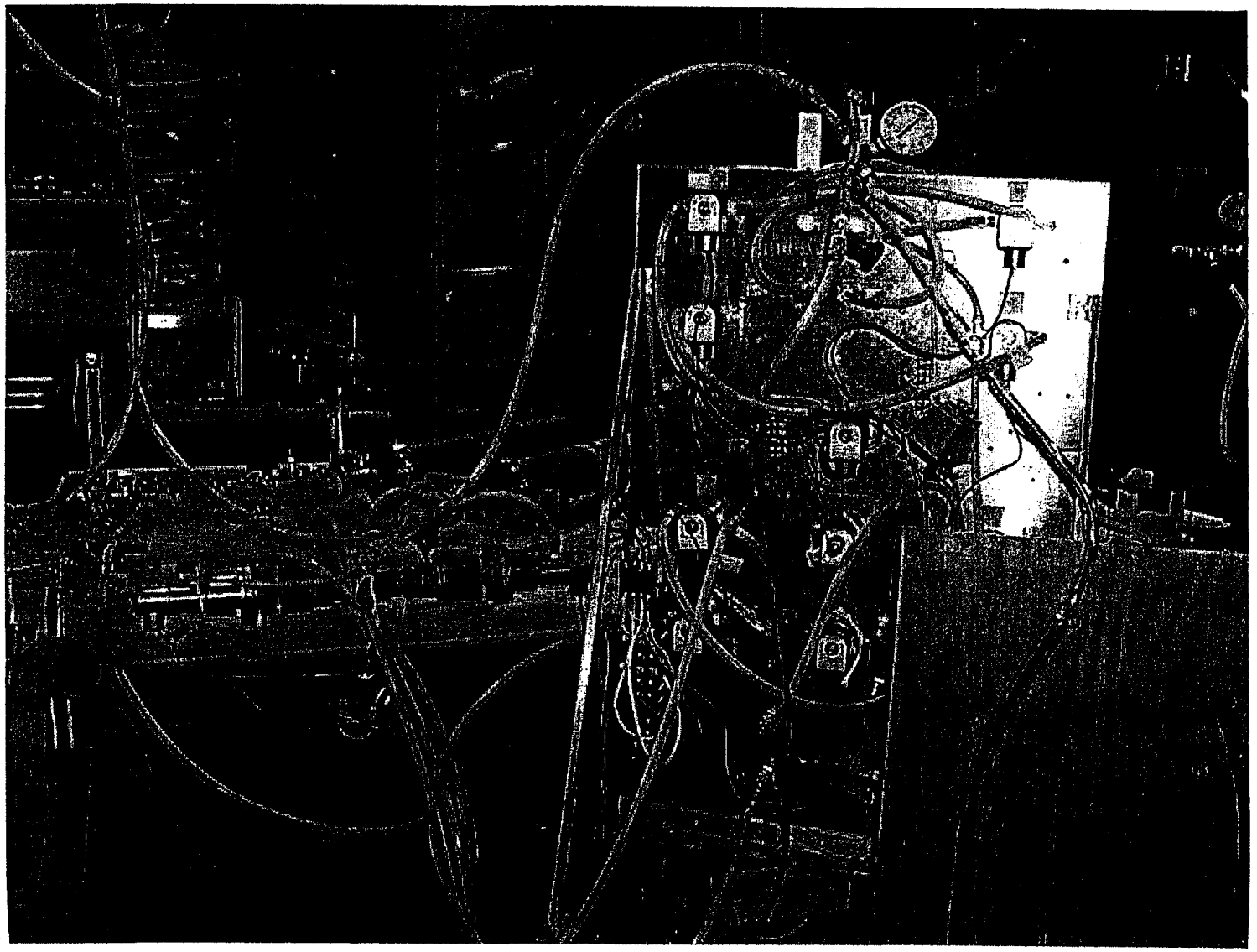


t1 + t2 = 10 minutes, t2 = 40 seconds

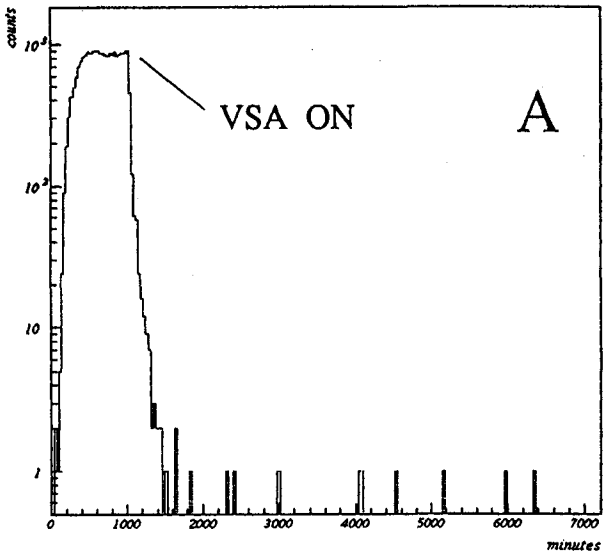
The VSA Prototype



The VSA Prototype

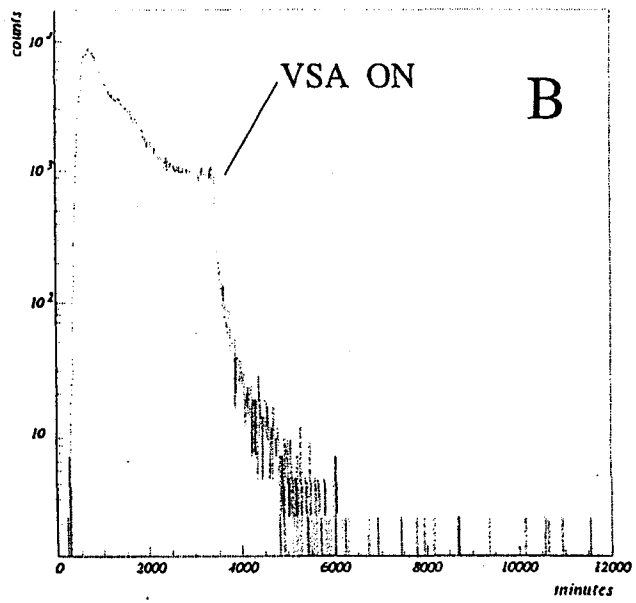


First Results from VSA system



Radon concentration reductions at the the required levels have been proved possible with the small–scale system

Reduction factor achieved: $> 10^4$



Contamination of one (A) or both (B) columns gives the same final concentration (steady–state)

Run parameters: Input flowrate: 1.7 m^3/h (28 sl/min)
Purge fraction: 10%
Feed time: 10 minutes
Repressurization time: 40 seconds
Feed pressure: 900 Torr (1.2 atm)
Purge pressure: 10–25 Torr

Characterization of the System and Choice of the Vacuum Pump

The above successful runs performed with a purge to feed volume flowrate ratio of 7.5. Can this be relaxed?

Several runs with purge to feed volume flowrate ratios ~3.5 (obtained with different combinations of purge pressure and purge fraction, after extensive calibration of the system) gave radon reduction factors ≥ 1000

Problem: pressure drops across the low pressure column limit the overall effective pumping speed obtainable

=> 850 m^3/h vacuum pump (volume flowrate gain of 5)

P = 1 ÷ 5 Torr

purge fraction = 1 ÷ 3.5 %

Full–Scale VSA System

→ design essentially complete

→ vessels, vacuum pump charcoal and parts ready to be ordered

→ should be complete by Fall 2000

Parameters: mass of carbon: ~ 135 kg/column

volume of each column: ~ 400 litres

(r=38 cm, length=1 m to contain pressure drops)

Good News ? (preliminary ...)

Plateout: nylon samples were exposed to high radon concentrations (~40 kBq/m^3) for some weeks in a mock–cleanroom. The 5.3 MeV α from ^{210}Po was measured after some months with a Si diode detector.

$$f < 1\%$$

(f=fraction of Rn daughters created above the sample)

Washoff: exposed nylon samples were exposed to Rn

i) cleaned with detergent and soaked for 10 hours in PC:

$$\text{washoff} = 0 \pm 2\ \%$$

ii) soaked for 10 hours in PC:

$$\text{washoff} =$$

A VSA Device as a Possible Low–Level Radon Detector

Principle: use the dependance of the mobility of radon on a charcoal bed on the volume flowrate and the large purge to feed pressure ratio to obtain a radon–concentrated air stream to measure

How:

i) feed a small amount of cleanroom air through one column
ii) radon will concentrate in the first portion of the bed
iii) purge the second column at very low pressure
iv) collect the purge gas, rich in radon, compress it back to atmospheric pressure and measure it
(=> radon concentrator, by a factor ~ feed to purge mass flow)

Advantages:

i) use existing knowledge in the group
ii) compact and cost–effective (reuse many components)
iii) should work with the radon concentration expected in the cleanroom

Radon Activity in the Clean Room: How to Monitor it ?

All runs with the prototype have been performed with a source at the input so that the radon concentration of the air stream was much higher than that of ambient air

Does this scale down linearly to low concentrations?

→ it should, since the radon partial pressure is low enough to prevent any saturation effect

→ have to worry about activity of the charcoal, ...

A direct measurement of the activity of the cleanroom air would be a very important achievement

→ our commercial solid–state detector can continuously measure radon concentrations ~ 0.1 Bq/m^3 or higher

Conclusions

A small–scale VSA radon filter has been operated and radon reductions of 10^4 have been achieved

A large–scale VSA system for supplying air to the cleanroom where the assembly of the Borexino nylon vessels will take place has been designed to achieve radon reductions $\geq 10^3$

Preliminary results, if confirmed, show that plateout of radon daughters on nylon and subsequent washoff into the scintillator are far from the "worst case" scenario

A VSA device could be used as a radon concentrator to continuously monitor the activity of the cleanroom air once the air–filtering system is operational

EXPERIMENTAL LIMIT ON Rn-222 ACTIVITY IN LIQUID Ar

V.D.Ashitkov[1], A.S.Barabash[1], S.G.Belogurov[1], G.Carugno[2], S.I.Konovalov[1], F.Massera[4], G.Puglierin[2], R.R.Saakyan[1,3], V.N.Stekhanov[1], V.I.Umatov[1]

[1] Institute of Theoretical and Experimental Physics, B.Cheremushkinskaya 25, 117259 Moscow, Russia

[2] Dipartimento di Fisica e INFN, Universita di Padova, via Marzolo 8, I-35131 Padova, Italy

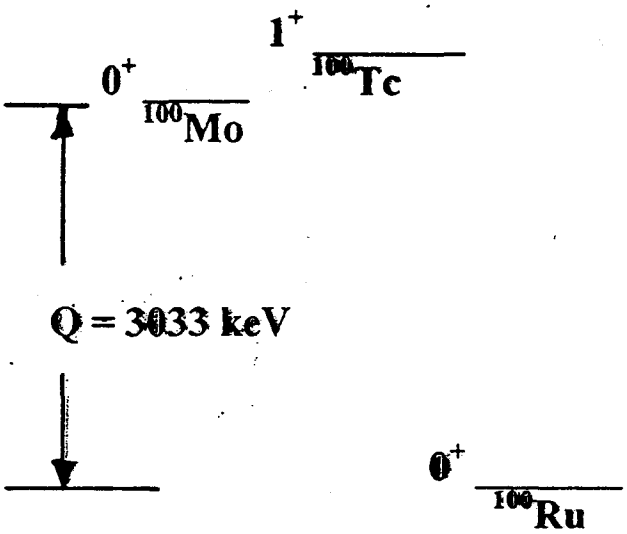
[3] Laboratori Nazionali del Gran Sasso dell'INFN, I-67010 Assergi (L'Aquila), Italy

[4] INFN, Sezione di Bologna, 40126, via Berti Pichat 6/2Ÿ, Bologna, Italy

(INTRODUCTION)

WHY ^{100}Mo? ⇒

1. High Q value – 3033 keV.
2. Large values of $M^{2\nu}$ (experiment) and $M^{0\nu}$ (calculations).
3. Interesting nucleus:
   1) to check low-lying-state dominance hypothesis (half-life, single electron spectrum, angular distribution);
   2) to check possible time variation of G_F.

