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HEALTH PHYSICS SYSTEM SCHEME FOR THE URANIUM PURIFICATION PLANT*

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ABSTRACT

After describing the two uranium purification processes used in the Chemical Engineering Division of the Instituto de Energia Atomica we examined the possible hazards and methods to control or eliminate them.

Since these purification process present several stages in each one of them we have evaluated the hazards and we have tried to give adequate solutions to protect both personnel and installations from the potential radiation hazards.

1 Introduction

This work has its object a description of the activities that the Health Physics and Dosimetry Service (SPRD) is developping in the Uranium Purification Pilot Plant of the Chemical Engineering Division (DEQ) of the Atomic Energy Institute (IEA)

With the intention to show the range and the importance of the services rendered by us in the uranium operation areas, we will concisely describe the two uranium purification processes developped in that Division

We start from the supposition that all work with radioactive substances can present three types of hazards namely

a) external exposure
b) external contamination
c) internal contamination

We try to evaluate in both of the purification processes the type or types of hazards and ways to control or to eliminate them

Since these purification processes present several stages in each one of them we have done the evaluation of the hazards and we try to give adequate solutions in a manner to protect from the potential radiation hazard both to the human element and to the instalations Up to now, there have already been taken a lot of measures in this sense and we can affirm that the result of our endeavour have been worthwhile even though other measures may be necessary

The present and future test of this scheme that is, giving satisfactory results, will orient us the future measurement of protection

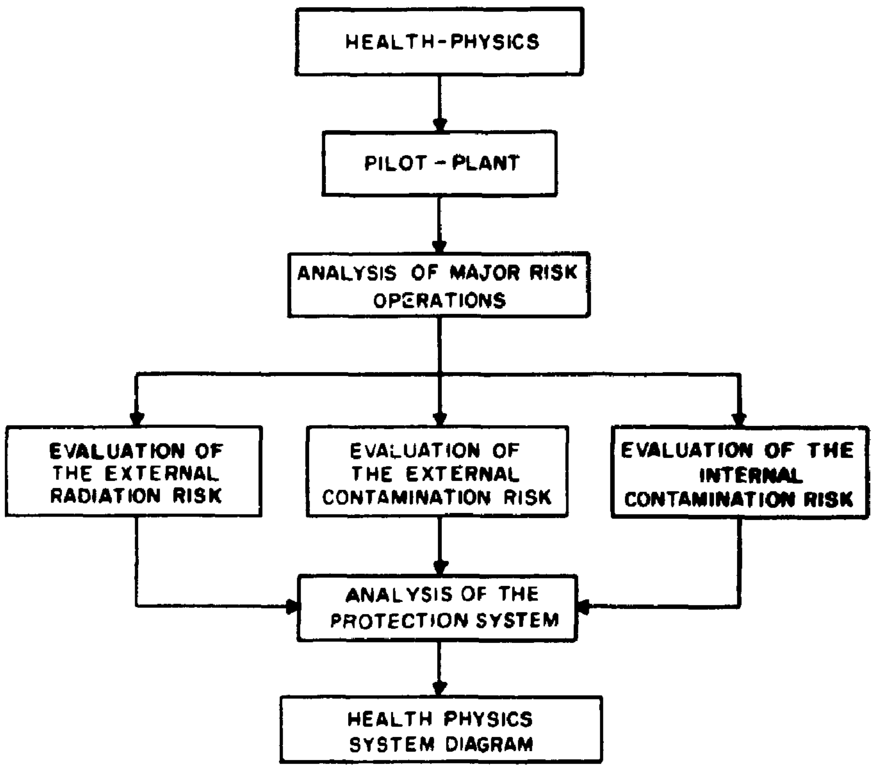
At the end of this work, we present a summary of the scheme of the Health Physics System established in this Pilot Plant

2 Uranium Purification Pilot Plant in I E A

The D E Q has installed two pilot uranium purification units, one using a purification method based in ionic exchange[1] and the other based on the conventional method of extraction with n butile

(*) Presented at the First Latin American Conference on Physics in Medicine and Radiation Protection, SP - Brasil 1972.

FLOWSHEET FOR THE PRELIMINARY INSTALLATION OF THE HEALTH PHYSICS SYSTEM AT THE URANIUM PURIFICATION PILOT PLANT OF ATOMIC ENERGY INSTITUTE IN S.PAULO, BRAZIL



fosphate (TBP) diluted These two units have been installed with the double purpose of training personnel in the uranium purification technology and developping the transformation and purification technologies for uranium compounds to obtain nuclear grade substances for utilization fuel elements for research reactors[16] The pilot unit whose central process is the purification of the uranil nitrate by means of ionic exchange in columns of cationic resin, is at present stopped, having already marked more than a year continuously colecting data It is a relatively small unit, with a capacity of 250 Kg of uranium a month, under the form of amonium diuranate The second unit does the uranium purification by means extraction of uranil nitrate with TBP-varsol in an environment of nitric acid, using three pulsed columns for the extraction, washing, and re extraction, with the aquous and organic stages in contact by opposing flow This units has a capacity to purify 5 kg of uranium in an hour In the two units the uranium solutions are, in the last stage, transformed in amonium diuranate, by precipitation with amonia gas Presently, it is this unit that is in operation[6] The D E Q has 42 workers, of which 8 are always working in the plant

The two pilot plants, until now have used only uranium concentrate, that is, sodium diuranate (SDU) derived from monazite sand chemical processing, being utilized as a subproduct in the industrial installations of the Administração da Produção da Monazita APM[14]

So that the reader can have a better idea of the Health Physics scheme, already implemented in the two uranium purification pilot plant, we will present next a diagram that summarizes the two chemical methods that the pilot plants apply Of course, this description give us a view about the motion of uranium in its different forms in the pilot plant areas (see annexed diagram)

3. Analysis of the operations of major risks.

From the point of view of Health Physics and only to facilitate evaluation, we will divide the D E Q in three sections:

a) Administration
b) Laboratories
c) Pilot Plant

The radiation levels in sections A and B oscillate between 0.02 and 0.06 mR/h Showing us there are no influences from the pilot plant Depending on the building material, size and room arrangements, these levels are normal for our environment

In view of this, our attention passed to the pilot plant

In view of the large use of acids and alcalis in the uranium purification process, all of the process is interesting, but there are six operations that are of the biggest interest, namely

OP 1 Handling of the sodium diuranate (SDU) in fine powder form, during the opening of the drums transfer to smaller containers (transparent plastic) and sample withdrawal which is done for weigthing

OP 2 The SDU transport in acrilic containers (25 kg of capacity) to supply of the dissolution reactor

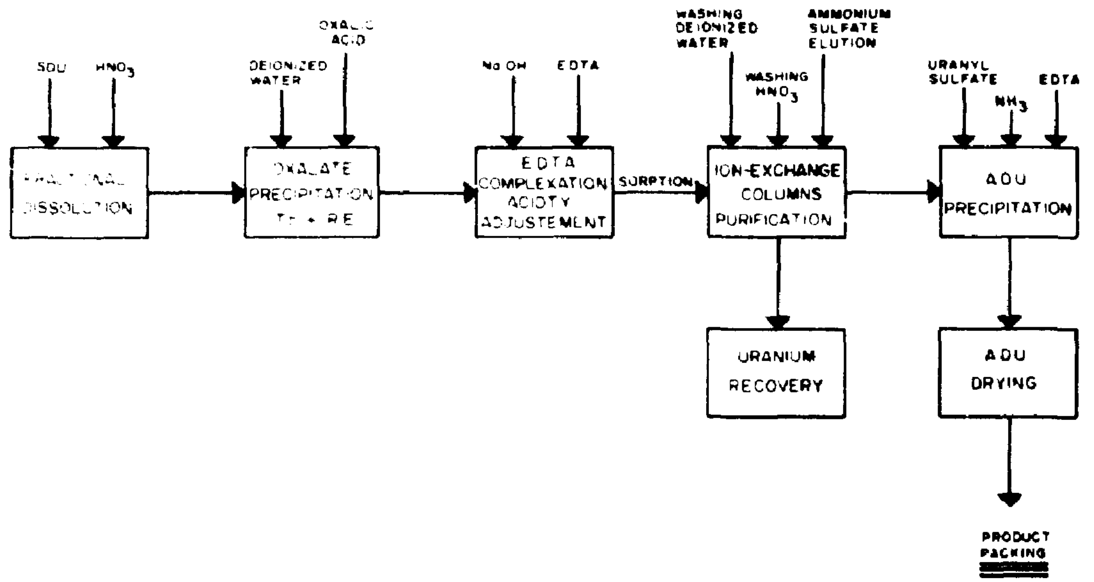
OP 3 Uranil nitrate withdrawal (in solution) for laboratory analysis

OP 4 Amonium diuranate (ADU) handling in a wet precipitated form and, the transport of this in a tray to the hot house

