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CONTRIBUTION OF IPEN-CNEN/SP TO THE SEMINAR ON MANAGEMENT OPTIONS FOR LOW AND INTERMEDIATE-LEVEL WASTES IN LATIN AMERICA (IAEA)

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DEPARTAMENTO DE CICLO DE COMBUSTÍVEL

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INTERMEDIATE-LEVEL RADIOACTIVE
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SUMMARY

page

CONTRIBUTION OF IPEN–CNEN/SP TO THE SEMINAR ON MANAGEMENT OPTIONS FOR LOW AND INTERMEDIATE–LEVEL WASTES IN LATIN AMERICA (IAEA)

Rio de Janeiro, Brazil
13-17 October 1986

FOREWORD

This publication represents the participation of the IPEN–CNEN/SP at the Seminar on Management Options for Low and Intermediate-Level Wastes in Latin America held in Rio de Janeiro during the period of 13-17 October 1986 and sponsored by the International Atomic Energy Agency.

From a total of 36 papers accepted for presentation IPEN–CNEN/SP contributed with 15 of them which were distributed among five of the six sections programmed. Those 15 papers were distributed in the sections according to the following subjects: techniques and practices for pre-treatment of radioactive wastes; treatment options for low and intermediate-level radioactive wastes; conditioning options for low and intermediate-level radioactive wastes; packing, transportation and interim storage techniques; and practices and disposal options for low and intermediate-level radioactive wastes.

The authors are indebted to a large number of people who assisted in the conduct of the experiments and in the production of the papers presented in this report. Engº H. R. Franzen provided invaluable support as program manager. Dr. R. Fulfaro provided valuable backing and encouragement in the realization of the projects. Thanks are also due to the technical people belonging to the Nuclear Fuel Cycle Department of IPEN–CNEN/SP and to Mrs. Walkiria G. dos Santos and Mrs. Thereza T. Iaria who carefully typed the papers presented here.

These works were supported by the Comissão Nacional de Energia Nuclear as part of the waste management program.

Dr. Achilles Alfonso Suarez

DESIGN OF A PACKAGE FOR ENCAPSULATION, TRANSPORT AND FINAL DISPOSAL OF DISCARDED RADIUM NEEDLES

J. P. Villalobos, R. Vicente, A. A. Suarez

ABSTRACT

This paper describes the development work aiming at designing and building a package for radium needles. These needles comes mainly from hospitals where they were used in radiotherapy, but industrial sources may be stored in this package. It was previously defined that the package should be in a form which is acceptable in final disposal site, and also conform the type A – non-special form, of the "Regulations for the safe transport of radioactive materials", safety series nº 6, 1985 Edition, published by the IAEA. As the final disposal site has not yet been defined, there is not a list of requirements to be fulfilled by the package. Hence the adopted criterion was to keep the source in a non readily dispersible form. The development work was undertaken to select packaging materials as shielding, structural and absorbing materials, to define package geometries and encapsulation process. As this waste will be temporally stored before transport to the final disposal site an additional requirement was considered: radon leakage from the package should be limited. To find out a figure for the maximum leakage rate, an interim storage scenario was defined. The incurred doses on operating personnel from the radon leakage should not exceed maximum permissible doses for workers. The package has an inner stainless steel cilindrical capsule, with two cubic decimeter in volume, in which the radium needles are enclosed with charcoal as an inert absorbing material. The shielding materials are a 0.5 cm thick lead layer inside a concrete shell 23 cm thick. The outer package is a 200 liters drum.

Pertinent type A package tests were performed. This include integrity of containment system and shielding.

INTRODUCTION

Radium 226 is one of the most hazardous materials known. Radium 226 replaces calcium in the bone structure and is a source of irradiation to the blood-forming organs. This, behavior along with its long half-life (1.620 years) and high radiation energies, places it in the highest radiotoxicity group. It also has the longest history of use of any radioactive material, and most of the standards for the effects of ionizing radiation on man are based on this material.

Radium 226 is used as a radioactive source in medical practice and industry. Its primary value has been in the treatment of cancers by insertion of encapsulated needle sources directly into the tumor or by means of moulded applicators that hold the source next to the tumor. Industrial applications have included radium for radiography, certain electronic values, switches, and luminous paints. The use of radium in all of these applications has been reduced greatly with the availability of safer and cheaper radioactive materials although many radium applications still exist.