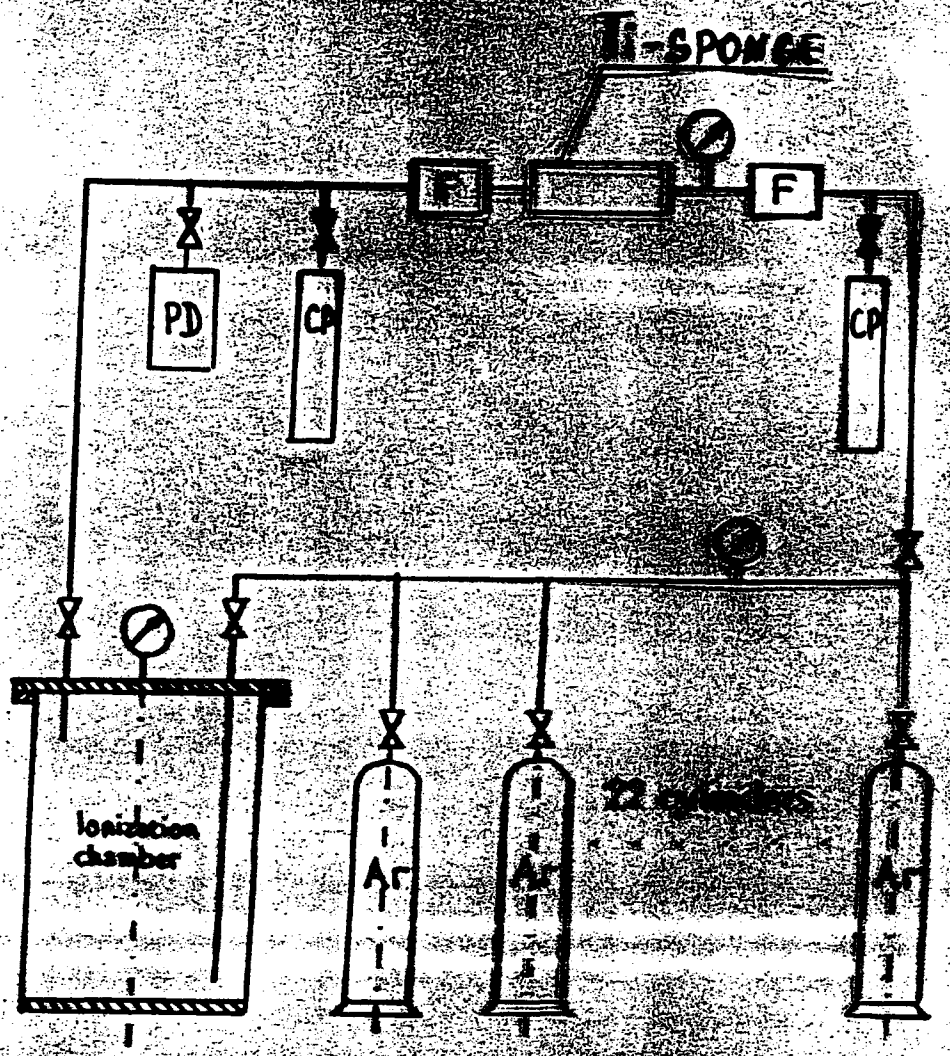


MAIN GOALS OF THE EXPERIMENT ⇒

1. To measure (independently and using new technique) half-life value of $2\beta(2\nu)$-decay of ^{100}Mo.
2. To improve limits on 0ν- and $0\nu\chi^0$-decay.
3. To investigate background properties of liquid argon (^{42}Ar, ...).

222Rn

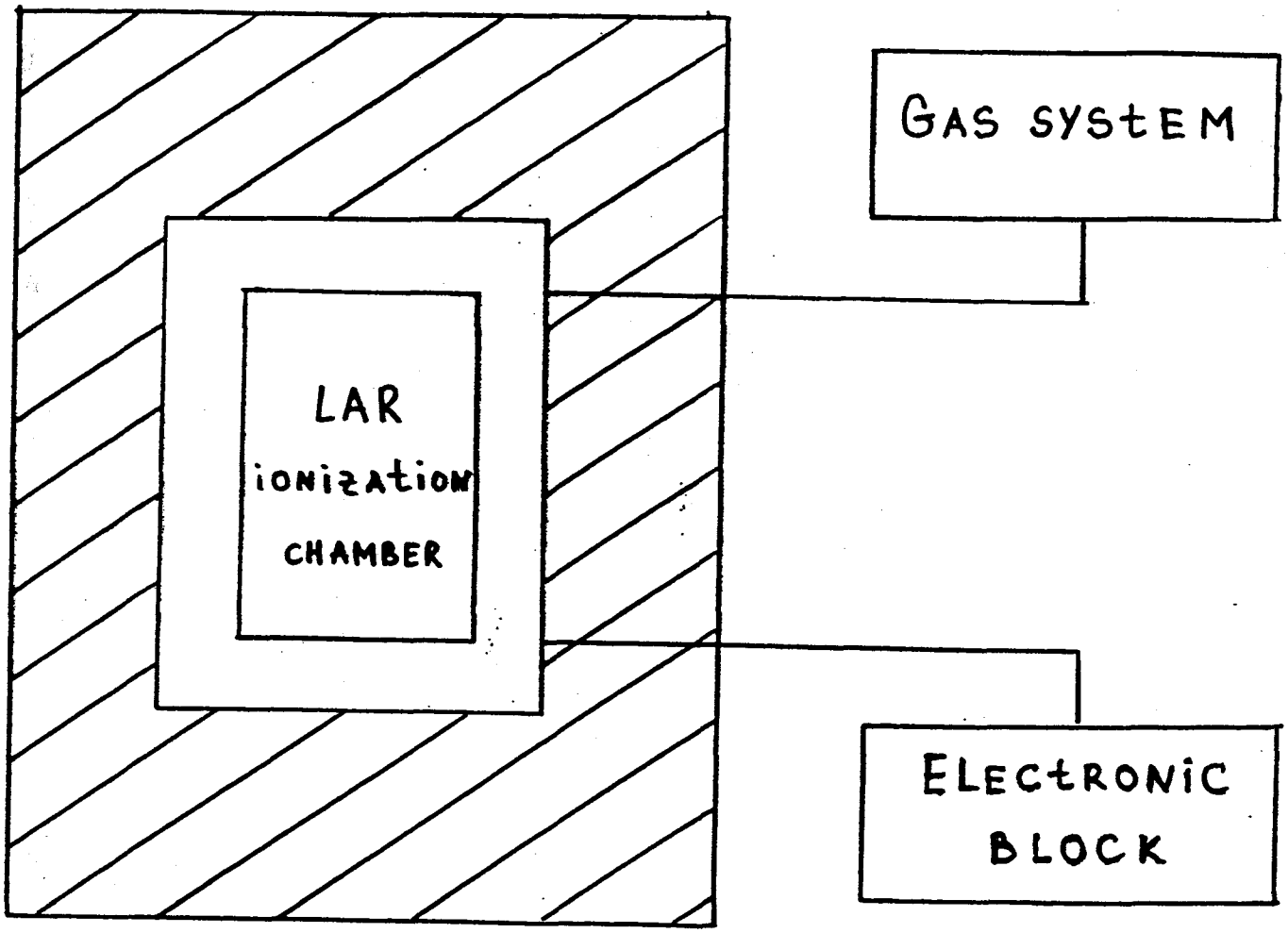
GAS SYSTEM



THE Ti-SPONGE PURIFICATION SYSTEM HAS BEEN CHOOSEN SINCE, FOR EXAMPLE, OXYSORB WILL GIVE A LOT OF ^{222}Rn. FROM THIS POINT OF VIEW Ti-SPONGE IS $10^2 - 10^3$ TIMES BETTER.

OUR PURIFICATION SYSTEM REDUCES THE IMPURITY CONTENT $\rightarrow 2\ 10^{-9}$ eq. O_2 $\rightarrow$ IT'S ENOUGH FOR US

SCHEME OF EXPERIMENTAL SETUP

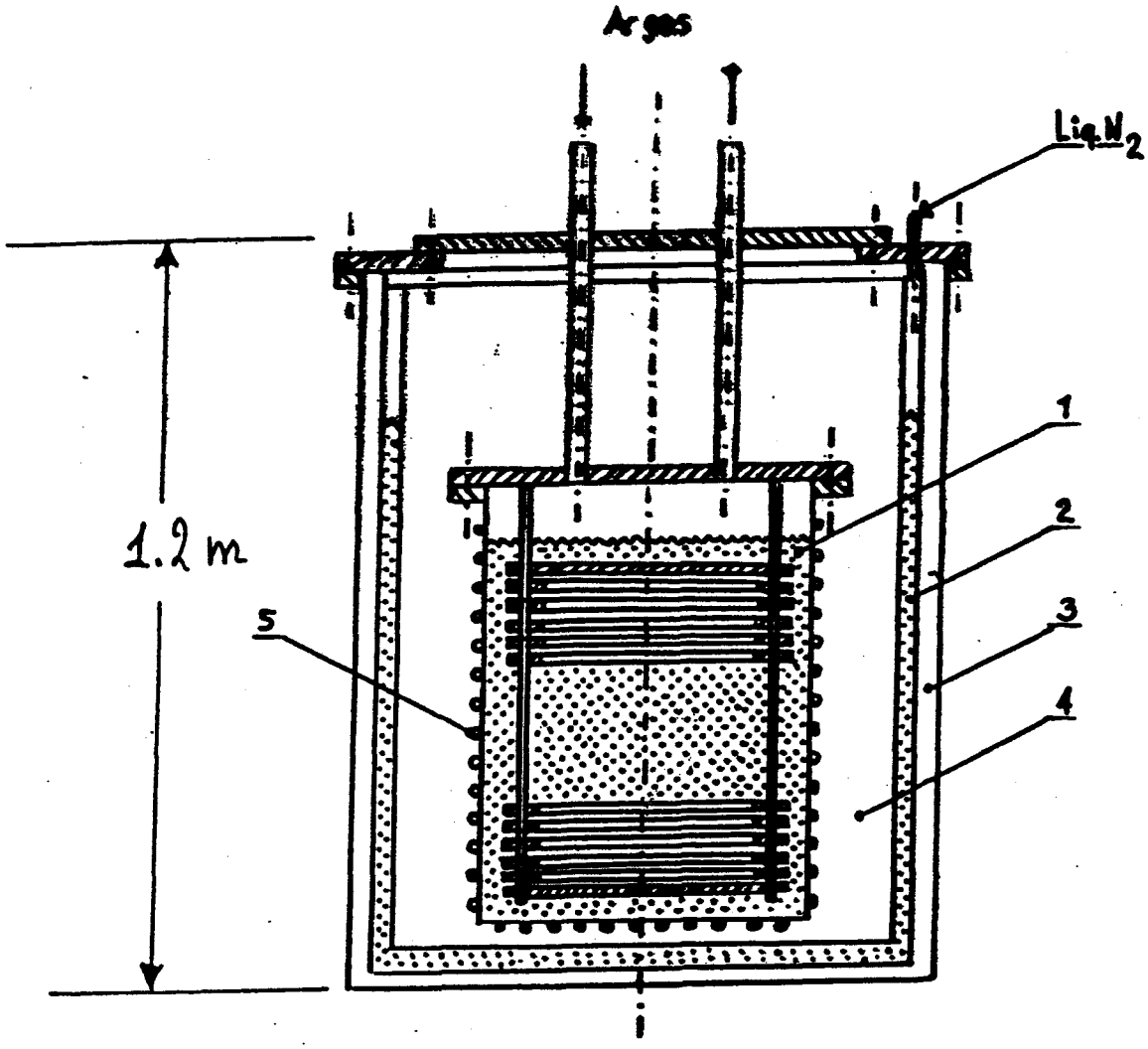


Pb ~ 15 cm ~ 28 t

SETUP IS PLACED IN LNGS (3500 m.w

Level of ^{222}Rn in the air is ~ 180 Bq/M^3
(and $\lesssim$ 9 Bq/M^3 inside of passive shielding)