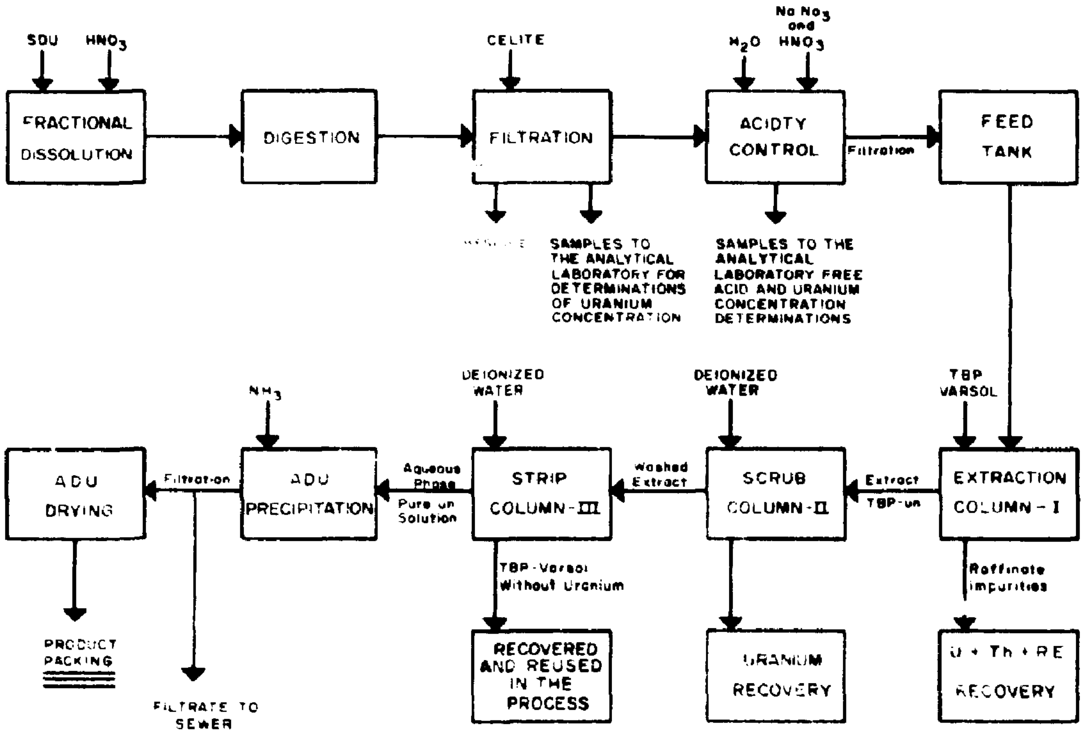
OP 5 ADU weighting and packaging placed in polietilene bags within cardboard cask

OP 6 SDU and ADU storage

SIMPLIFIED DIAGRAM FOR THE URANIUM PURIFICATION PROCESS BY ION EXCHANGE TECHNIQUE .



SIMPLIFIED DIAGRAM FOR THE SOLVENT EXTRACTION URANIUM PURIFICATION PROCESS.



Appendix A shows us, in a level plan of the D E Q reduced in a factor 1/100, the radiation levels in sections A and B as well as the place of the operations and of the monitoring points that we will mention in the subsequent explanations (tables) for section C

4 Radiation external hazards

4 1 Exposition levels

Several monitoring points are defined in accordance with the operations of major hazards

In table 1 besides the points, we put the mean values of the exposure in mR/h presented during 1971

Table I

Exposure levels in the uranium purification pilot plant in the I E A

POINT	mR/h	POINT	mR/h	POINT	mR/h
A	0.10	J	0,30	S	0.20
B	0.40	K	7 00	T	0 10
C	0 25	L	0 20	U	0 10
D	0 30	M	1.50	V	0 70
E	1 40	N	1,80	X	0 20
F	0 10	O	0.30	Y	0,80
G	0 10	P	0.30	W	0.40
H	0,10	Q	1.30	Z	0 20
I	0.40	R	0 10		

These exposure levels have been determined by use of a portable Geiger Mueller survey meter with a window of 1 5 to 2 mg/cm² in thickness and a dead time of 200 μsec The background radiation in the pilot plant was about 0.10 mR/h

As we can see the exposure levels in our pilot plant are relatively low

These monitorings are done daily with the purpose of control any alteration in the exposure levels

In those operations in which the personnel exposure excede 2 mR/h or there is uranium in powder we also monitore during each operation to avoid that the received dose is bigger than the maximum permissible established by the ICRP[10.8]

4 2 - Personal Dosimetry

This dosimetry is done through the film badge The results give us a good idea about the beta and gamma exposure levels, but not alpha, because the film envelope is sufficient to absorb all alpha particles It is obvious that an external personal dosimetry for alpha particles is not essential when the levels are so low as in our case, because besides the dose being low, the energy of the alpha particles is not sufficient to pass the epiderm However, even so in the future we will try, taking as model the Oak Ridge National Laboratory dosimeter to introduce an exposure measurement of the alpha radiation[18]

4 3 - Definition of personal permanence areas

Definition of the permanence areas for workers, is related to the obtained evaluation about the exposure and internal and/or external contamination levels

Table 2 presents the number of workers and the annual exposure accumulated.

Table II

YEAR/EXPOSURE	0,10 R	0,1 - 0,5 R	0,5 R
1967	8	1	0
1968	20	4	0
1969	19	7	0
1970	26	7	0
1971	34	8	0

Once the hazards are evaluated we can determine:

1 Permanence zone for non professionals.

This is defined as those places that the exposure or contamination hazards are practically nonexistent In the pilct plant this should be the path for visitors

2 Continuous permanence zone for workers

In this zone there is exposure and/or contamination hazard but it is well controled In the pilot plant this should correspond to the places where uranium purified

3 Conditional permanence zone

The workers permanence to do special works is conditional on the constant presence of an S P R D member At the present, this zone is formed by the places where the special operations specified in chapter 5 occur, and also in the SDU and ADU storage room

4 Forbidden permanence zone.

Those are the zones where the hazards are imminent or already exist The workers permanence is forbidden without express invitation This is only given after S P R.D autorization Until now, this zone has only appeared in contamination cases

5. - External contamination hazard.

5.1 - Contamination of persons, clothes, areas, etc.

The person's chance of external contamination is associated with accidents in the pilot plant, principally those resulting of the operations in which the workers mantain a close contact with the radioactive substances

Until now the personnel contamination in the pilot plant have been rare Worker leaving the plant are always monitored and in the end of each day they take a bath and the clothes are collected for monitoring Clothes contamination is more frequent, and once detected the clothes are collected and the user is monitored

The determination of area contamination is also done through area monitoring with the portable survey meter already mentioned and mantained at a distance of 1 cm from the area.

The mean levels of radiation in the pilot plant are presented in Table 3 The background radiation is 150 C P.M

Table III

Contamination levels in the ground of the Uranium Purification Pilot Plant

POINT	CPM	POINT	CPM	POINT	CPM
A	150	J	300	S	450
B	150	K	300	T	300
C	225	L	300	U	150
D	150	M	300	V	225
E	300	N	300	X	300
F	300	O	150	W	225
G	150	P	300	Y	300
H	300	Q	300	Z	450
I	300	R	225	-	-

100 CPM = $4{,}4 \times 10^{-9}$ μCi/cm^2

5.2 - Decontaminations

From the limits for surface contaminations that probably will be adopted by the Comissão Nacional de Energia Nuclear [2], we should have as maximum values given in the Table 4, but as we adopted the basic norm of the International Commission on Radiological Protection (ICRP) to mantain the levels as low as practicable, we proceed to the decontamination of any utensil, article, clothing on area that presents levels higher than the normal

Table IV

Limits for surface contaminations

Surface Type	Activity (μCi/cm^2)
Inactive and low activity areas	10^{-4}
Active areas. Special clothes not normally used in inactive areas	10^{-3}
Skin	10^{-5}

The pilot plant floor is decontamined once every weeks using a solution of HNO_3 (0.02M) followed by detergent soap; this is done whether the floor is not contaminated.

About the clothes and the utensils, they ser... aste disposal and decontaminations laboratory, where decontamination is carried out

5.3 - Protective Clothing

To select protective clothing for the workers of the pilot plant we have considered several factors, a few of general character and others specific, namely:

a - worker confort during use
b - case of decontamination
c - radioactive agent contamination, in the case, sodium diuranate, uranil nitrate, wastes

resulting from the chemical processing and amonium diuranate

d presence of acid and alcalis.

For the pilot plant, two types of clothes are being selected depending on whether or not the worker is connected to the operations that present radioactive hazards. In this case, the clothes are made of thick cloth and boots of thick leather, with elastic on each side and neopreme sole with double reinforcement (fig. 1)

In the case that the worker is to be connected with operations that present radiation hazards his clothes consist of: 1) PVC lab coat (resistant to the acid and alcalis, besides giving protection against alpha radiation) sewn with nylon thread, long sleeve with elastic fastener, and completely closed in the back. 2) Rubber or PVC gloves, of several sizes, adequate for the type of operation done. 3) boots, already described (fig. 2)

For both types of clothes, when necessary, as for instance, to avoid head impacts or contaminations provoked by possible aerial ducts leakage, that one meet in the plant and contains acids, uranil nitrate solutions, etc. the worker uses a fiber helmet (fig. 3)

6 Internal Contamination Hazards.

6.1 - Air Contamination.

Air monitoring is been done, having in view evaluation of the possible air contamination, for those operations defined as the major hazards.

For equipment, a Millipore pump with filter of diameter 47 mm of the same mark was used.

In each place the pump remained on 30 minutes with an out let of 0.5 feet^3/minute.

The Millipore Filter was counted in a Philips apparatus formed by a Geiger Mueller tube, a power supply, a scaler and a rate meter.

The G. M. tube of the own Philips is the model 18505 that has a window thickness between 1.5 and 2.0 mg/cm^2 and has a useful diameter of 0.5 cm, giving in this manner the possibility of to measure alpha particles with an energy larger than 3.5 MeV.

It happens however that the G. M. tube shielding is not very efficient and the background radiation is too high, equivalent to more or less 1×10^{-10} μCi/cm^3. This is already above the maximum permissible concentraion for natural uranium, that is, 6×10^{-11} μCi/cm^3. In this manner we can only obtain results relatives to this background radiation for the places considered of the major importance. Such results can be seen in Table 5.

The monitoring places are in the appendix A.

In view of this, we bought and installed an air-borne continuously monitoring device, and now we are calibrating it to obtain the answer in terms of concentration.

Table V

Radiation level, above the Back ground in the air of the Pilot Plant.