This paper describes the design and building of a package acceptable to hold discarded radium sources in non special form for interim storage, transportation and final disposal.

PACKAGING DESIGN

A schematic drawing of the packaging for interim storage, transportation and final disposal is shown in figure 1. It is composed of different materials layers in order to promote an adequate leaktightness, shielding and mechanical integrity. The source material is enclosed in a stainless steel cylinder with 0.5 cm thickness and two cubic decimeter in volume. The remaining space of the cylinder is filled with charcoal as an inert absorbing material whose function is only to retard the difusion of the radon gas

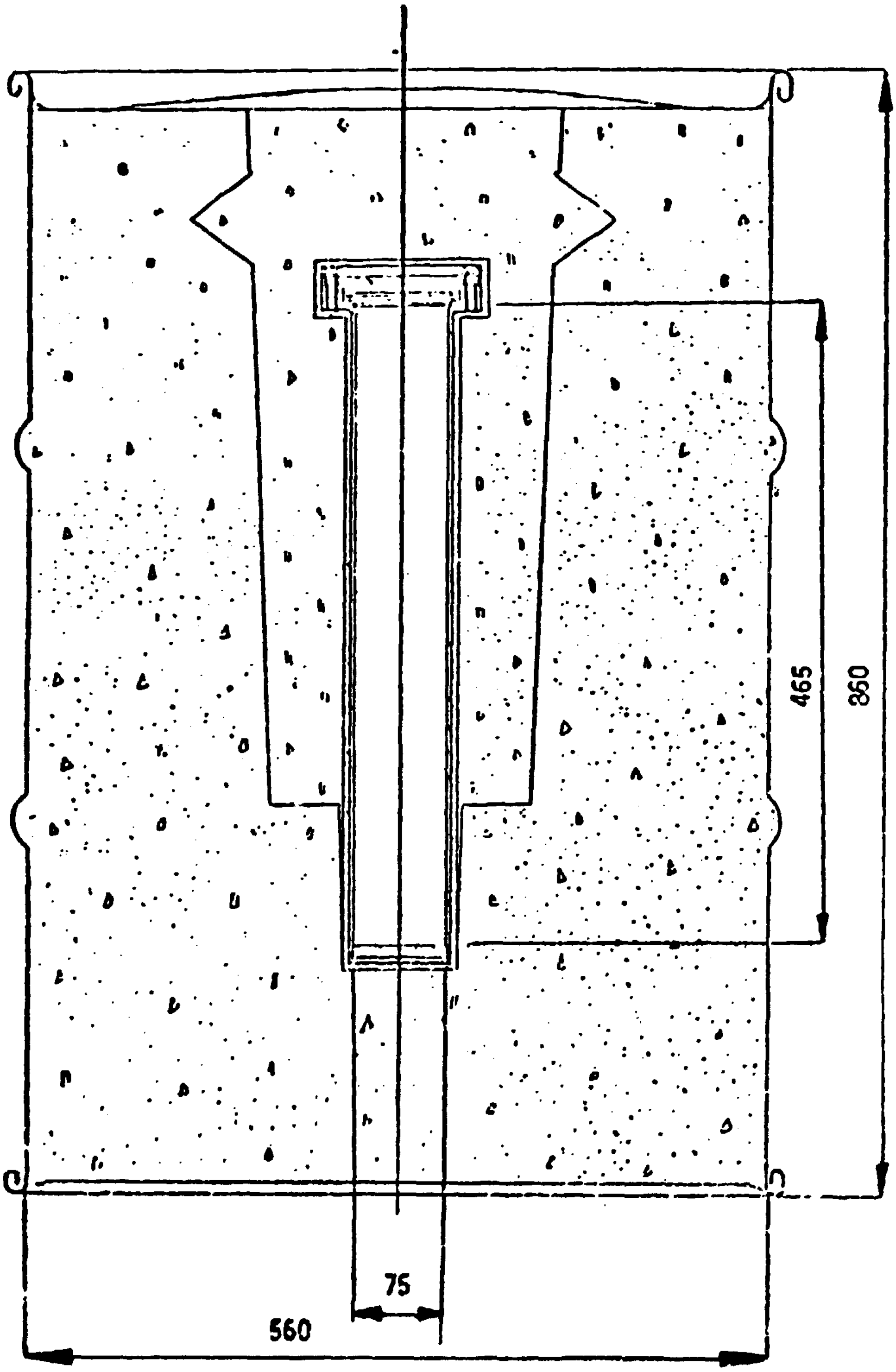

Figure 1 – Packaging to enclose radium sources
(dimentions in mm)