IONIZATION CHAMBER



1 - VESSEL WITH INNER VOLUME $\varnothing 40 \times 70\ cm^3$

2 - "LIQUID NITROGEN" VOLUME

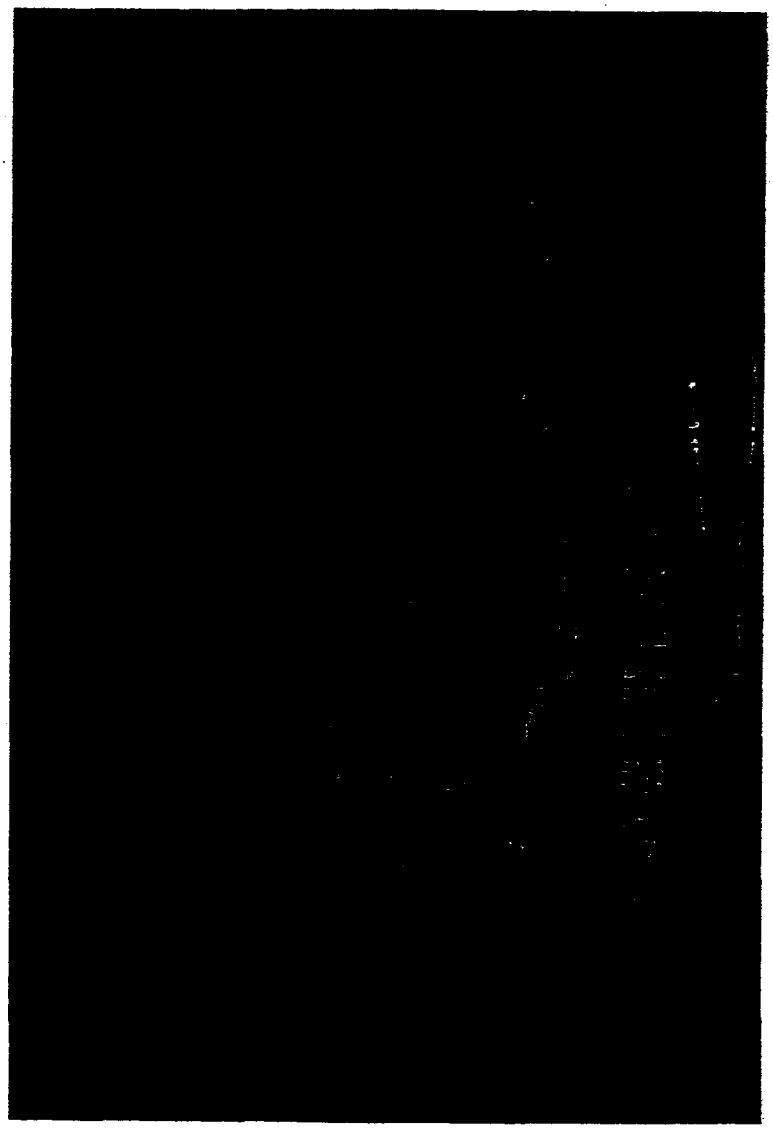
3 - VACUUM CASING

4 - "VAPOUR NITROGEN" VOLUME

5 - HEATERS

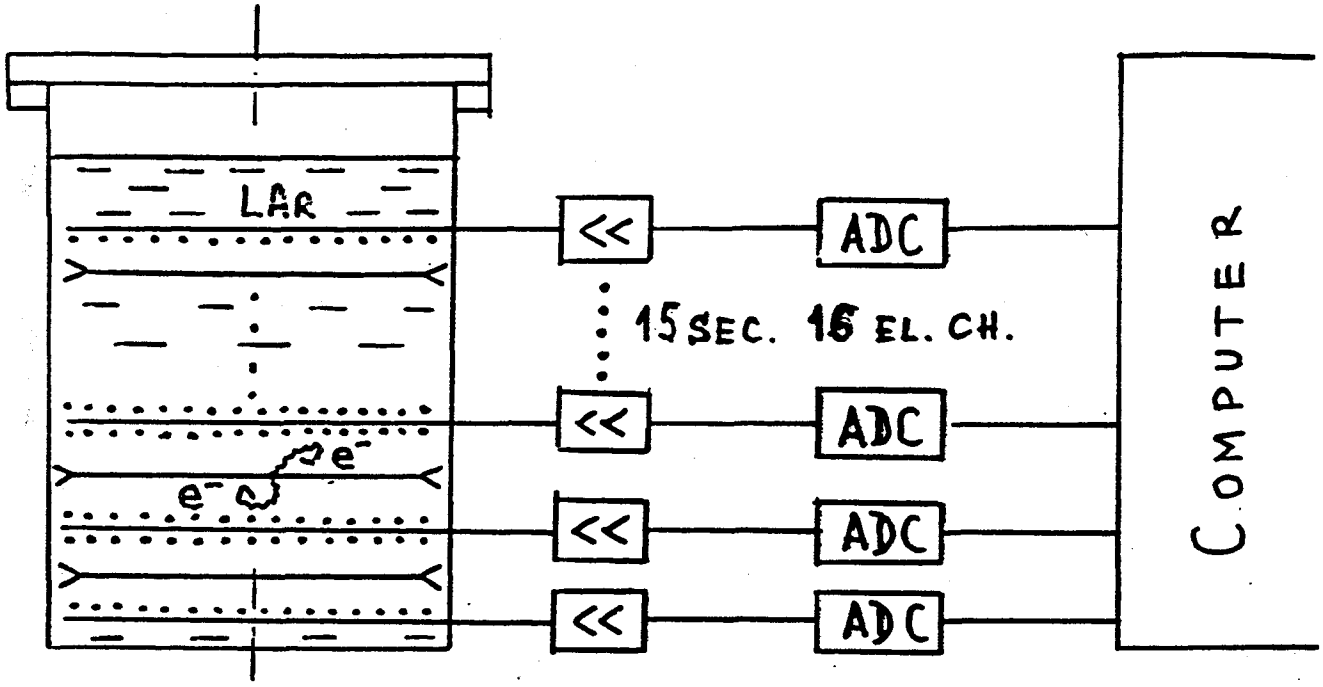
THE CHAMBER IS FILLED UP WITH AR.

EVAPORATION TAKES ABOUT 3 DAYS.

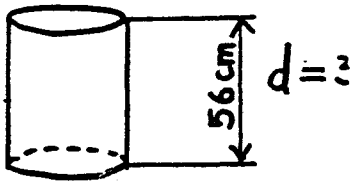


[redacted]

PRINCIPLE OF THE EXPERIMENT



1. USEFUL VOLUME is 40 L OF LAR



2. EACH SECTION ⟹ two PAIRED FLAT IONIZATION CHAMBER WITH SCREENING GRIDS AND COMMON CATHODES. THE CATHODE is Mo-FOIL ~ 50 μm.

3. NOW THE CHAMBER CONTAINS 8 ENRICHED CATHODE ⟹ ~306 g OF ^{100}Mo. ENRICHMENT is 98.37 %
(1-st measurements were done with 4 cathodes – 138.7 g of 10

4. TWO TRIGER CONFIGURATION ARE AVAILABLE:
   a) THE SIGNAL EXCEEDS THE THRESHOULD AT LEAST ON THE ONE OF ANODS

EXPERIMENTAL CONDITIONS OF THE MEASUREMENTS

1. The chamber contains 8 cathodes of enriched Mo ⇒ **306 g of ^{100}Mo.** These cathodes were put into the working volume alternatively with the 6 cathodes of natural Mo.

2. Electrical fields:

E_1 = 1.93 kV/cm for the cathode-grid gap

E_2 = 4 kV/cm for the anode-grid gap

3. Energy resolution:

$\Delta E/E$ = 6% FWHM (for E = 3 MeV)

4. Trigger – at least on one of anodes the signal exceeds a threshold

Threshold – 500 keV

5. Measurement time:

Run 1 - 202 h;

Run 2 - 238 h;

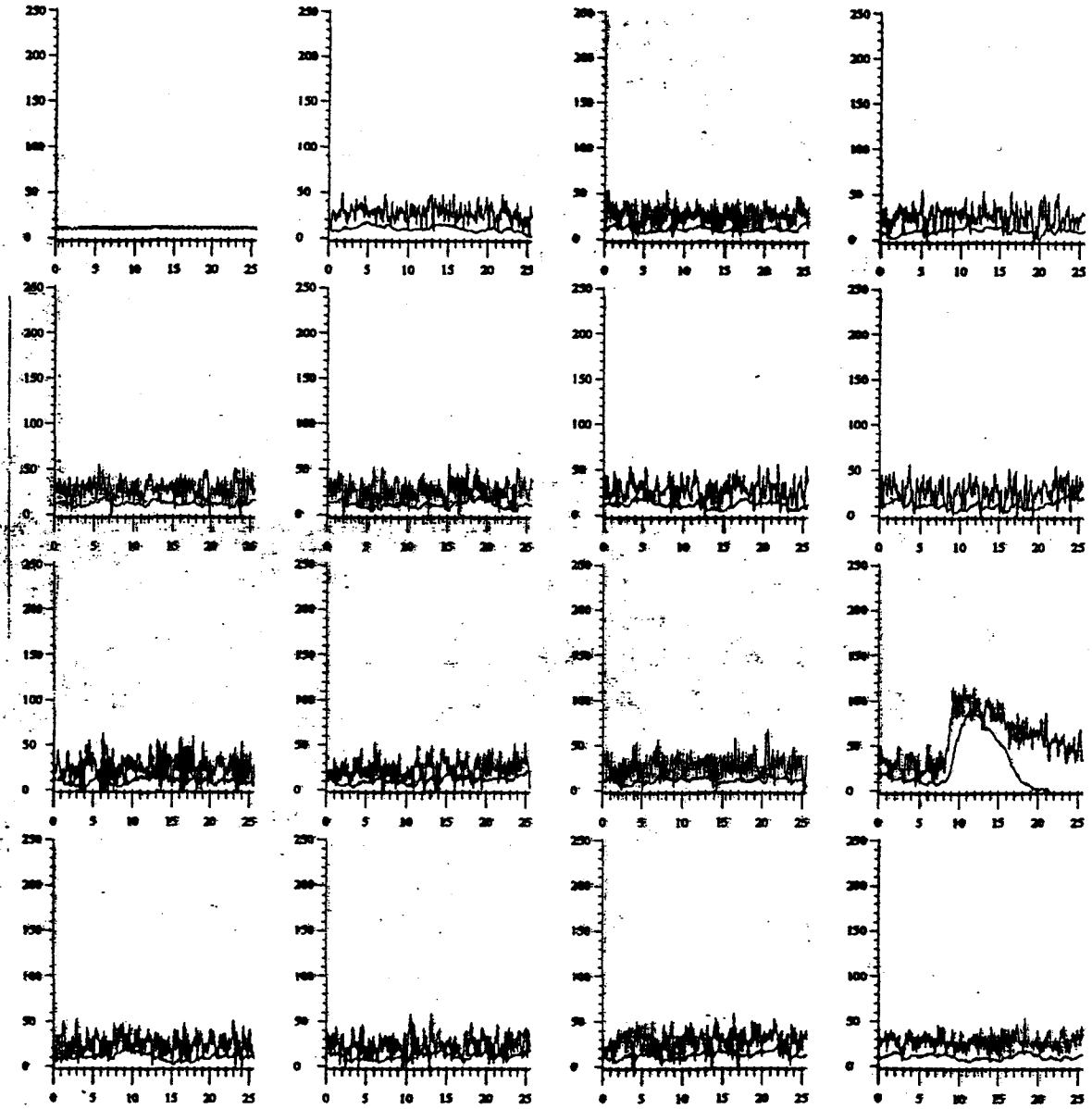
Run 3 – 313 h
Run 4 – 1355 h
} M·t = 458 kg·h for ^{100}Mo and 303 kg·h for ^{nat}Mo

Run 5 - 708 h (anti-neutron shielding)

6. ^{222}Rn concentration:

Run 1 - ~ 50 Bq/m^3;