Place	Number of times higher than the B G
A_1	2
A_2	10
A_3	1
A_4	1
A_5	2
A_6	4
A_7	1

At the same time, we are trying to improve the G. M tube and shielding, to see if we can lower the background

6.2. - Urine Analysis.

6.2.1. - Internal Contamination Control by determining the Uranium content urine.

In spite of a lot of controversy, due to the large number of variables that enter in the uranium quantity excreted, this type of analysis serves as support, reinforcing the other precautions taken. Although there is doubt concerning the correlation between the values in the urine and the exposure to the uranium powder, the urine analysis serve as a forewarning, because high results drive us to verify the environment conditions, and once on adequate program is established, the results go to indicate, within certain limits, if the exposure has been higher than that considered as the maximum permissible [5]

The first tests develop a uranium analysis method for urine, here in the I.E A, date from 1968 The technique used is based on others already in use, although modified and adapted to our conditions in the Chemistry Engineering Division [5]

In this work we present for the first time, the results obtained for the urine of the personnel since March 1971, the date since which the analyses have been done by S P R D. personnel

6.2.2. - Toxicity and maximum permissibles concentrations.

Uranium presents chemical and radioactive toxicity Both of these are important, depending on the solubility of the uranium in question

The critical organs for natural uranium are:

U(nat) soluble - critical organ: kidneys-Chemical toxicity

- U(nat) insoluble - crit organ: lungs-Radioactive toxicity

After intake, the uranium is eliminate, preferentially in the urine.

Based on recommended values from different Health Physics Organizations and data reported in several works[3,4,7,9,11,13,15], we adopt the following parameters:

- Maximum level of uranium in the urine for follow up analysis (investigation level) 50μg U/1 urine
- Maximum level of uranium in the urine for immediate measures (action level) 100 μg U/1 urine

6.2.3 - Analysis method[5]

The selected analysis procedure is fluorimetric in view of its sensitivity[17].

The method is based on the characteristic fluorescence of the uranium salts when exposed to the ultra-violet light. The light intensity emitted is a linear function of the uranium quantity present in the sample, in a range of 10^{-10} to 1μg per container[19]. The uranium fluorescence is intensified by his fusion with solids as NaF and LiF.

The fluorescence intensity is measured in a fluorometer made by "Jarrel Ash" with a melting mixture of two carbonates and one fluoride, namely, Na_2CO_3, K_2CO_3 e NH_4F, in the mass rate 45.5 : 45.5 : 9 and give us quite reproducible results.

To reduce the influence of the extinction provoked by certain elements, determination by the direct analysis of urine in small samples has been selected (0,1 ml of urine previously mineralizated) followed by the adition of a standard (two additions of 0.1 μg uranium). The uranium concentration in the sample is obtained as shows in the figure 4.

Point 0 corresponds to the reading obtained through the analysis of the sample; the points 0,1 μg and 0.2 μg represent the addition of two standard done in the original sample in the same container, in sequence, conserving the same experimental conditions. The x value shows us the uranium concentration present in the sample, having eliminated as much as possible, the several interferences.

Note that of the reading we compare to the value of a blank made only with the fused flux.

All the analysis are done in duplicate.

The method has been shown to be satisfactory for the control in question, with reprodutible results. The experimental error in works of this type of measurement is between 5 and 50%, our error is about 25%.

6.2.4. - Results.

The results presented here are preliminary and come from urine analysis of 42 persons, during the year of 1971.

We have done in the start of the program a survey of the uranium level in the urine of all the personnel, after we pass to do routine analysis only for the Plant personnel and other persons that are in contact with the uranium and sporadicly, the rest of the personnel only for control.

In the Table 6, the uranium quantities excreted in the urine by the personnel that work in the D.E.Q. are summarized.

Table VI

U excreted in the urine by the personnel of DEQ

D E Q Sections	Sample Number	Sample number put in the several intervals of U concentration (μg/l)					
		0 10	11-20	21 30	31-40	41-50	51 80
Plant (8 persons)	45	5	7	7	14	2	10
Analitic Laboratory (23 persons)	71	29	14	17	2	5	4
Plant Technical Assistance (10 persons)	34	10	8	9	4	3	0
Secretary (1 person)	3	3	0	0	0	0	0
TOTAL	153	47	29	33	20	10	14
Note: Above 80 μg U/l results = 0					–	–	–

One note, as one hopes, the biggest levels are for the personnel that work in the Pilot Plant and that it has not exceded the value of 80 μg U/1

If we observe the data from several countries[3,4,7,9,12,13,15], we note that most samples are in the range of 0 to 10 μg of U/1 of urine Our values, meanwhile, in spite of having the biggest number of samples in the range of 0 to 10 μg U/1, present several distributed in the following ranges up to 50 μg U/1.

Another fact is that if although there is not an official rotation among the Plant personnel, there is rotation of work. In this manner, we avoid big values like those determined in other countries, sometimes higher than 100 μg U/1[5]

Only 14 analysis gave results in the interval of 50 to 80 μg U/1, being that 10 belong to 5 of the 8 person that work in the Plant and the other 4 to 3 persons, over 23, that belong to the Analitical Laboratory The detailed distribution is:

Plant:	TOTAL n° of Analysis
2 persons with 3 analysis > 50 μg	6
1 person with 2 analysis > 50 μg	2
2 persons with 1 analysis > 50 μg	2
TOTAL	10

Analitic Laboratory:	TOTAL n° of Analysis
1 person with 2 analysis > 50 μg	2
2 persons with 1 analysis > 50 μg	2
TOTAL	4

Of these analysis, only 2 are subsequent to our starting use of masks, proving in this manner the efficiency of these masks

6 3. Respiratory Protection

The operations that present the largest air contamination hazards are accomplished in closed rooms with air exhaust pipes

These pipes draw in the air for a cycle filters (fig 5) that absorve all the powder content in the air before releasing to the air in the atmosphere

Concerning the worker, we have put in use two respiratory system, namely: a) mask with filter against gases and radioactive powders; b) protection helmet with compressed air to breath

The mask is used for isolated operations, of short duration and which present the problems of injurions vapours and radioactive powders (fig. 6)

The mask body (Panoramic-S-drager) is made of black neopreme, having an acoustic membrane that permits communication an inner little mask covering the mouth and nose, Plexigum panoramic visor and an in and out valve

The filter (B62OST DRAGER) is compound of obstacles that retain radioactive particles and acid and organic vapours

The respiratory protection helmet with pressed air is used in the exhaust rooms where there is ADU and/or SDU handling and also during reactor charge where the SDU dissolving is done

To use this type of equipment, we have to use extention of the compressed air from the Pilot Plant to the exhaust rooms

Before entering the room, an air purification system has been introduced, with filter R-0500 and filter with purger After the filter we have done a division of the pipes to the two rooms In each room a compressed air suppriment valve has been installed. A third valve has been installed externally to the rooms, and is used during the reactor charge In this manner, the worker that uses the helmet connects his extention to this valve to receive the filtered air The helmet, done in fiberglass has its own regulating system for the passing air The use of this protection helmet in indicated for a long duration operation and the worker protection is complete Its use has been very efficient in the following operations:

a) SDU weighting and transference operation (fig 7)
b) Dissolution reactor loading with SDU (fig 8)
c) Packing of the ADU in small card board casks (fig 9)

For emmergency operations we maintain a protection mask with its own compressed air suppriment The air charge has a mean duration of 50 minutes and the assembly has an alarm system that warns the end of the air charge, giving time to the user to withdraw from the emmergency operation before the end of the charge (fig 10)

7 - Conclusions.

Although the protection system that is already in a large part implemented for the environmental and personal control in the areas of the uranium purification pilot unit in the D E Q of the I E.A is not the final one, it has permitted an efficient control for the work conditions of the workers of that area. The permissible levels of radiation and the uranium presence in the urine are not being exceeded

The determination of the uranium in the urine of the pilot plant personnel is done in a routine manner, although one has not selected a definitive method about the samples taking and their frequency Nevertheless, the number of samples already taken and analyzed assure us that the environment conditions of work, are under control and that the personnel is conscious about the potential radiation hazards and operate within good protection norms

Equipment such as the air borne continuously monitoring (fig 11), the hand and clothes contamination detector and portable survey meter with zinc sulphete cristal (fig 12), all of them for alpha radiation are already been adquired and one finds them in the places of use, although not all of them are turned on.

With partial implantation of the protection system, the personnel of the pilot plant are to have protection clothes more functional and adequated for the type of work, such as plastic lab coat, gloves, boots and mask. All this can be noted by urine analysis.

Of enormous importance, we want to emphasize in this work, that besides colaboration to create a permanent control in the uranium working areas, maintaining all the places cleaned and decontamined, and assuring tranquility to all personnel and supervisors, the presence of the health physics staff has had the worth to create a profissional conscience in reference to environmental and also personnel hygiene with relation to the radiation for all the D.E.Q. personnel.

Concluding, the scheme implanted for health physics in the uranium purification pilot unities in the D.E.Q. of the I.E.A., represents for us, a pioneer experience in the country, having now staff specifically trained for this type of work, and that will endeavour to continue to improve this system which is giving us positive results. It is obvious that we will not stop our endeavour to improve our technique and to try to reduce the radiation levels even more.