generated. The shielding materials are a 0.5 cm lead layer inside a heavy concrete shell 23 cm thick. The outer package is a commercial 200 liters drum.

It was intended that the package should be in a form acceptable in the final disposal site and also meet the requirements for type A packages described in the regulations for the safe transport of radioactive materials. According to that regulations the activity limit for non special form Ra 226 sources is 20 GBq (~ 0.5 Ci).

SHIELDING

Beyond the activity limitation per package the regulations limit the radiation level at any point on the external surface of the package to 2 mSv/h (200 mrem/h) and 0.1 mSv/h (10 mrem/h) at any point 1 m from the outer surface of the package.

To attend this determination the required thickness of all shielding materials was evaluated by using the computer code HGAMMA I(1) develloped for cylindrical geometries. The range of values for the parameters data used in that calculations are shown in table I and the obtained results for a concrete shielding density of 3.0 g/cc are in table II.

LEAKTIGHTNESS OF PACKAGES

The leak test used to verify the tightness of the stainless steel capsule was the helium spray probe technique. The equipment utilized was a VEECO-MS-17AB with a detection limit of 6×10^{-11} atm x cm^3/s and a sensibility of 5×10^{-9} mbar x ℓ/s. The detector background during all the measurements was less than 0.6×10^{-9} mbar x ℓ/s.

For all tested capsules the leakage rate was lower than 5×10^{-9} mbar x ℓ/s.

With this leakage rate value it is possible to evaluate the dimensions of a closed room eventually used as an interim storage in order to comply with the radiological protection rules and the limits of inhalation of radon by workers.

The radon leakage rate equivalent to the observed air leakage is 1.81×10^{-9} mbar x ℓ/s. Considering that the derived air concentration (DAC) for radon and daughters is 1.5×10^3 Bq/m^3(2) it is possible to demonstrate that for each radium source packaging is necessary to provide a free space of at least 5 m^3. For example this means that an interim storage room has to have at least 50 m^3 to accomodate 185 GBq (5 Ci) of radium sources without incurring in excessive doses for the workers due to radon leakage. Of ccurse any forced ventillation will decrease the space necessity in terms of radon concentration limits.

PACKAGE TEST PROCEDURE

In order to demonstrate the compliance with the performance standards required by the regulations it was prepared a prototype of the packing. It was submited to the free drop test as required for type A packages.

The integrity of containment system was verified using the same equipment utilized for testing the leaktightness of the capsules.

The shielding integrity as well as the containment integrity evaluations were both carried out before and after each individual test.

Table I

Parameter values used to evaluate the shielding of the packaging

Layer Material	Layer thickness (cm)	Density (g/cm^3)
Source	3.75	0.00125
Inox Capsule	0.5	7.8
Concrete	23.75	2.0; 2.4; 2.6; 3.0; 3.4
Lead	0.5; 1.0; 5.0	11.3
Drum	0.115	7.8

Table II

Activity limits for packaging according to shielding materials thickness

Shielding thickness (cm)		Maximum activity per package-type A	
Lead	Concrete	(GBq)	(Ci)
0	23,75	14.3	0.4
0.5	23.25	19.5	0.5
1.0	22.75	24.5	0.7
5.0	18.75	130.00	3.5

CONCLUSIONS

The described package satisfy requirements for transportation and interim storage and gather enough properties to fulfill final disposal sites criteria.

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2. INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION. Limits for inhalation of radon daughters by workers. Mar. 1981. (ICRP Publication 32).