Run 2, 3, 4, 5 - < 2 Bq/m^3



N

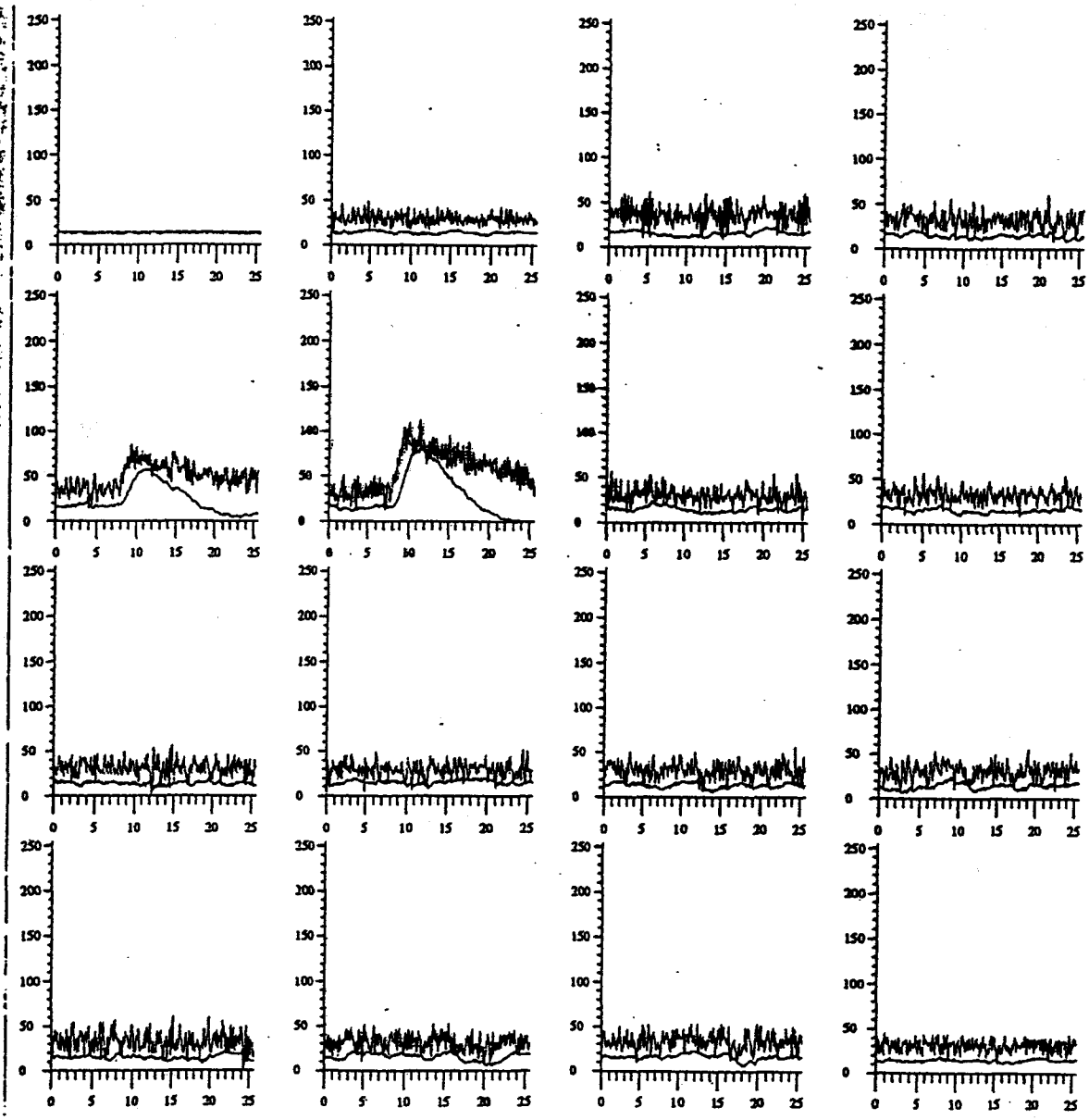
PREVIOUS

FIRST

EXIT

00012

Single electron event



NEXT

PREVIOU

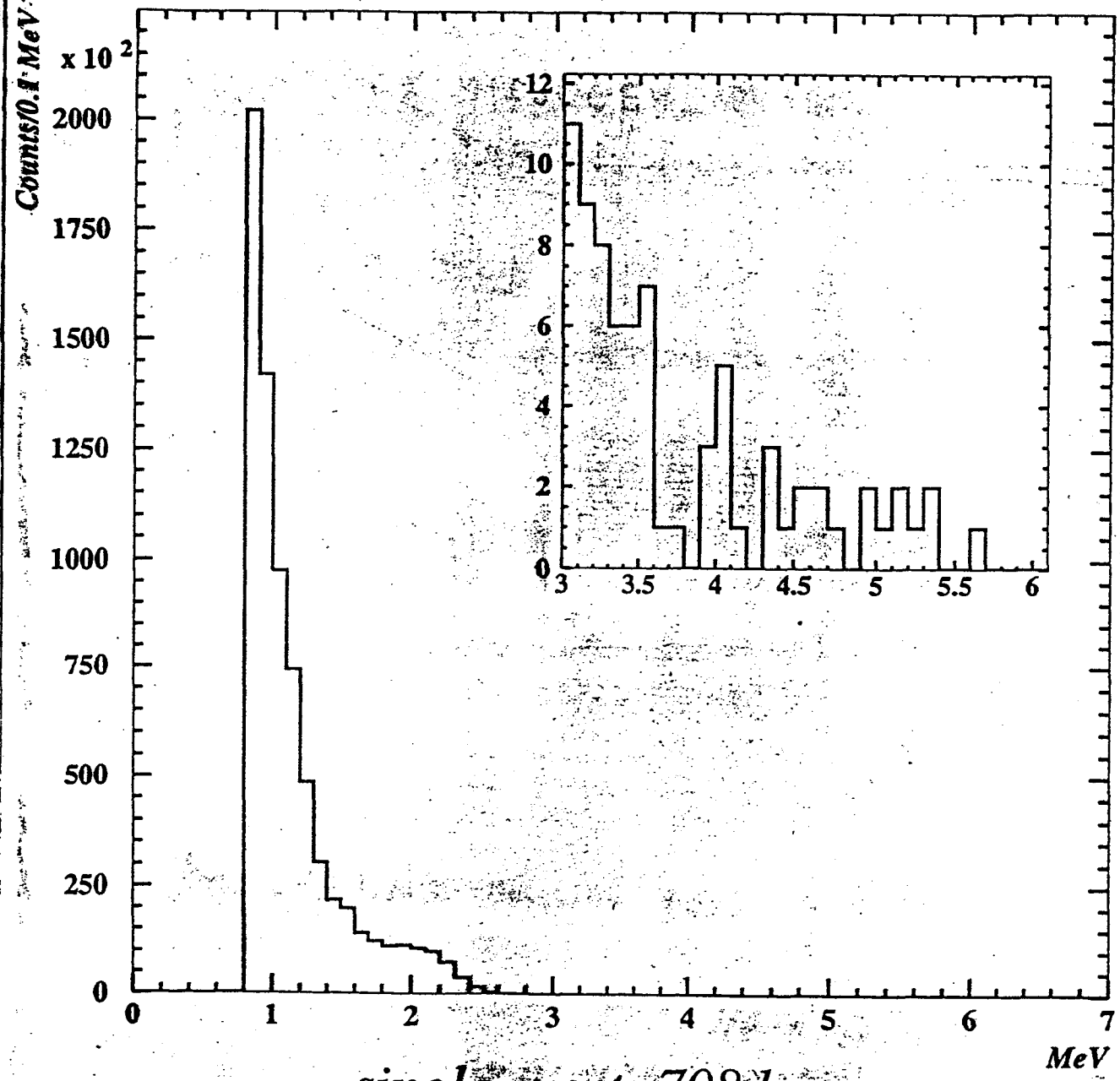
FIRST

EXIT

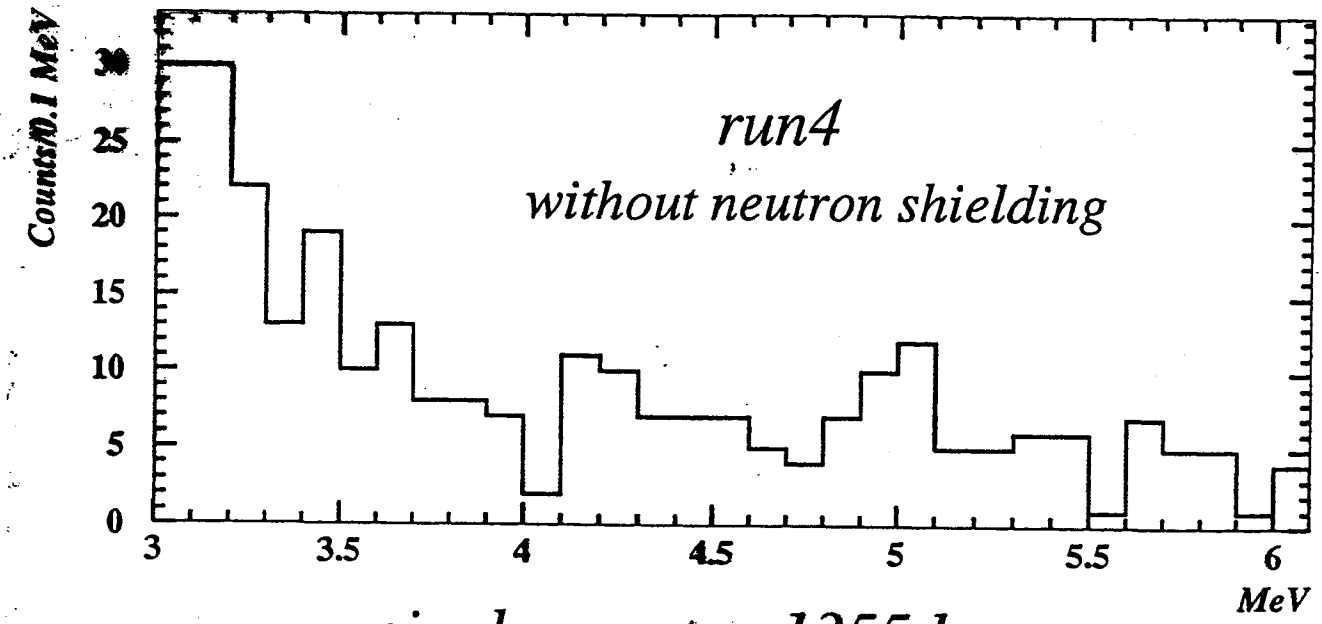
02542

Double electron event

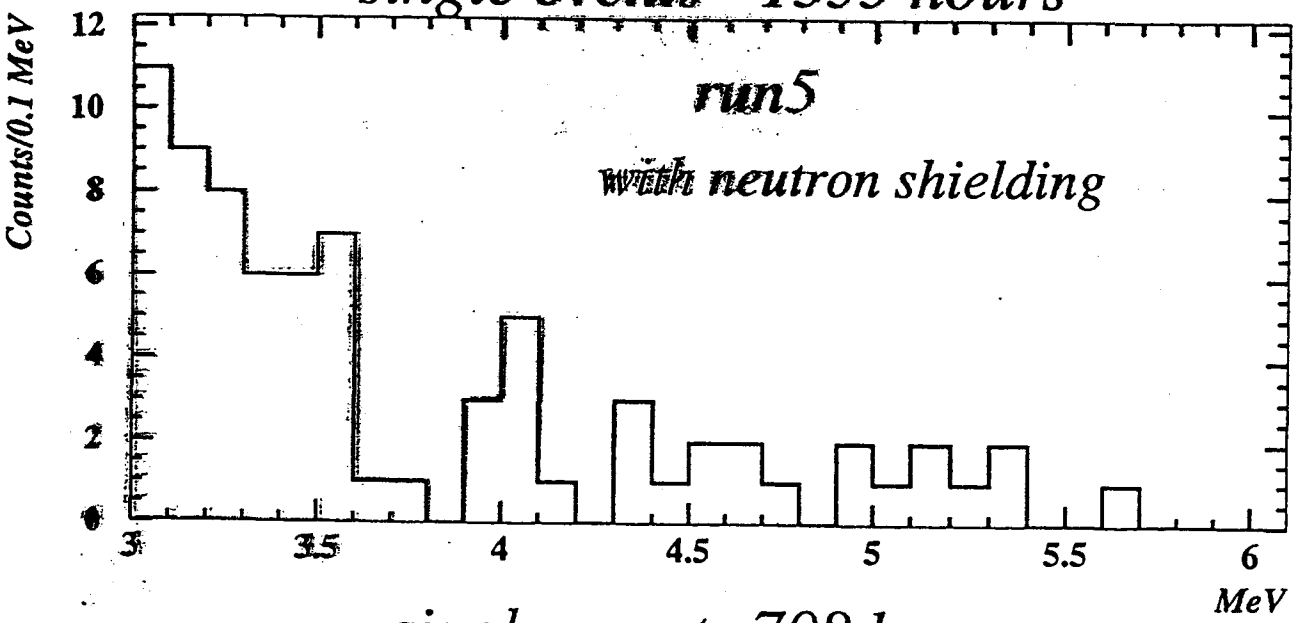
Run № 5



single events 708 hours



single events - 1355 hours



single events 708 hours

$$^{222}Rn \rightarrow \ldots\ldots \rightarrow {}^{214}Bi \xrightarrow{\beta} {}^{214}Po \rightarrow \ldots$$

$E_\beta = 3.26$ MeV

Energy interval [3 -3.5] MeV - 34 events/708 h

$\varepsilon = 0.025\%$

mass of Ar - 52 kg

Activity of Rn-222 in liquid Ar is < 1 mBq/kg

^{42}Ar : $C < 4 \cdot 10^{-21}$ g/g (90% CL.