HEALTH PHYSICS SYSTEM DIAGRAM AT THE URANIUM PURIFICATION PILOT PLANT, ATOMIC ENERGY INSTITUTE, S PAULO BRAZIL

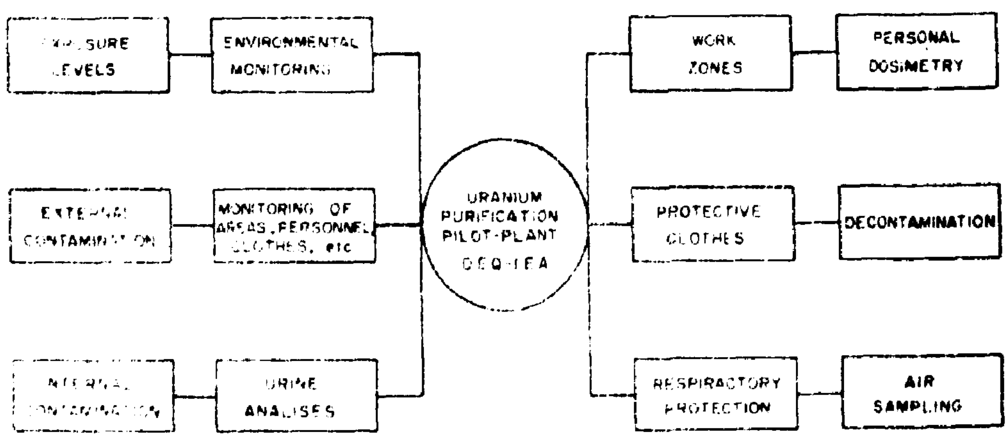


FIGURE CAPTIONS

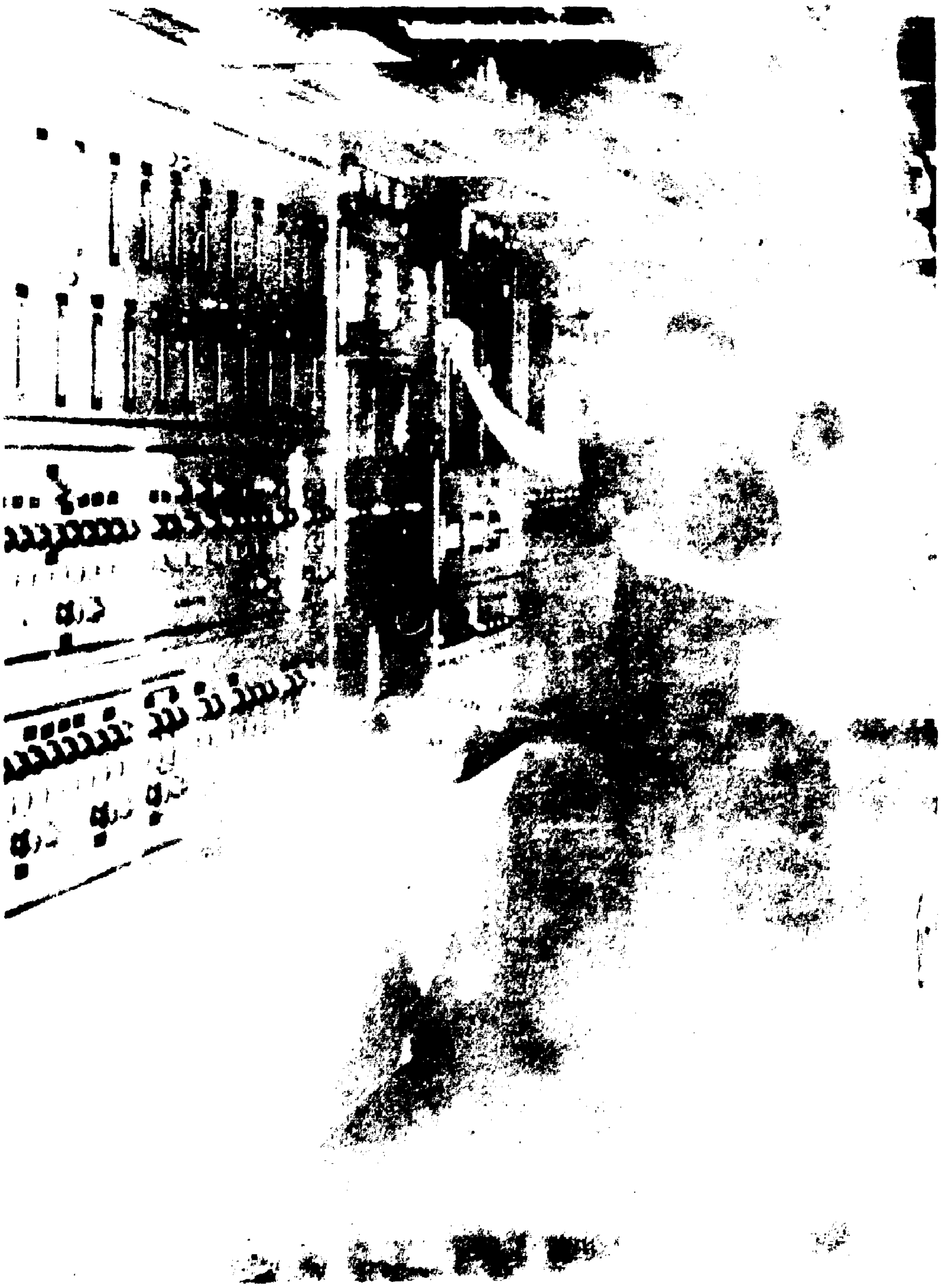


Figure 1

Figure 2

Figure 3

Figure 4

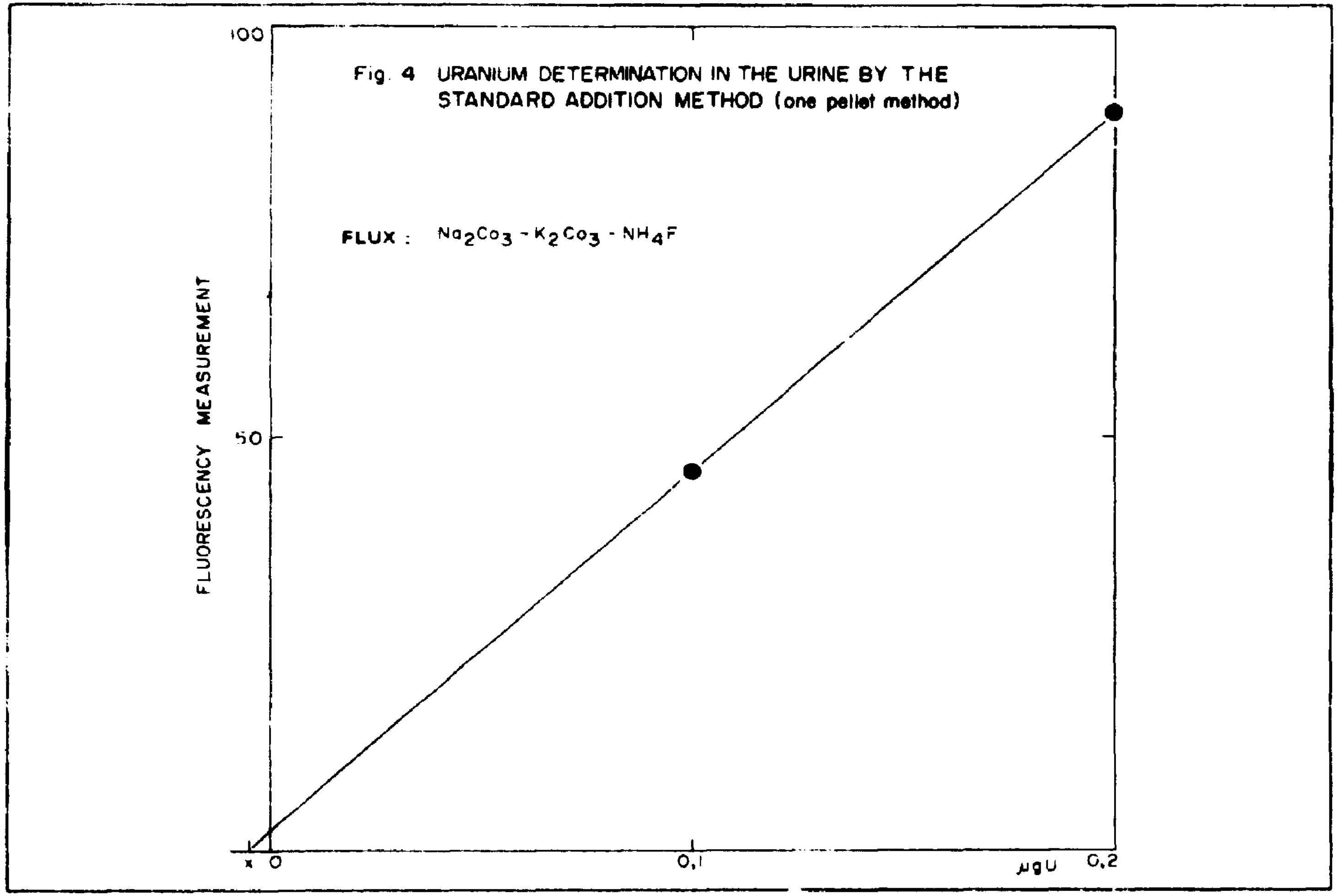


Fig. 4 URANIUM DETERMINATION IN THE URINE BY THE STANDARD ADDITION METHOD (one pellet method)