DESIGN AND DEVELOPMENT OF A LIQUID WASTE STORAGE TANK

Roberto Vicente

ABSTRACT

The paper presents the work undertaken to develop the concept of an intermediate-level waste storage tank and the laboratory work aiming at knowing and improving the performance of the associated instrumentation. Results of tested liquid agitator, liquid level indication and alarm devices are presented.

1. INTRODUCTION

Liquid wastes are stored in tanks before treatment and immobilization. In many cases the storage time can extend over periods of months or even years. Wastes classified as intermediate-level wastes, may present some problems in the selection of materials and equipments that will be in direct contact with the liquid.

Corrosion of materials, and reliability of equipment must be carefully evaluated in the concept and in the detailed project of a storage tank because the access for maintenance is hindered by the radiation field. Also, safety related equipment must work in passive form ensuring a high general safety standard of operation.

In the design of a storage tank for ILLW some functions must be forseen: agitation of the liquid to keep the waste homogeneous and to prevent suspended or dissolved solid matter in the liquid to settle and incrustate in the tank walls; indication of the level of the tank content and high liquid level alarm; indication and alarm of liquid leakage; temperature indication and alarm; decontamination etc.

In this work we have defined arbitrarily a set of conceivable waste properties and based in these properties we have made a tank concept and undertook laboratory work to support the choice of ancillary equipment for this tank. We have searched for configurations that resulted in reliable equipment, with reproducible behavior, and at the same time that was simple and cheap.

2. CONCEPT OF THE TANK INSTALLATION

A tank installation was devised in which a tank is placed underground in an engineered vault to facilitate radiation shielding.

The liquid wastes are transferred by pipeline from the laboratory or laboratories where they are generated to the storage tank. These pipelines are double-walled to prevent any liquid leakage from the active pipeline which could cause contamination of soil, buildings etc. In the case of waste leakage, an alarm indicates the event.

The tank is surrounded by a safety jacket to collect any liquid leakage from the stored waste. Bellow the tank, a safety tray prevent water to intrude in the installation or waste to contaminate vault structure. The space above the liquid is vented to an exaust system with absolute filters. The liquid is permanently agitated. Devices for level indication and alarm as well temperature indication are foreseen. Decontamination of the tank after it is emptied is performed by spray nozzles. Welded edges in direct contact with the liquid are kept to a minimum to reduce corrosion. All tube conections are made in the top of the tank in a flanged lid.

It follows a more detailed description of some of the tank features.

2.1. Materials Selection

Low carbon austenitic stainless steel is the recomended material for those parts in contact or that could be in contact with the waste. We selected the AISI 304-L stainless steel to built all tank parts and associated equipment.

Allowance for corrosion over the tank life-time is 1 mm.

2.2. Agitation of the Liquid

Agitation of the waste is necessary to prevent solid matter suspended in the liquid to settle in the tank botton. The settling of suspended matter cause the formation of sludge layers in the tank walls (which could lead to increased localized corrosion) and inhomogeneities which make difficult to sample and retrieve the stored waste for treatment.

A ballast tank or pulsator or pulsation column was selected as agitation system for the proposed storage tank. This device has advantages over other agitation systems normally used, namely air spargers or air-lift circulators. These last two systems intensify the evaporation rate in the liquid and increase the formation of radioactive aerosols which overload the air filtration systems. The pulsator is a tube sunken in the liquid waste with a nozzle in the botton and a compressed-air connection in the top.

A pulse of compressed-air makes the liquid contained in the pulsation column to be ejected through the nozzle sweeping the tank botton and agitating the liquid. This keeps the solids suspended in the liquid.

The work was directed to design an efficient pulsation column and a reliable air pulse generator.

Pulsation Column

Aiming at get expertise in the design of a pulsation column some premisses were adopted and a tentative column was designed and the optimum operating condition were determined empirically.

The pulsation column designed is a tube, coaxial to the tank axis, with a convergent section in the lower part and ending in a nozzle close to the tank botton. It is shown in figure 1. The choice of a pulsation column as the agitation system determined the tank geometry. The conic botton of the tank was select to improve agitator performance.