Alpha background from radon decay in the UK dark matter search experiments

V. A. Kudryavtsev, N. J. C. Spooner,
P. K. Lightfoot, J. W. Roberts

Department of Physics and Astronomy
University of Sheffield

Contents

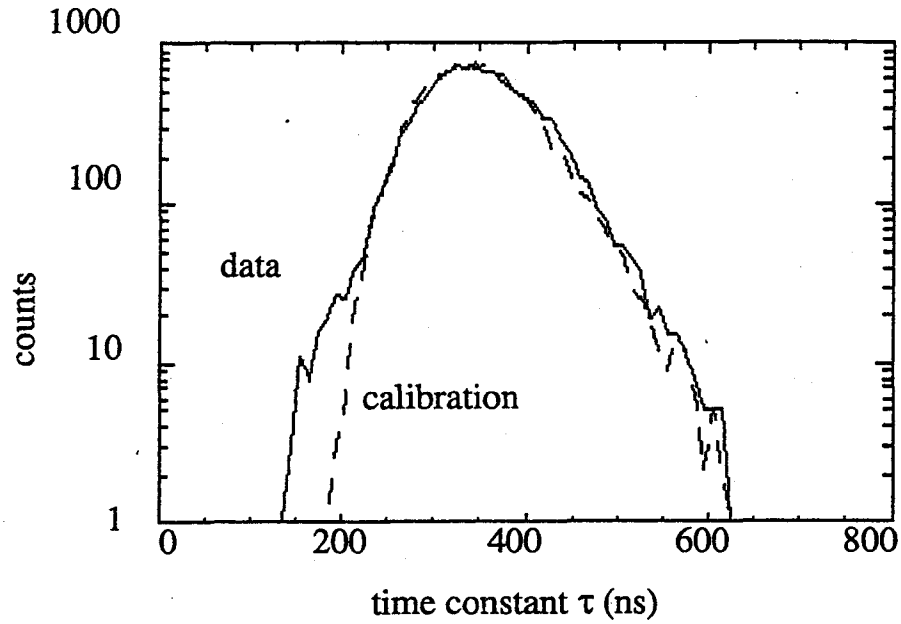
1. UK Dark Matter search experiments. Introduction.
2. Anomalous fast component in NaI(Tl)/CsI(Tl) detectors.
3. Alpha background as a possible explanation of anomalous component. External alphas versus internal (surface) alphas.
4. Recent test with CsI(Tl) crystal
   - Spectrum of fast component
   - Decrease of intensity after polishing
   - Rn versus U/Th contamination.

   Special features of alpha background
5. Conclusions

UK Dark Matter search experiments

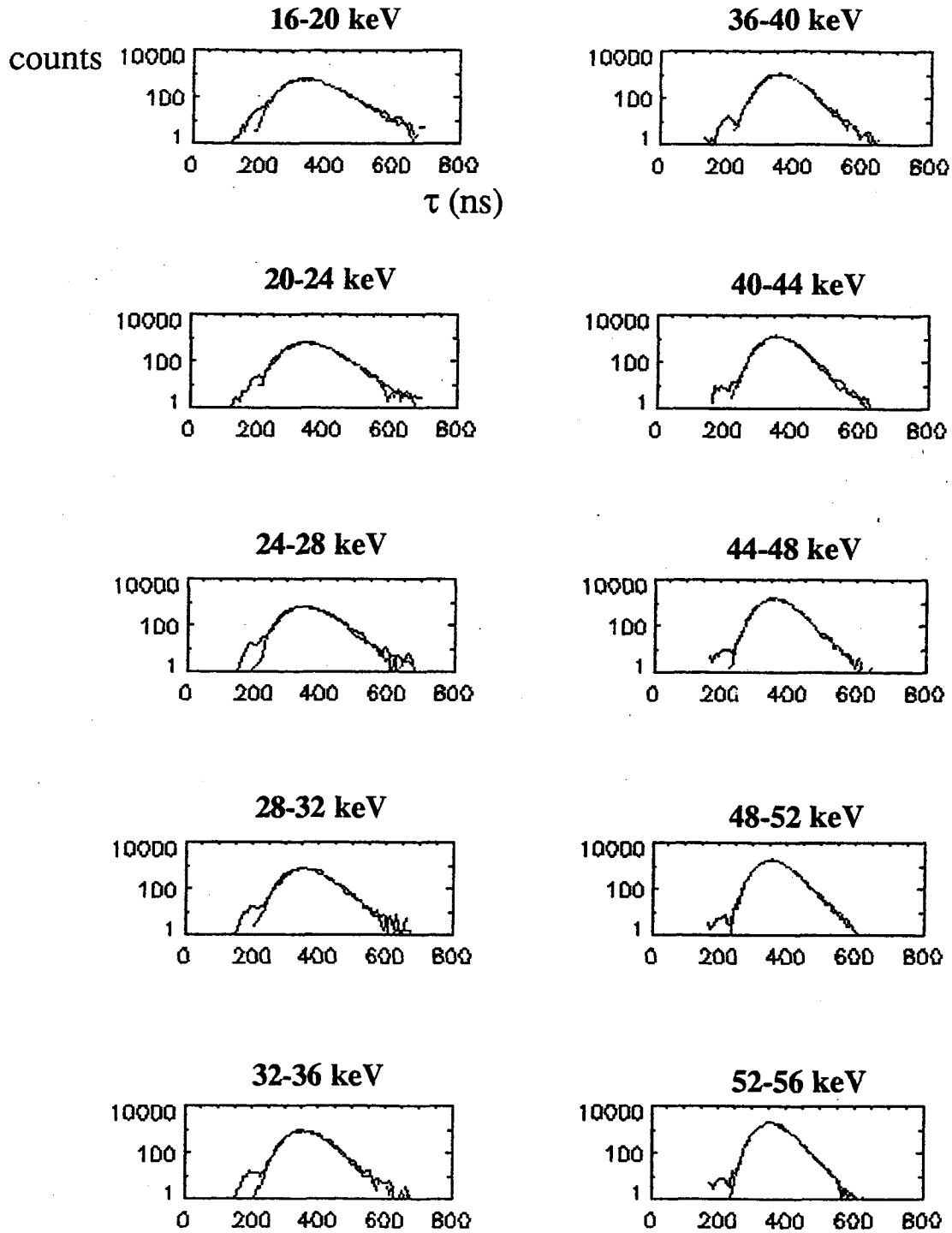
- At present several NaI(Tl) detectors are running in the underground laboratory in the Boulby mine.
- Time profiles of integrated pulses from the PMTs are digitised and fitted to the exponential to derive pulse parameters, such as amplitude (energy) and time constant.
- Time constant distributions of observed events reveal the presence of anomalous fast population of events. The mean time constant of this population is smaller than gamma/electron time constant and even smaller than nuclear recoil time constant.

Time constant distributions from NaI(Tl) crystal

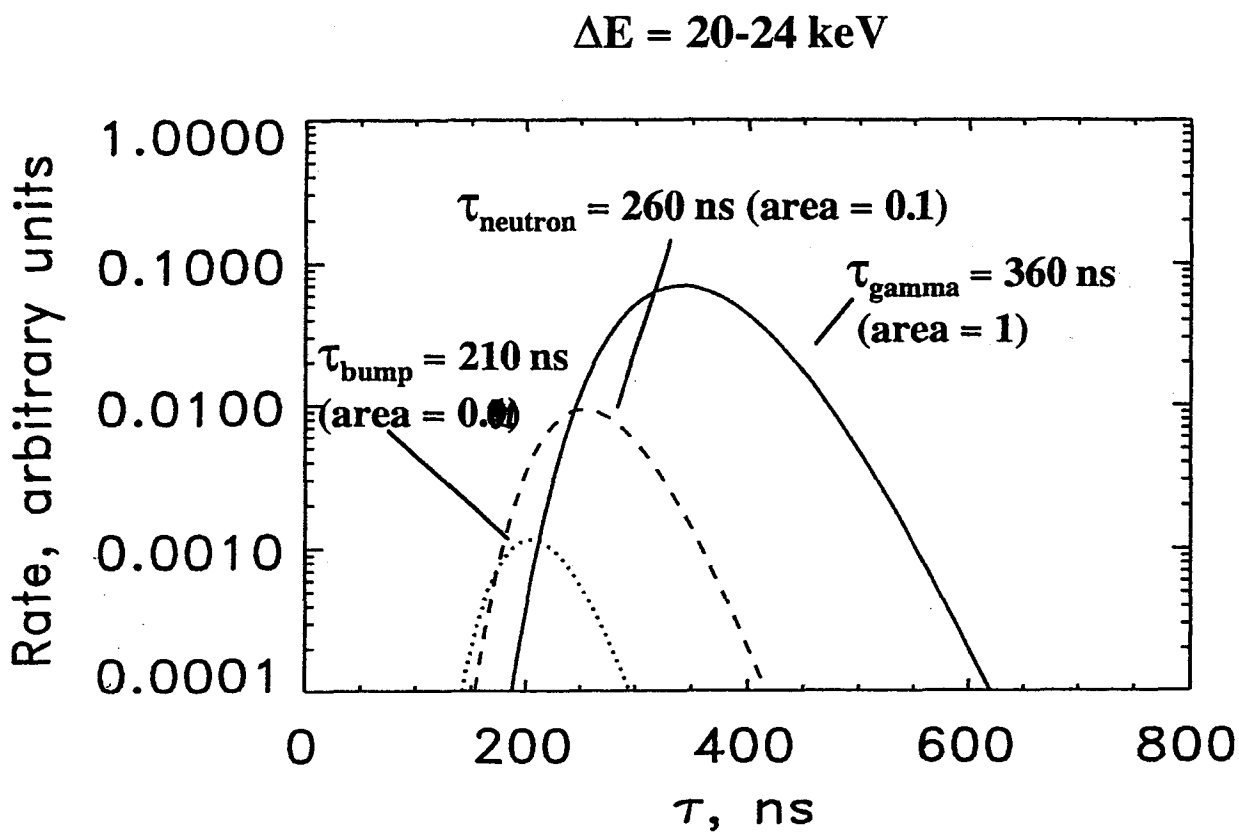


Solid line - data
Dashed line - calibration with ^{60}Co source
'Bump' at low τ is caused by anomalous events - 'bump' events (alphas?)