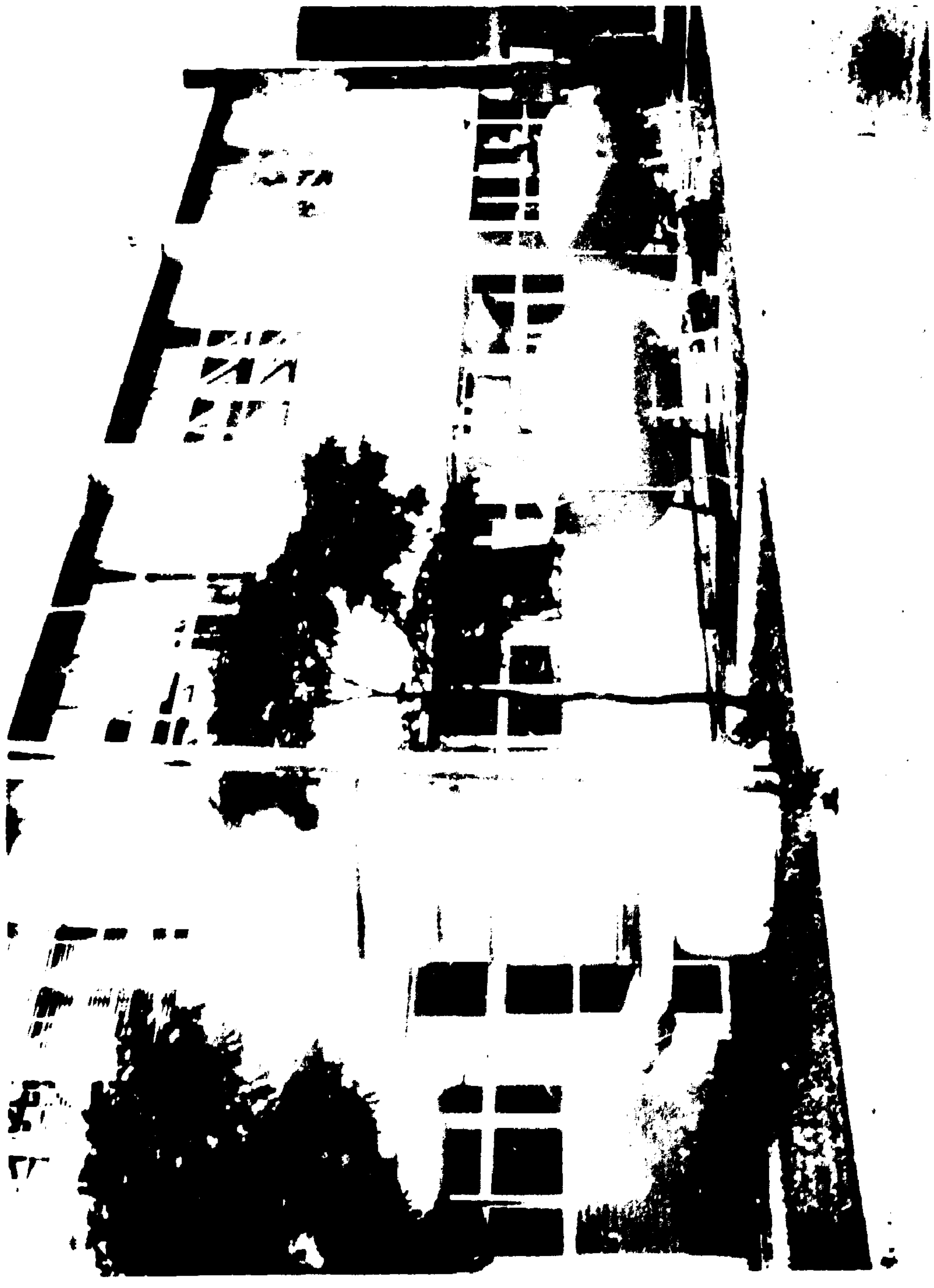


Figure 5

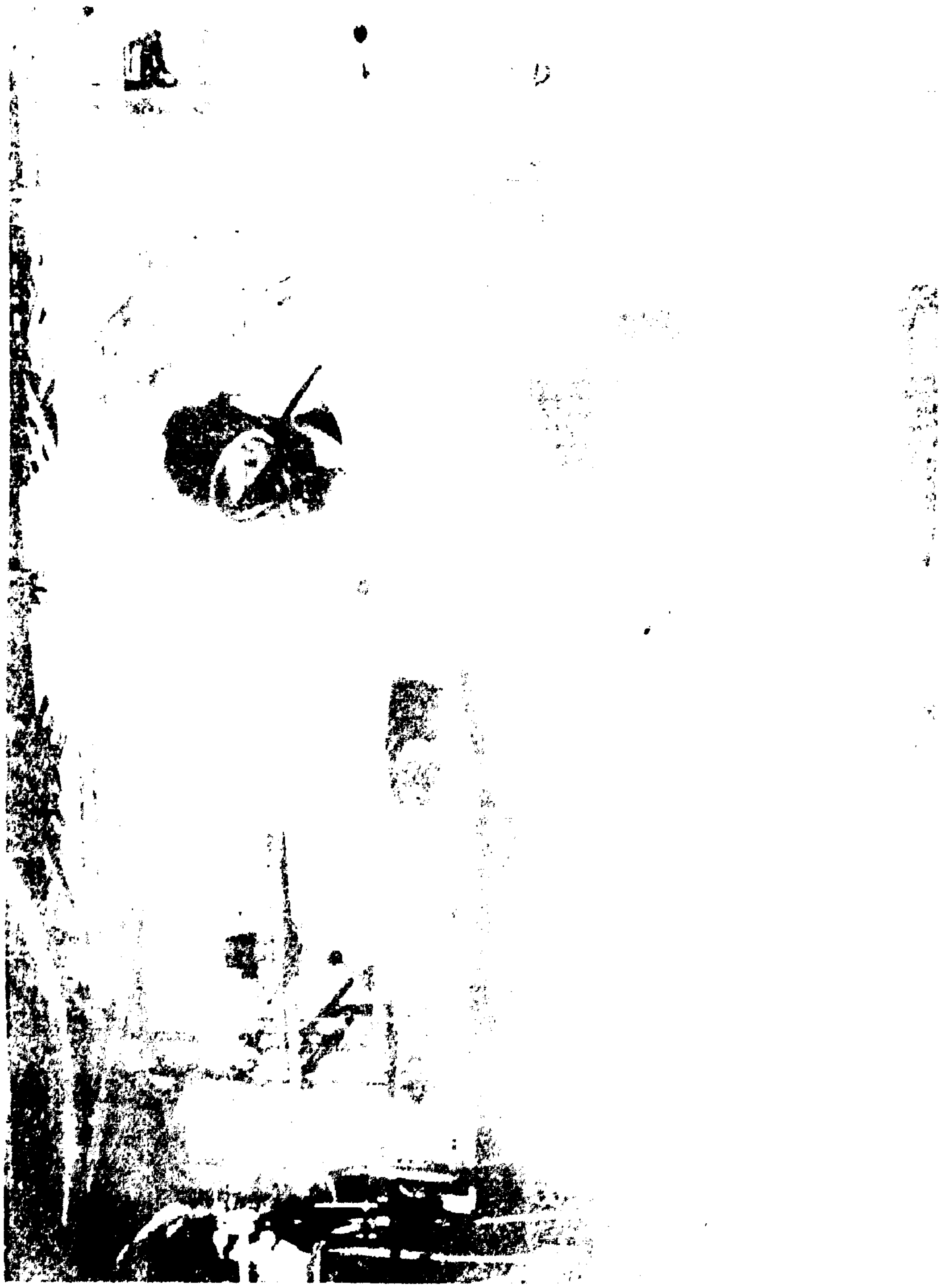


Figure 6

Figure 7

Figure 8

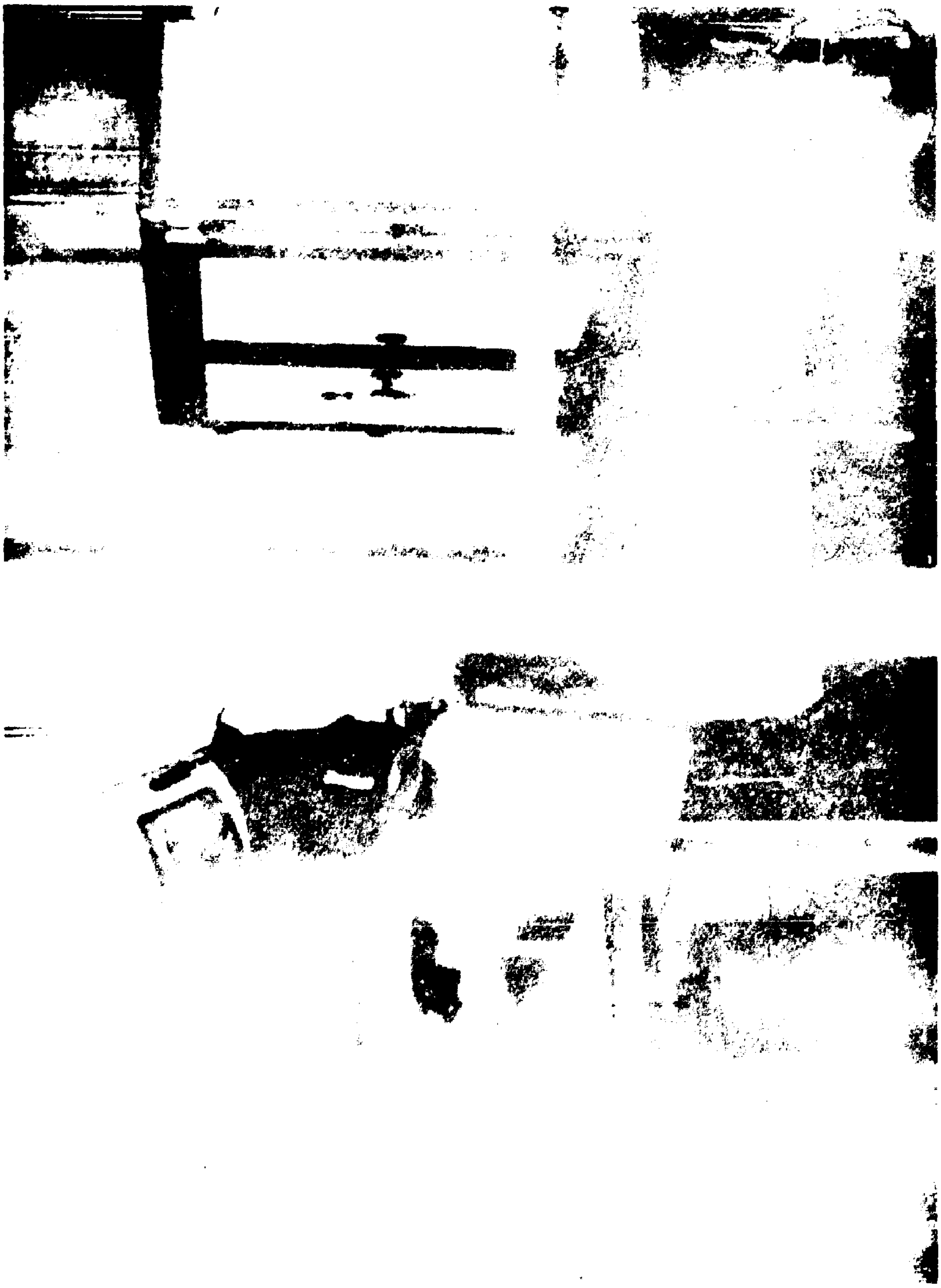


Figure 9

Figure 10