A model of the tank was built for the laboratory tests which included:

a) determination of pulse duration as function of air pressure, nozzle-to-botton gap, air scape orifice diameter and liquid level;

b) determination of the lower pulse period as a function of the foregoing set of variables; and,

c) least conditions of pressure, nozzle gap and orifice diameter which keep a suspended solid tracer homogeneously distributed in the liquid, as a function of liquid volume.

The figures 2 and 3 show the curves that fit data from air pulse duration determinations and figure 4 shows the minimum pulse intervals. From these results it was plotted the curve which correlates air pulse pressures and liquid volume at a fixed pulse duration and the curve which correlates pulse frequencies and liquid volume (figures 5 through 7).

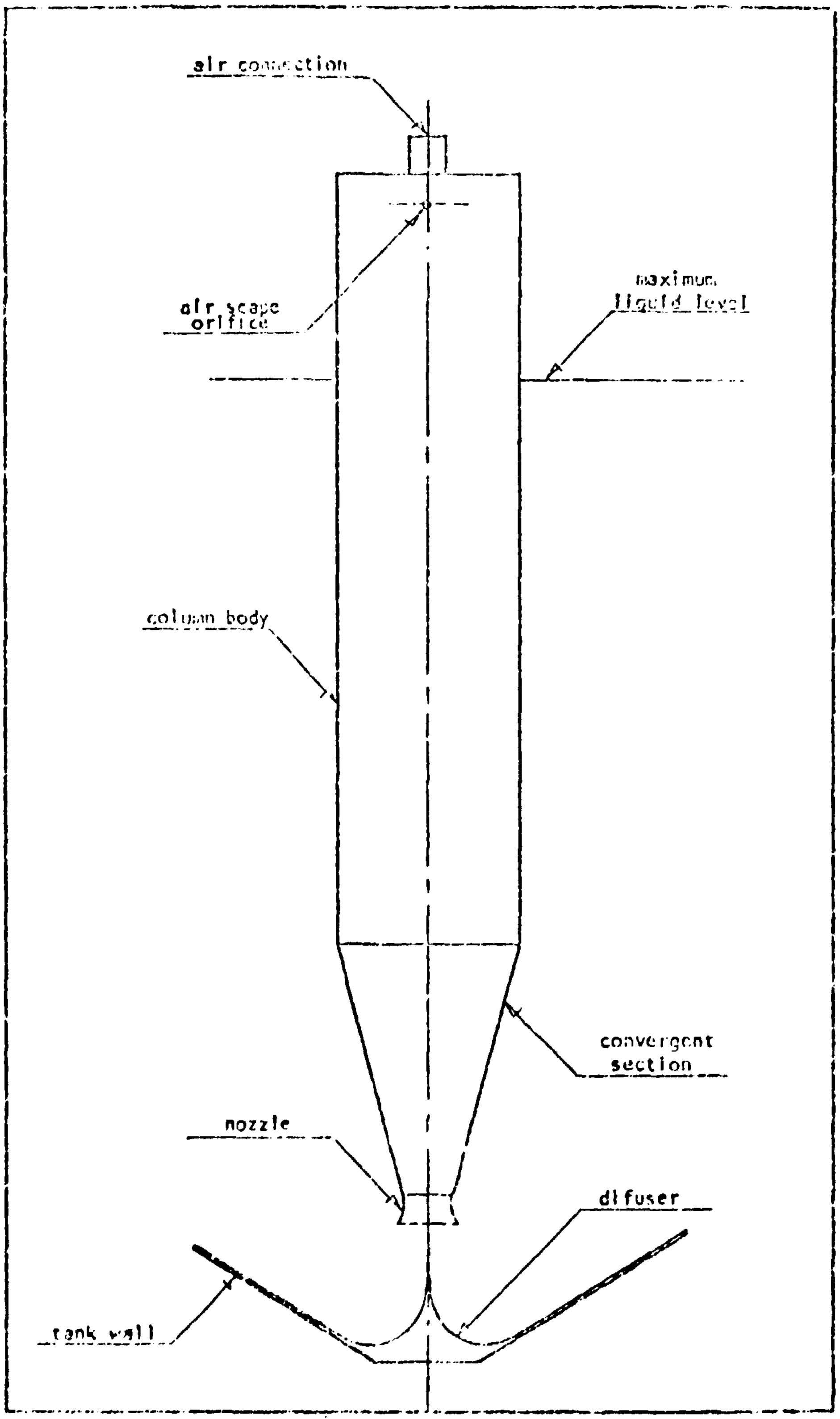


Figure 1 – Pulsation column

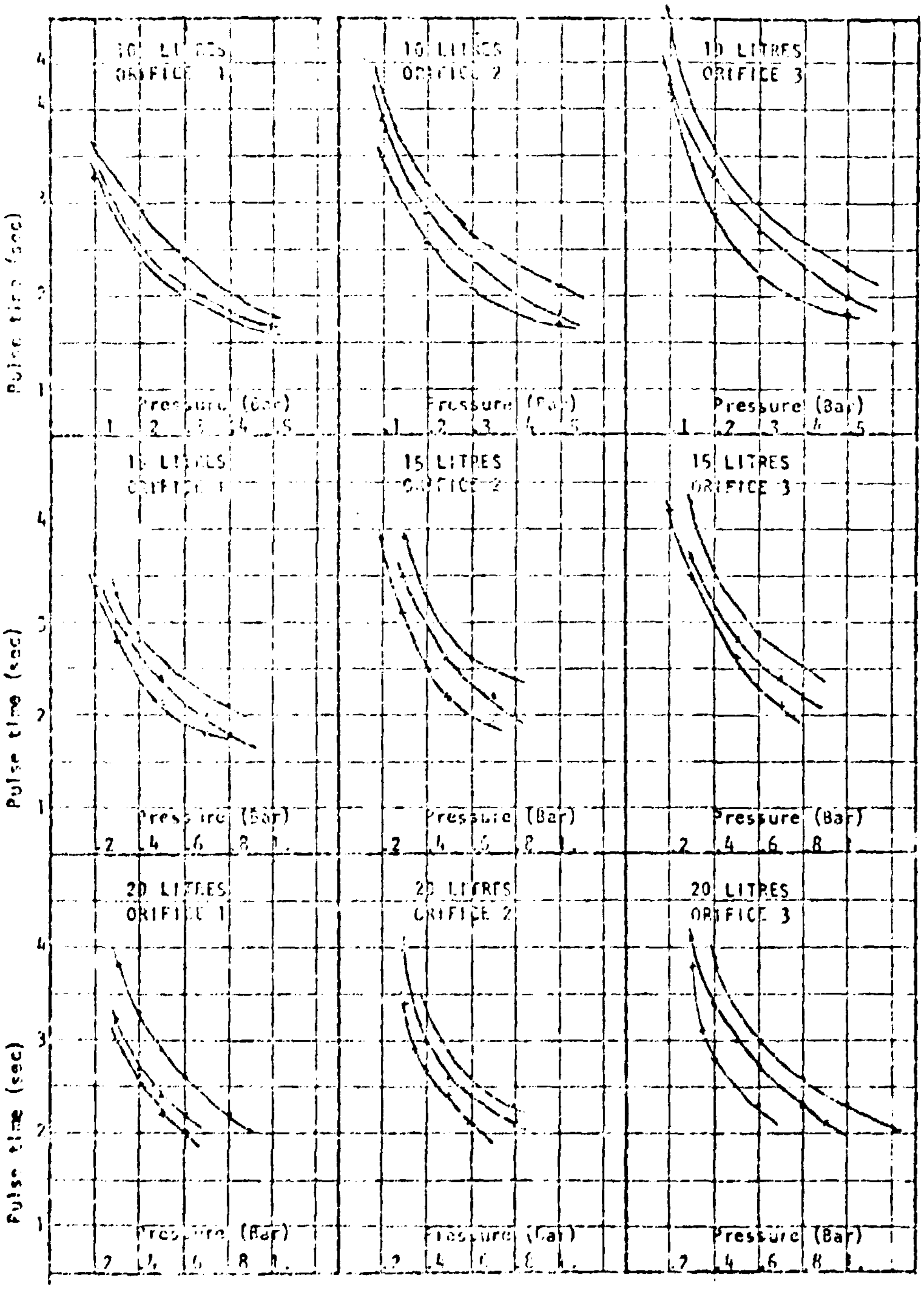


Figure 2 – Pulse time as a function of pulse pressure
(Key to symbols: 2 mm gap ●; 3 mm gap ▲; 5 mm gap +).