DM 46 data - 725 kg.days
data plus Compton calibration

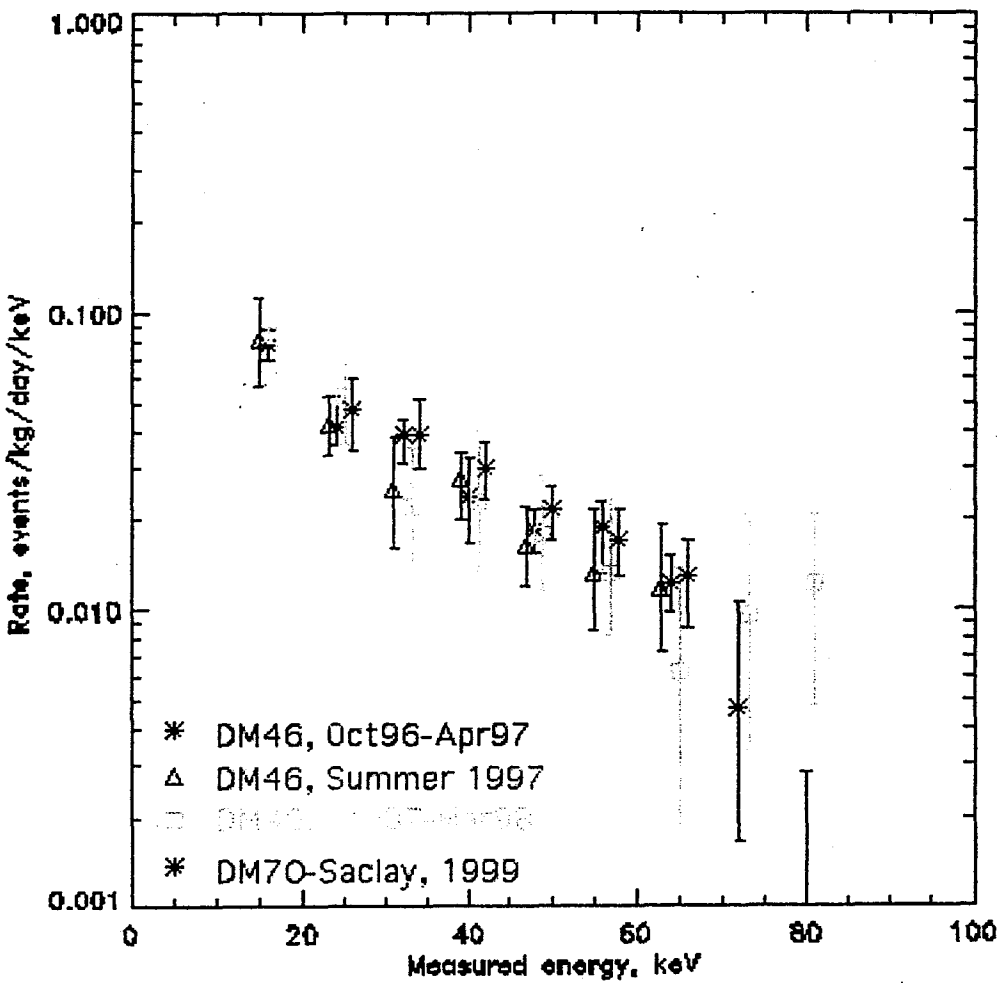


Example DM46 recoil calibration

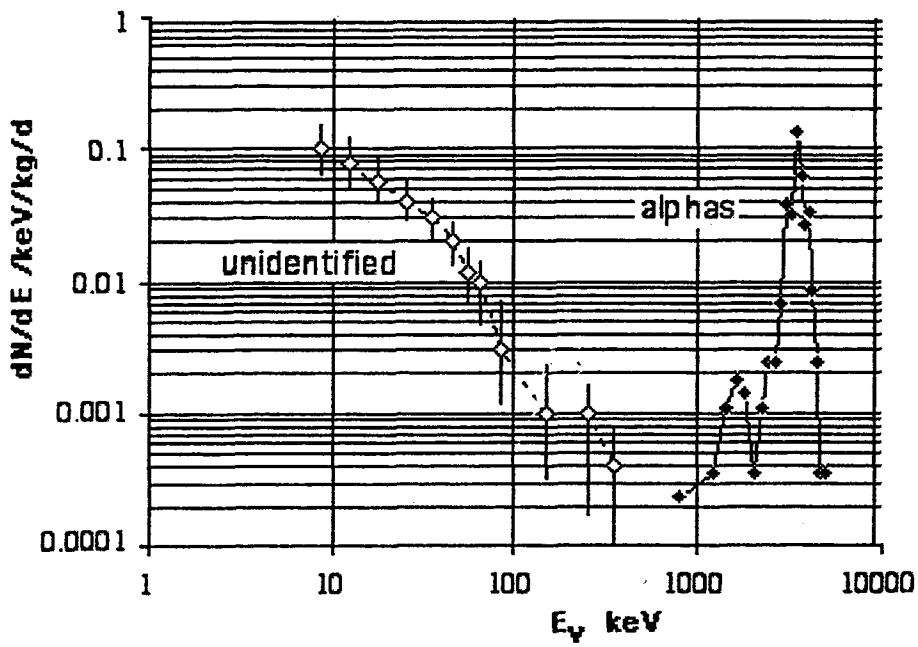


curves are fits to the time constant histograms
fit errors typically +/- 2 ns

Spectra of anomalous ('fast') events in various NaI(Tl) crystals



DM26 (NaI) spectrum: fast component (low energies) + alphas



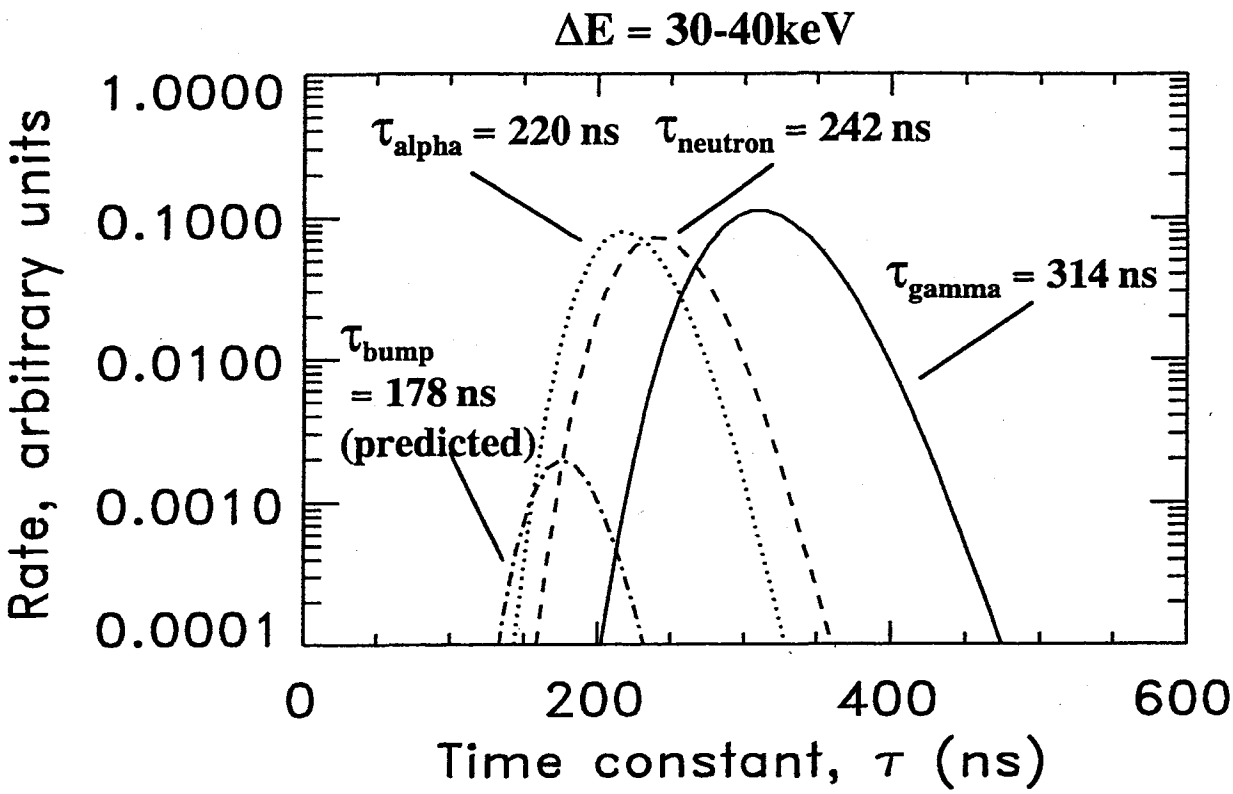
Spectrum of anomalous fast events in DM26 2kg crystal from data covering several energy spans. The MeV range peaks correspond to the expected alpha spectrum from U/Th
Graph from P. F. Smith et al. Phys. Rep., **307** (1998) 275

External versus internal (surface) contamination

- Alpha contamination looks like the best explanation (P. F. Smith et al. Phys. Rep., **307** (1998) 275)
- But internal bulk U, Th level is too low to account for the high rate at low energies.
- External incoming alphas from U, Th outside the crystal (PTFE?) needs ~1 ppm and very fine tuning of dead layers and NaI efficiency. Moreover, time constant of external alphas is not matched well to that of fast component.
- Internal (surface) contamination of crystal by alpha emitting isotope(s) was discussed recently by
- N. J. T. Smith et al. (to be published in Phys. Lett. B). Nuclear recoils from Rn decay can be implanted into the crystal surface. This creates a thin (0.1-0.2 microns) alpha emitting layer.
- Alternatively, crystal surface can be contaminated by U/Th.
- In both cases high concentration of alpha emitting isotope is needed (0.1-1 ppm).
- Test of surface alpha hypothesis - polishing crystal surface and run experiment again. This is impossible for encapsulated NaI crystals. Better discrimination in CsI crystals despite higher background offers such opportunity.

NaI surface alpha test result (DM29 detector)

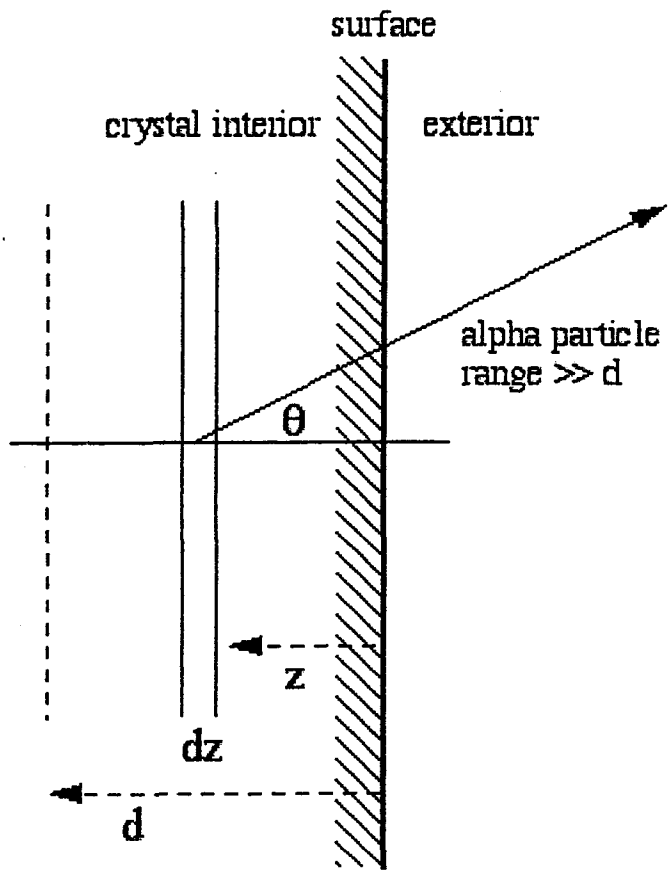
Note alpha events are low energy surface alphas from a source. There is no evidence that a sufficiently high rate of such alphas exists in the UKDMC experiment.



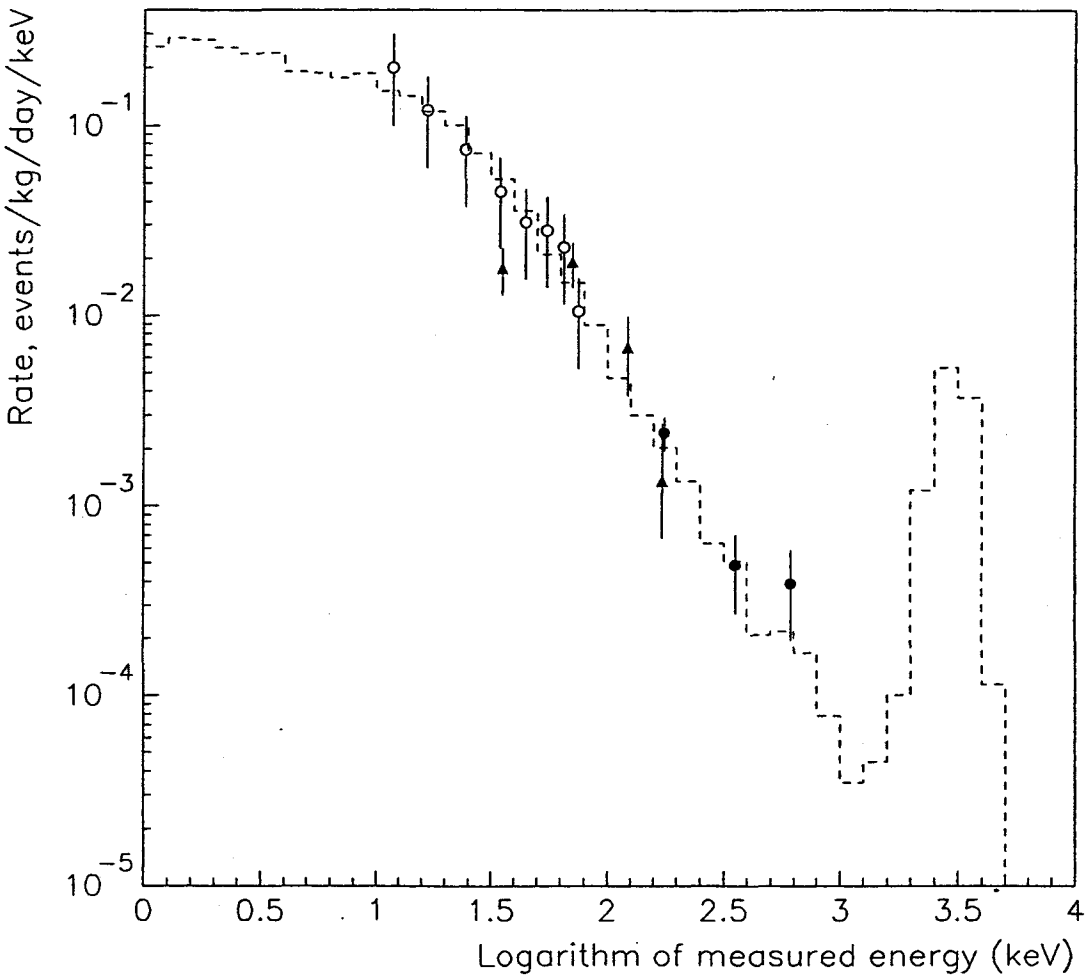
curves are fits to the time constant histograms
errors typically 2-5 ns
temperatures kept stable to ~0.1C