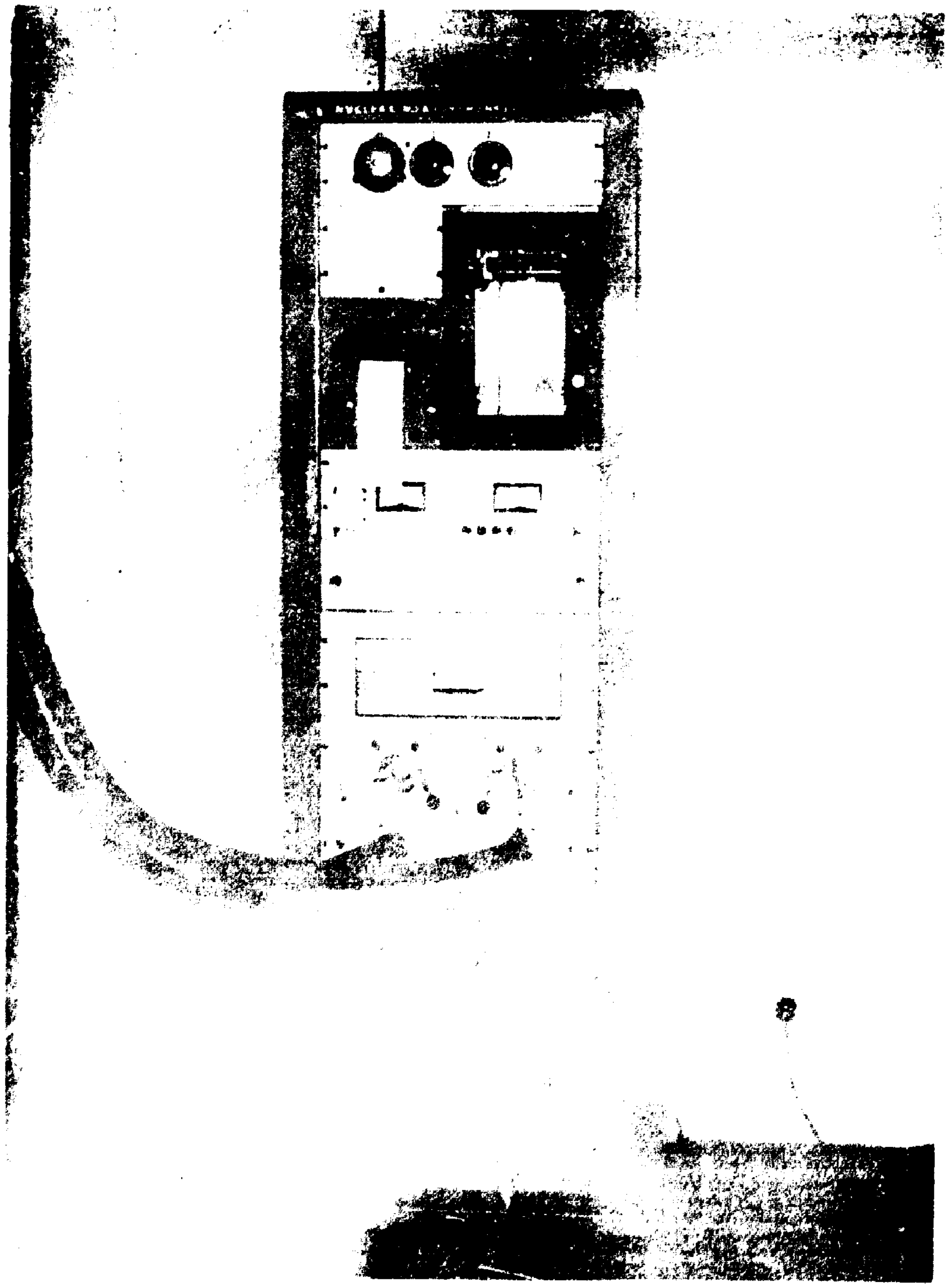


Figure 11

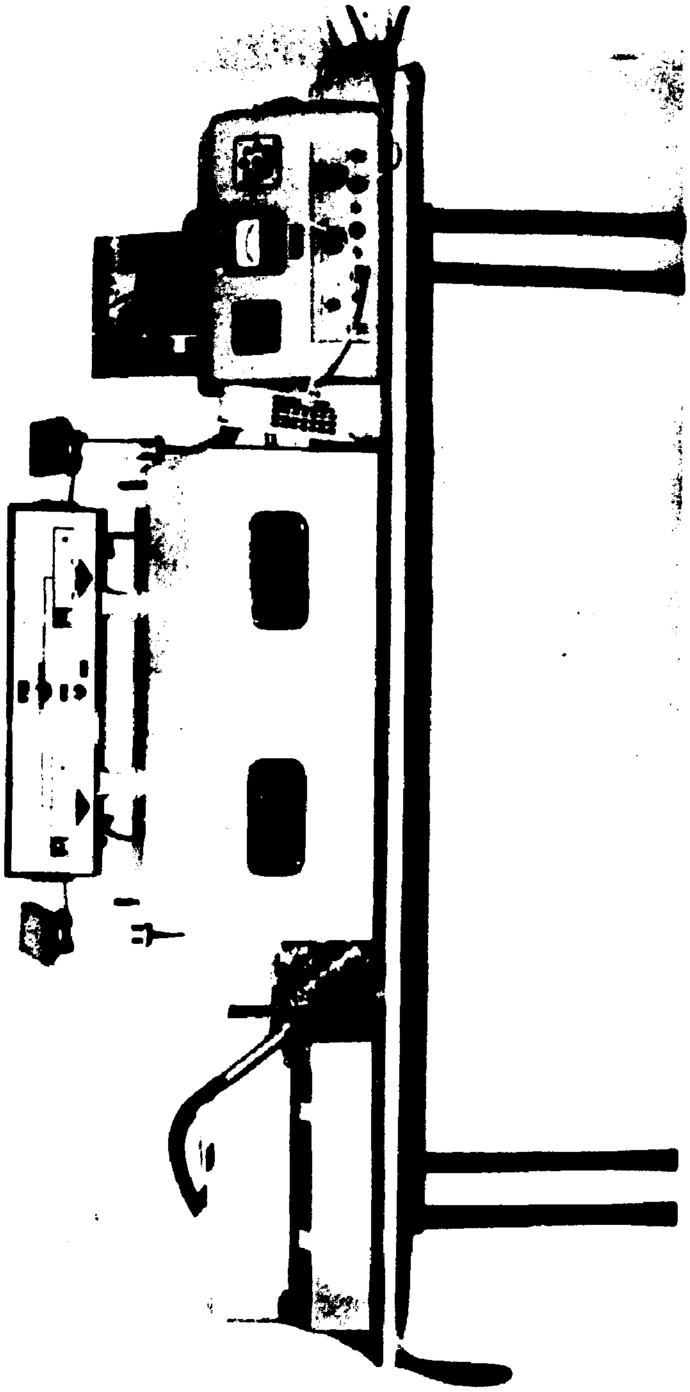
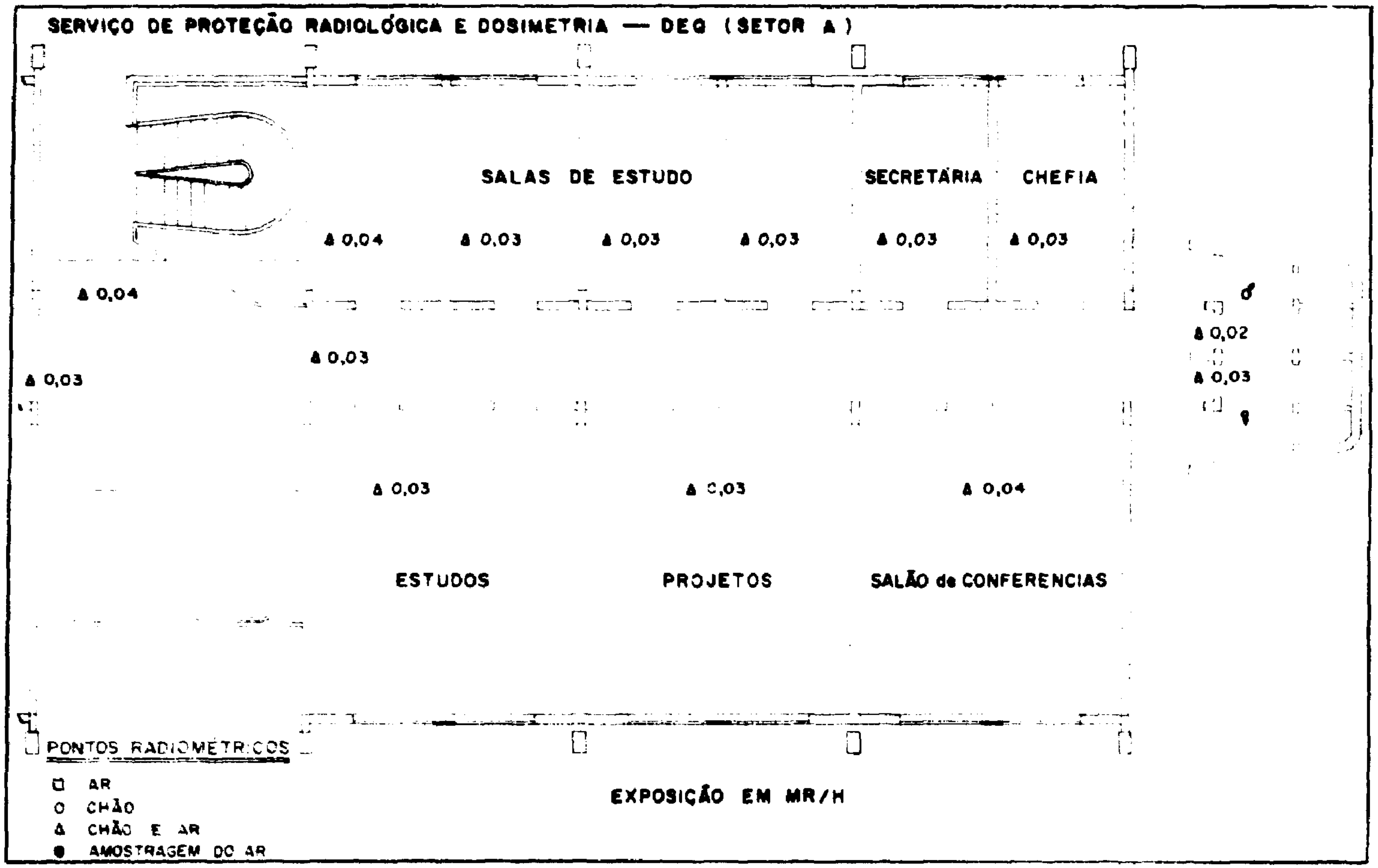
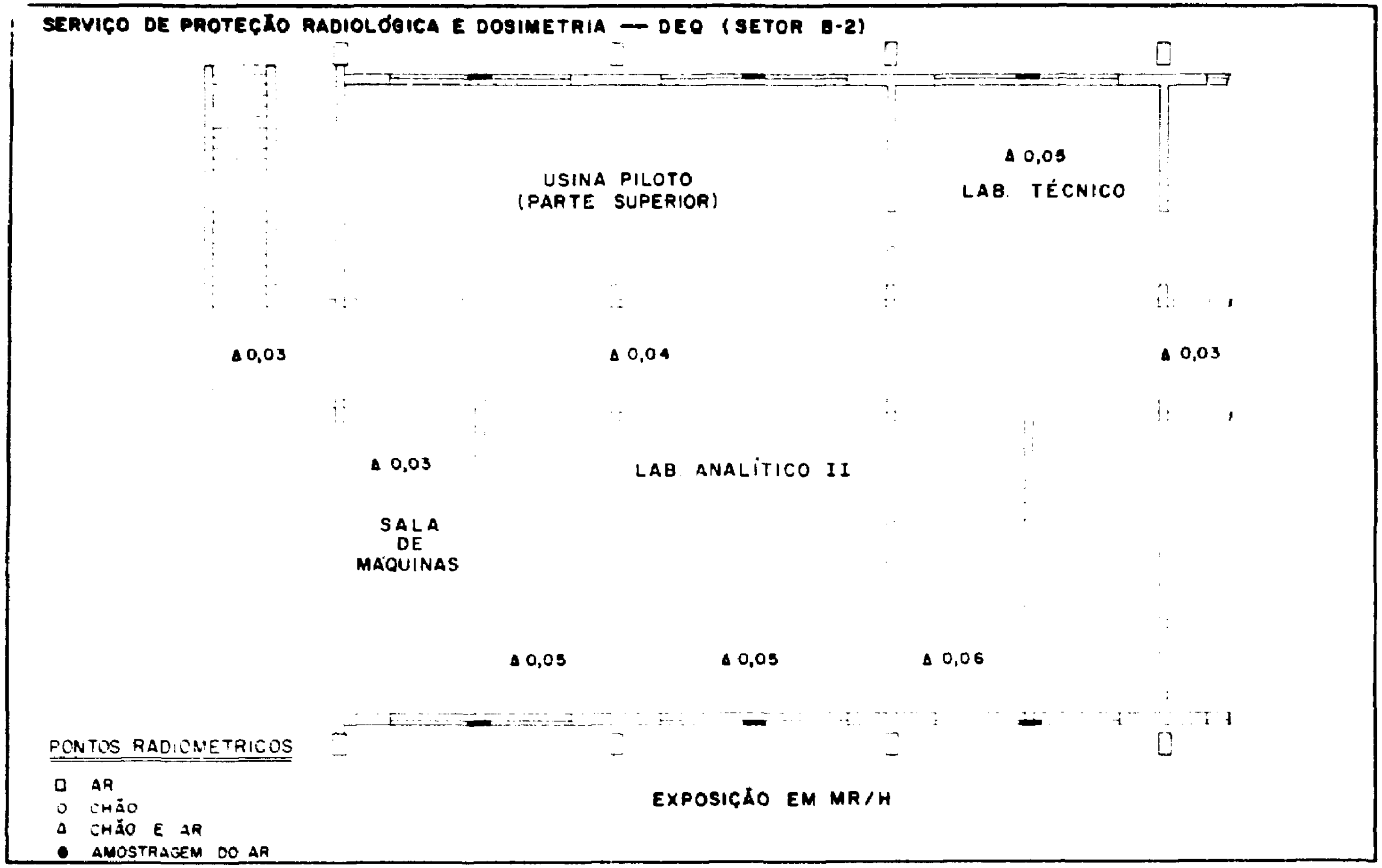