(results in: V. A. Kudryavtsev et al. PLB **452** (1999) 167)

Internal (surface) alphas

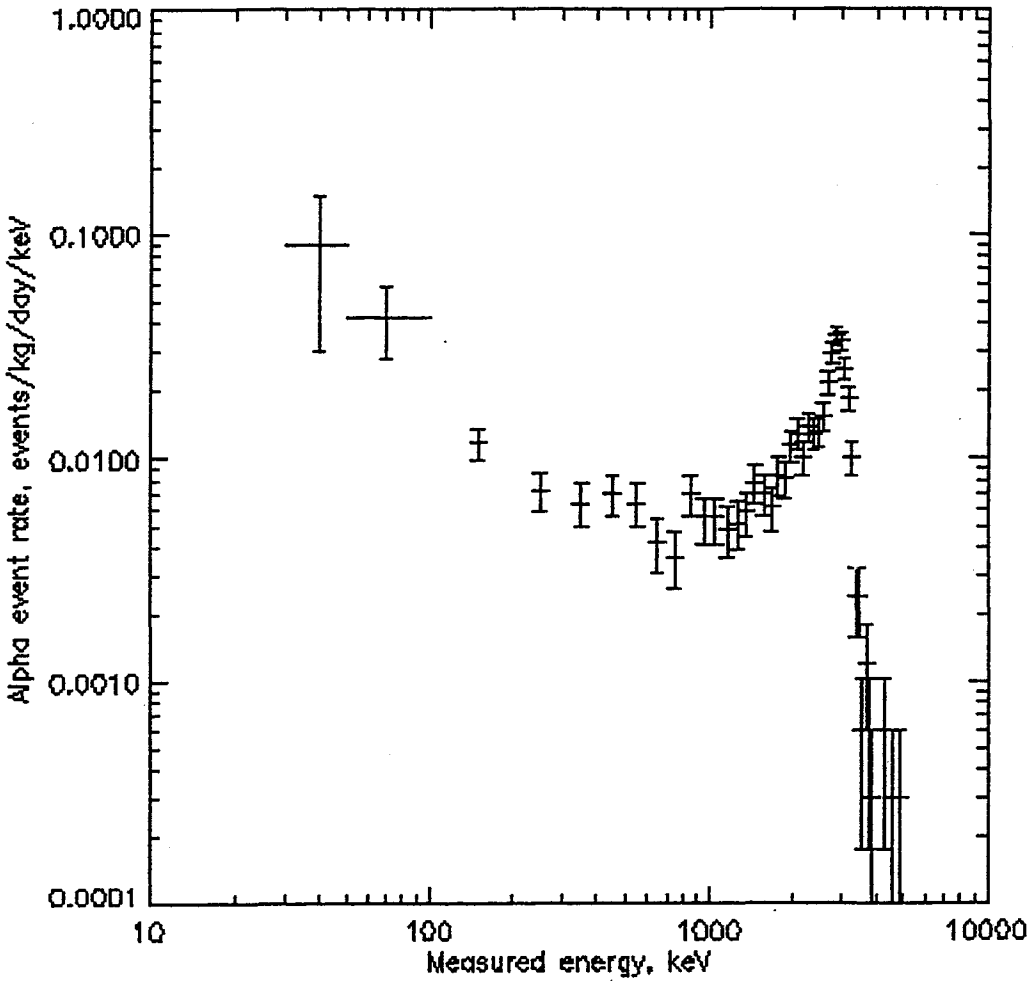


Graph from N. J. T. Smith et al. (to be published in Phys. Lett. B).

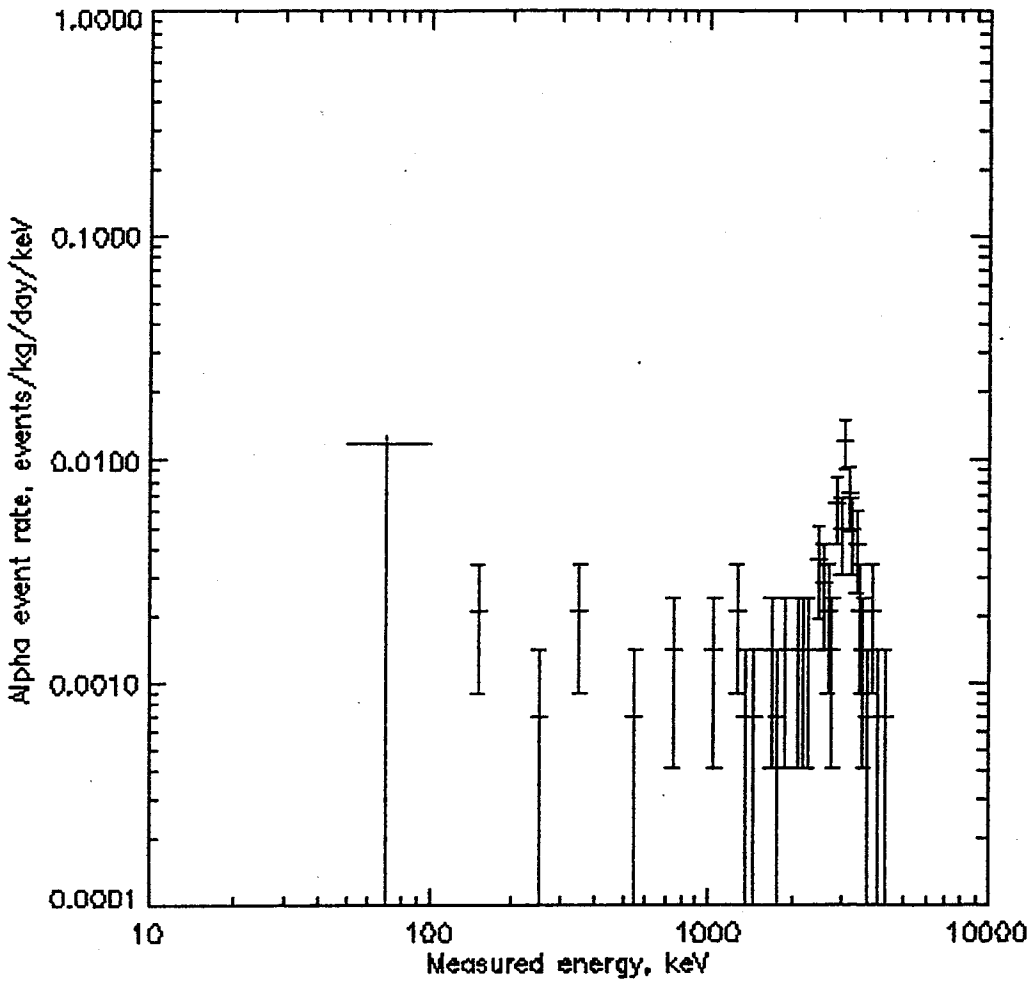


Measured spectrum of the bump and energy loss of alphas

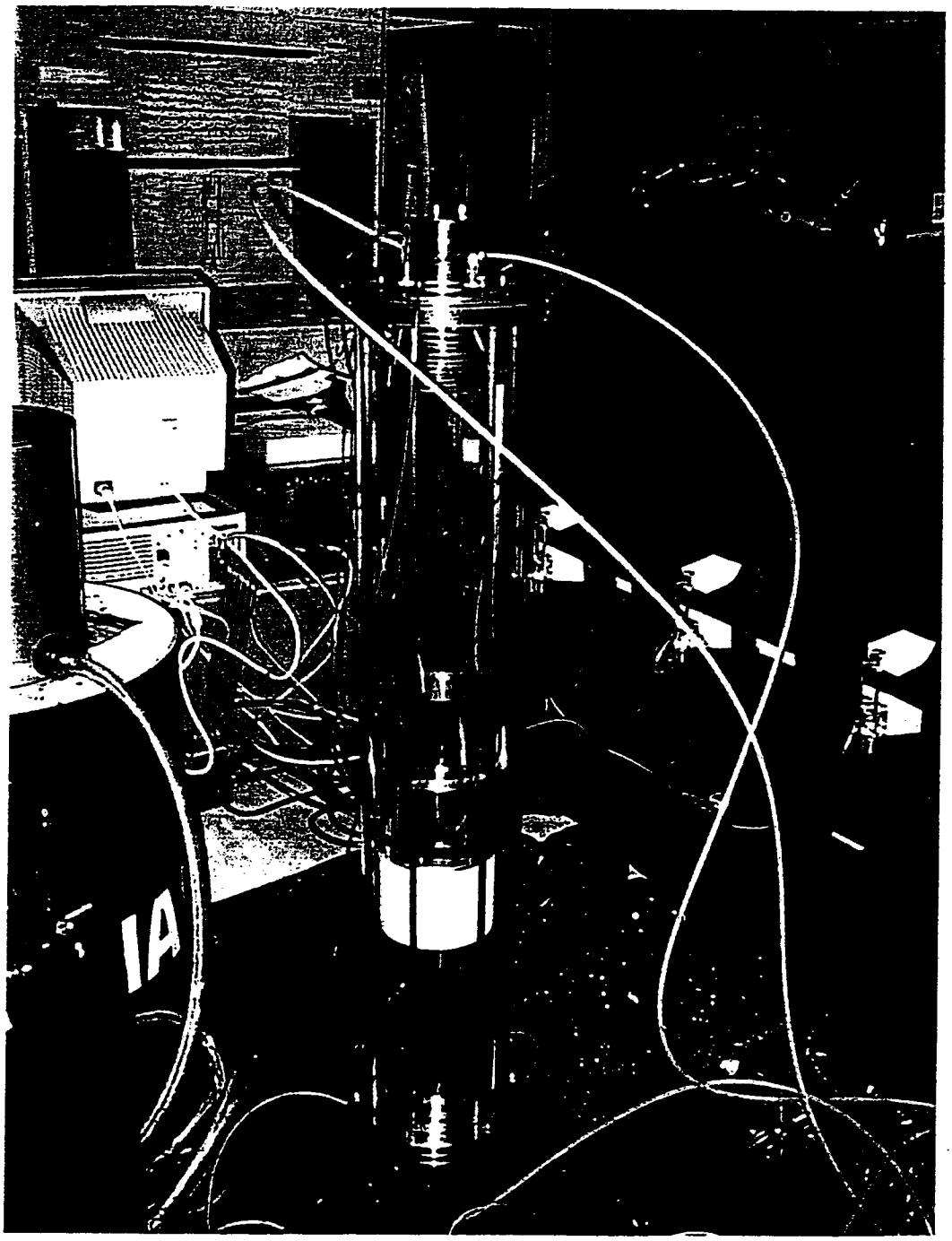
Alpha spectrum from CsI(Tl)