Figure 12



Appendix A
Monitoring places in the Pilot Plant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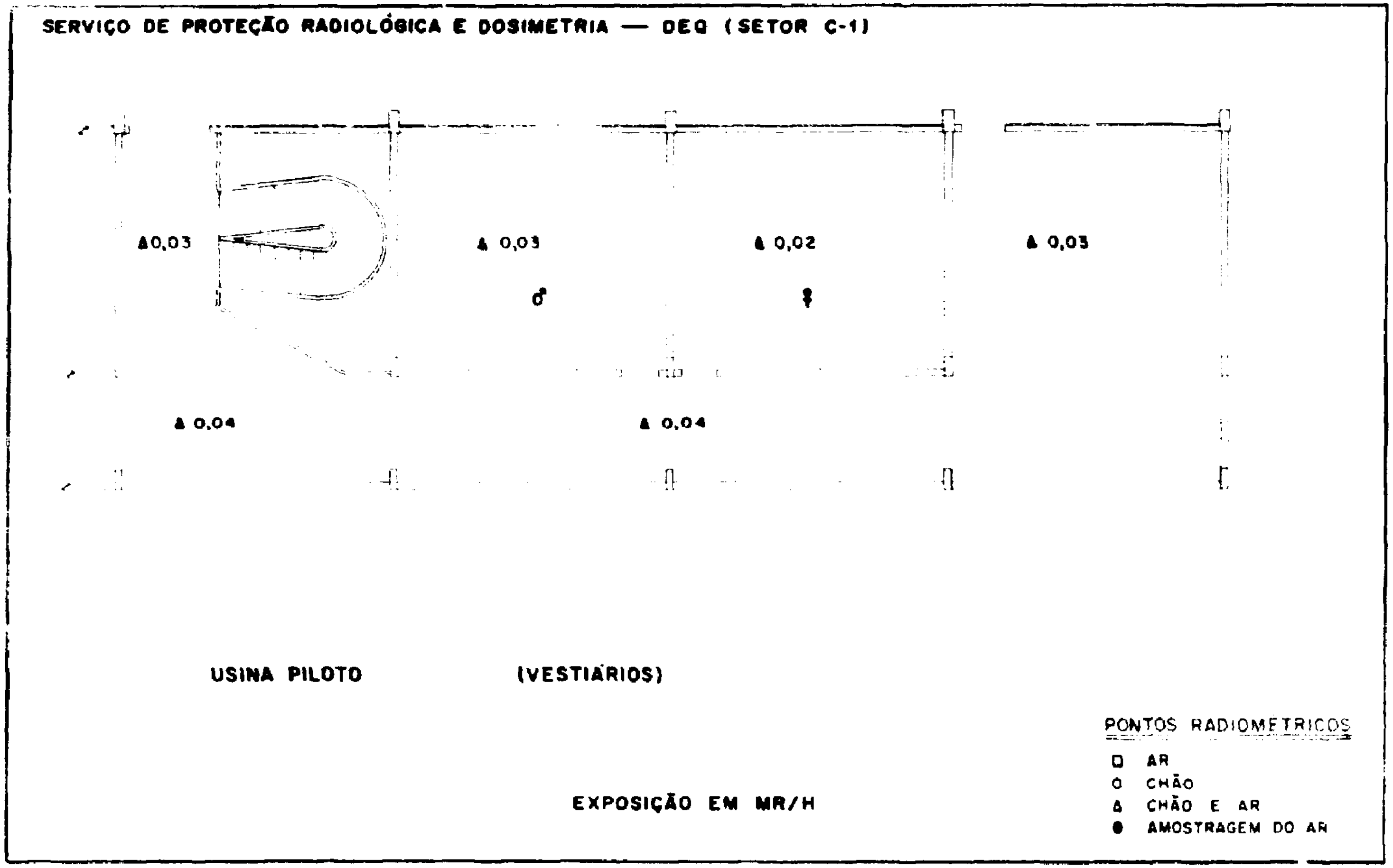
● AMOSTRAGEM DO AR
EXPOSIÇÃO EM MR/H