Alpha spectrum from CsI(Tl) after polishing

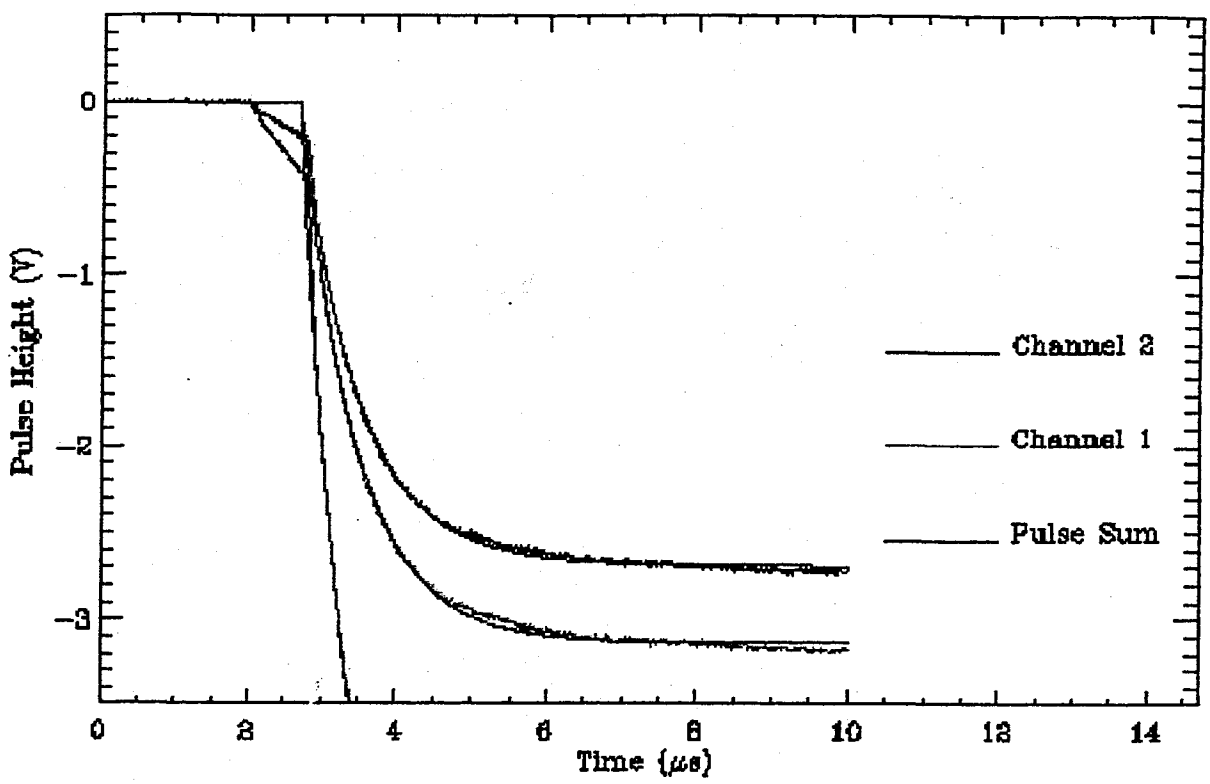


Non-encapsulated vertical detector



High energy pulses from CsI(Tl) crystal

$^{220}Rn \rightarrow ... \rightarrow {}^{212}Bi \rightarrow {}^{212}Po \rightarrow {}^{208}Pb$

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tau =  7.90141e-07   energy =      6889.13   conv =   1   chi^2 =   0.00662526
tau_mean =  3.14521e-06   t0 =  2.68335e-06  amp =      3.13913
tau =  7.75933e-07   energy =      6499.24   conv =   1   chi^2 =   0.00194443
tau_mean =  3.31798e-06   t0 =  2.67719e-06  amp =      2.68853
tau =  8.02663e-07   energy =      6721.32   conv =   1   chi^2 =   0.00146419
```

17 events from this chain, 15 of them - during 1st day

Half-life of ^{212}Bi is 10.6 hours.

Half-life of ^{212}Po is 0.3 microseconds.

These events are caused by ^{222}Rn (or daughter nucleus) contamination of the crystal surface during installation.

Rn contamination versus U/Th contamination

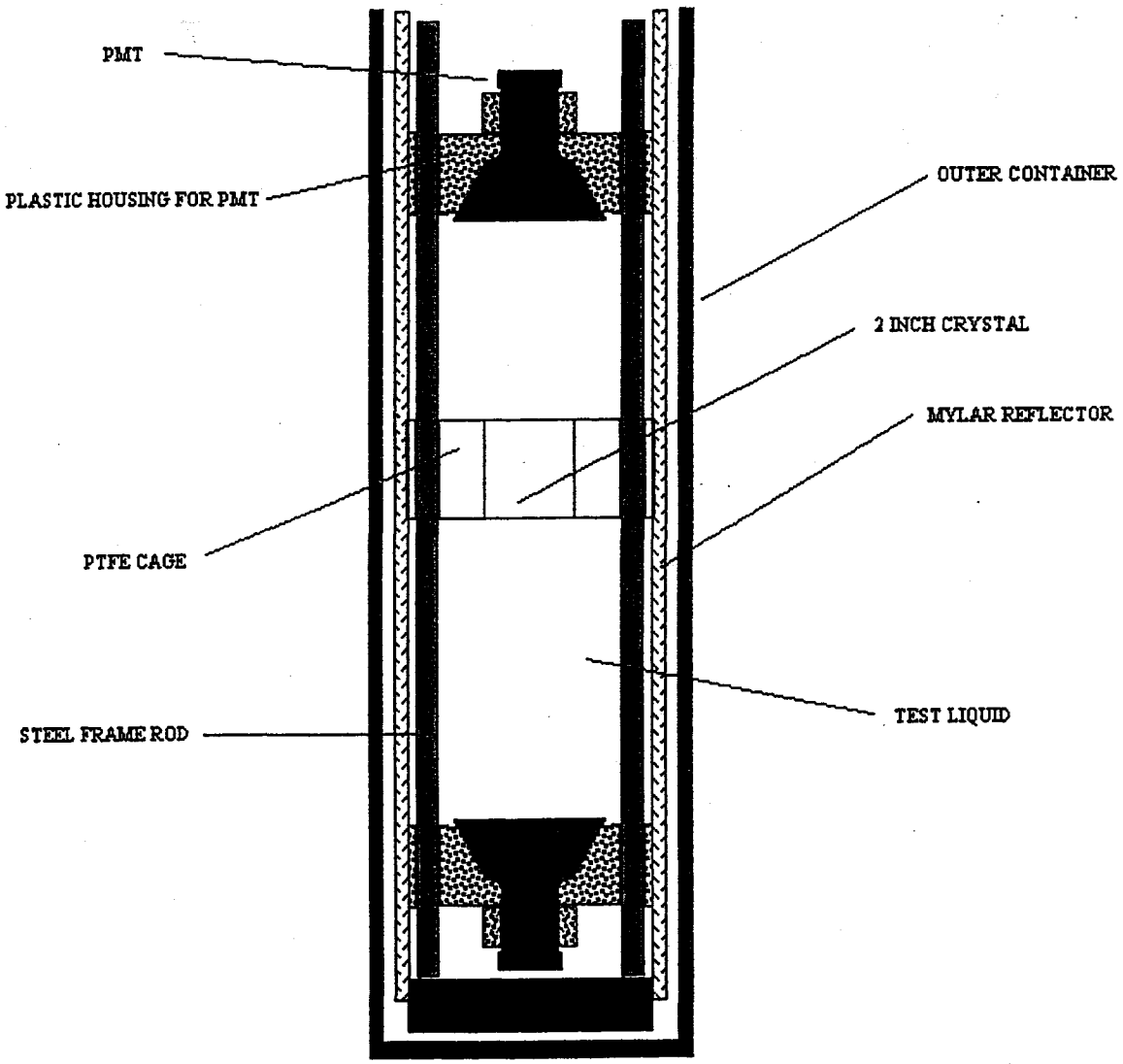
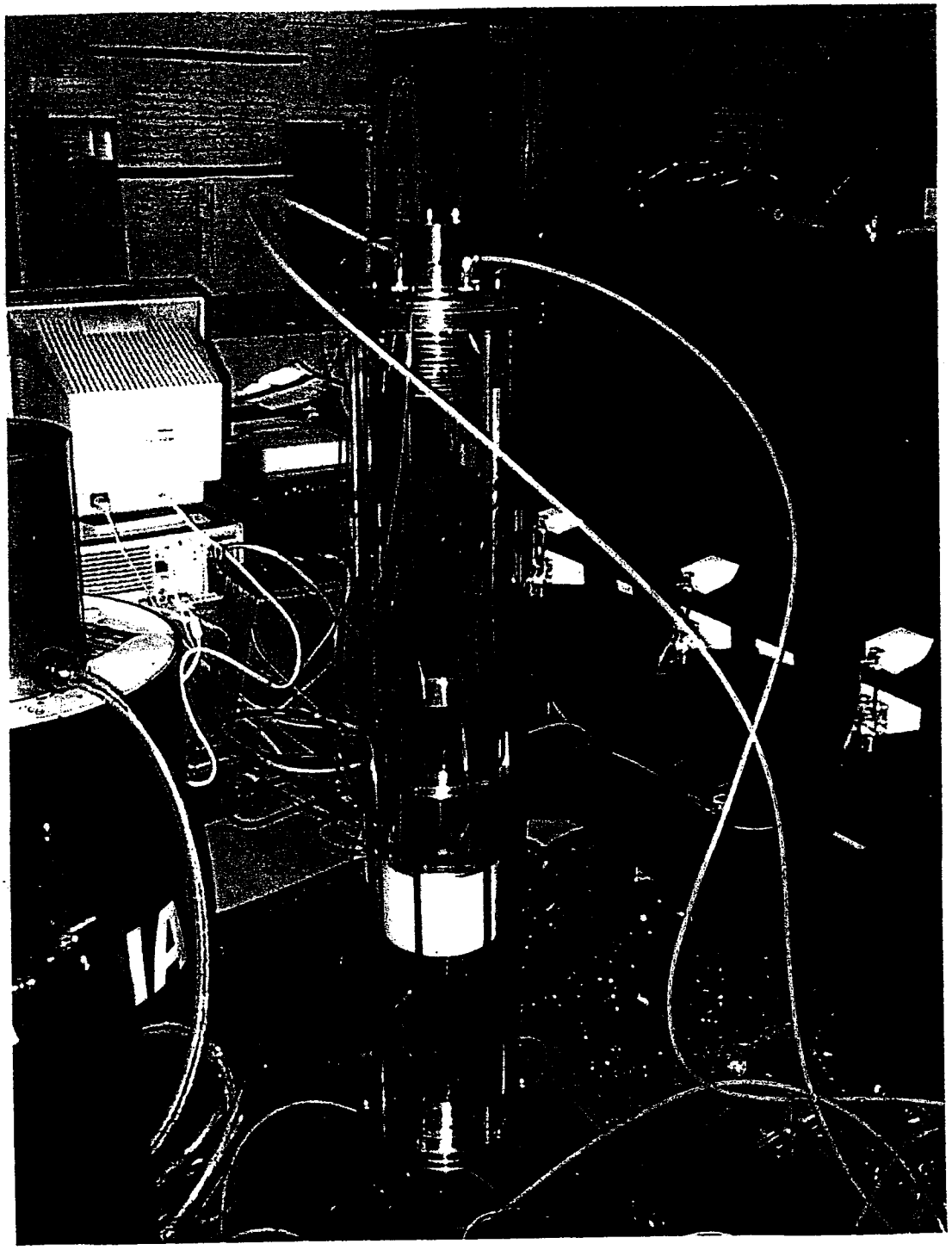
- Only one prominent peak is seen on the spectrum. The peak is probably due to $^{210}Po \rightarrow {}^{206}Pb$ decay (5.3 MeV alphas).
- No decay chains $^{222}Rn \rightarrow {}^{218}Po \rightarrow {}^{214}Pb$
  (2nd pulse delayed by about 3 minutes) or
  $^{224}Ra \rightarrow {}^{220}Rn \rightarrow {}^{216}Po \rightarrow {}^{212}Pb$
  were seen before or after polishing.
  Conclusion:
  The concentration of U and Th in the bulk of the crystal is low (less than 0.5 ppb).

- An exposure to radon of at least 1 year is needed to explain the main alpha peak. This figure comes from the observation of 17 events from $^{212}Bi \rightarrow {}^{212}Po \rightarrow {}^{208}Pb$ decay chain (exposure of crystal to air was about 2 hours during crystal installation). 1 year exposure to radon is not unusual for the old unencapsulated CsI(Tl) crystal. At least several month exposure to radon is needed to explain alpha peak (and low-energy fast events) in larger size encapsulated NaI(Tl) crystals.
  How is it possible?
  Radon diffusion through the windows of the detector?

Conclusions

- Anomalous fast events observed in the UK Dark Matter search experiments can be explained by the contamination of the crystal surface by the products of radon decay. It is unknown, however, how and when radioactive nuclei were implanted into NaI(Tl) crystals.
- Preliminary results show that polishing crystal surface removes at least major part of alphas and possibly of low-energy events.
- UKDMC has started a new program to run unencapsulated NaI(Tl) crystals in a liquid (paraffin) or in nitrogen atmosphere. The crystals can be taken off the detectors and re-polished.

Design of an unencapsulated detector



Non-encapsulated vertical detector