SERVIÇO DE PROTEÇÃO RADIOLÓGICA E DOSIMETRIA — DEQ (SETOR C-1)
▲0,03
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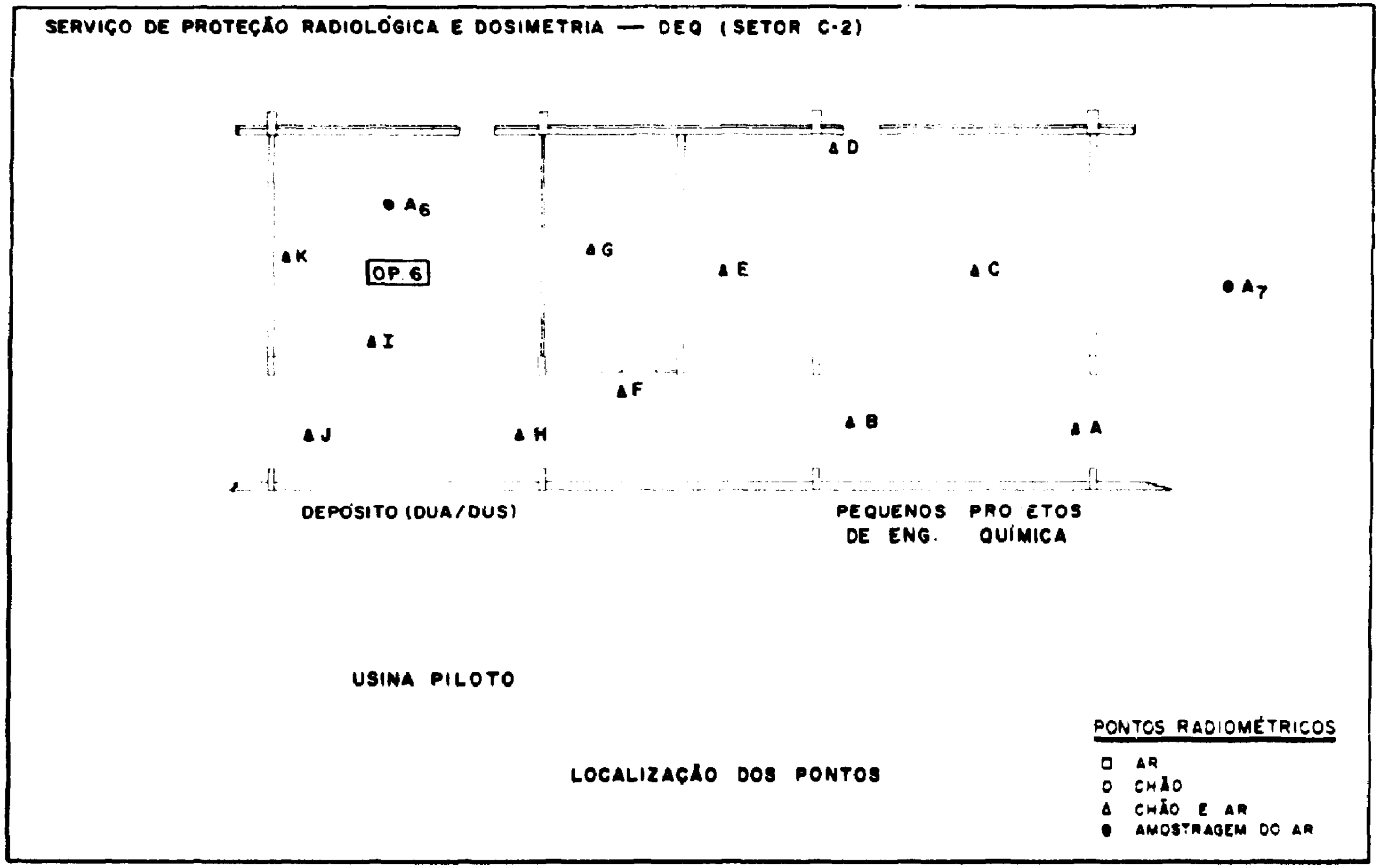
USINA PILOTO
(VESTIÁRIOS)
EXPOSIÇÃO EM MR/H
PONTOS RADIOMETRICOS
□ AR
○ CHÃO
△ CHÃO E AR
● AMOSTRAGEM DO AR


SERVIÇO DE PROTEÇÃO RADIOLÓGICA E DOSIMETRIA — DEQ (SETOR C-2)
D
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F
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A
J
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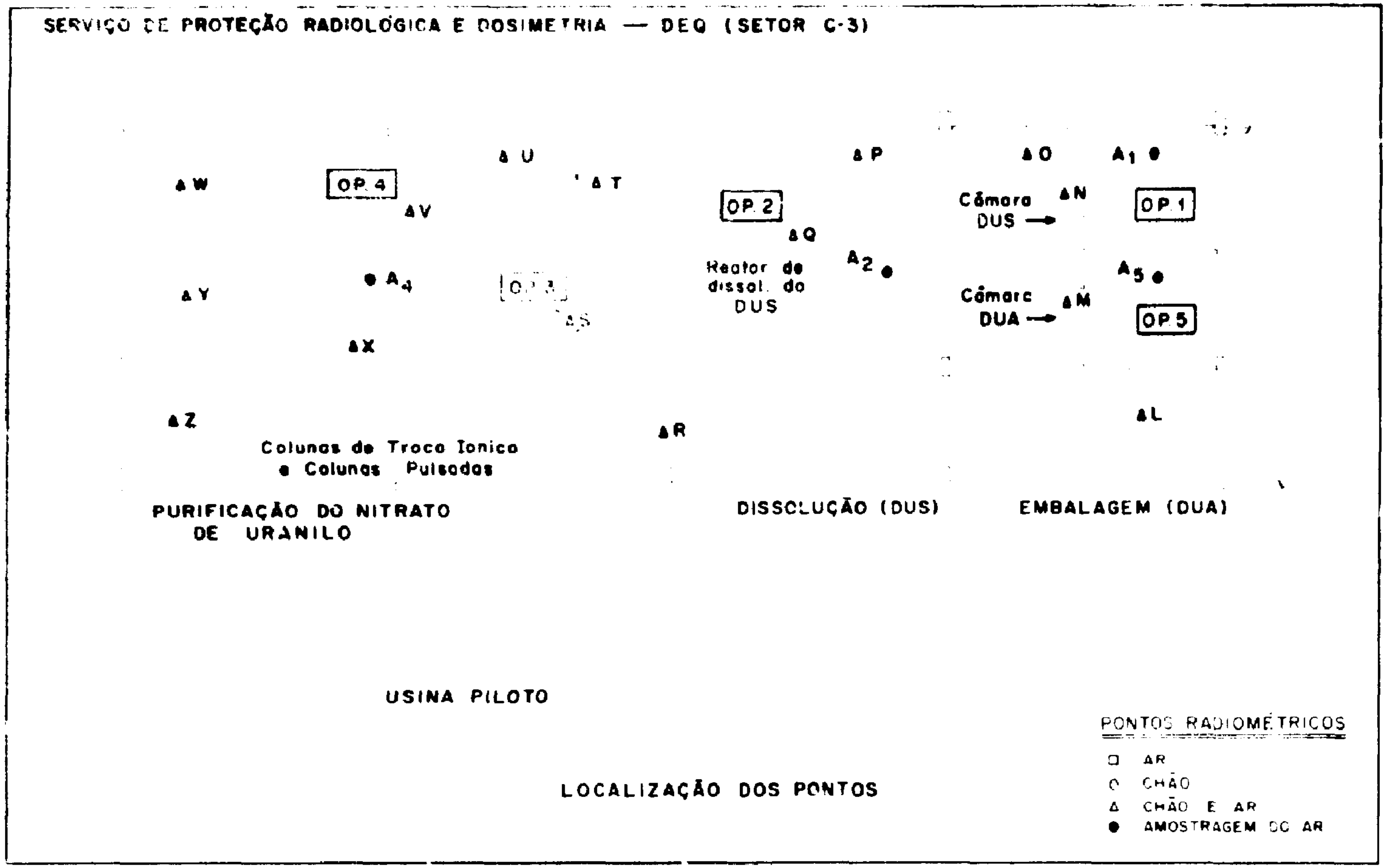
DEPÓSITO (DUA/DUS)
PEQUENOS PROJETOS
DE ENG. QUÍMICA
USINA PILOTO
LOCALIZAÇÃO DOS PONTOS
PONTOS RADIOMÉTRICOS
AR
CHÃO
CHÃO E AR
AMOSTRAGEM DO AR


SERVIÇO DE PROTEÇÃO RADIOLÓGICA E DOSIMETRIA — DEQ (SETOR C-3)
U
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OP.1
Reator de dissol. do DUS
A2
A5
A4
OP.3
Y
S
Câmara DUA
M
OP.5
X
L
Z
R
Colunas de Troca Ionica e Colunas Pulsadas
PURIFICAÇÃO DO NITRATO DE URANILO
DISSOLUÇÃO (DUS)
EMBALAGEM (DUA)
USINA PILOTO
PONTOS RADIOMÉTRICOS
AR
CHÃO
CHÃO E AR
AMOSTRAGEM DO AR
LOCALIZAÇÃO DOS PONTOS

RESUMO

Apos descrevermos os dois processos de purificação usados na Divisão de Engenharia Química do Instituto de Energia Atômica, nós examinamos os possíveis danos e os métodos adequados para controlá-los ou eliminá-los. Como estes processos de purificação apresentam várias etapas, em cada uma nós avaliamos os danos e tentamos fornecer soluções adequadas para proteger ambos, pessoas e instrumentos, dos perigos potenciais de radiação.

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