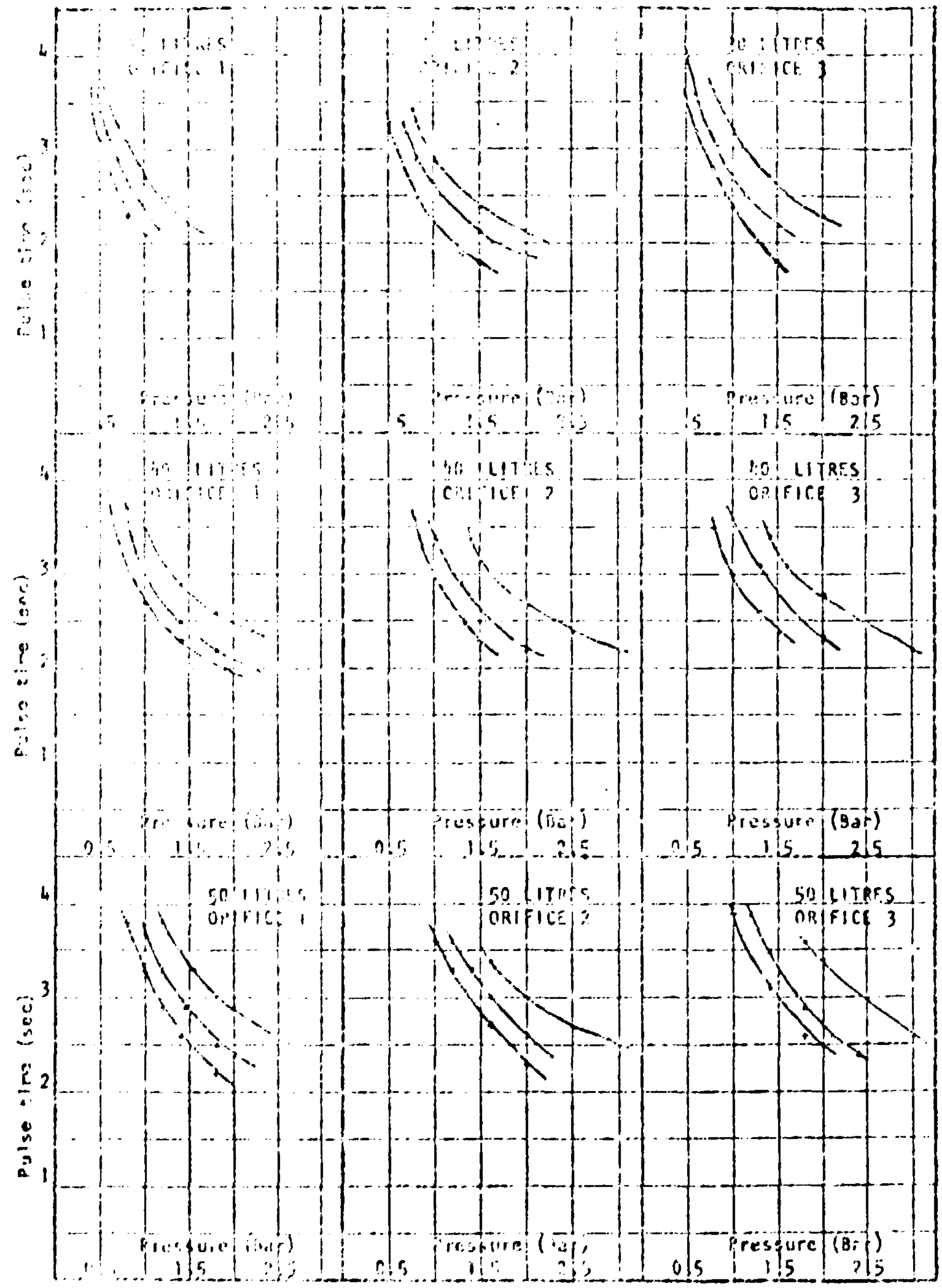


Figure 3 – Pulse time as a function of pulse pressure
(Key to symbols: 2 mm gap ●; 3 mm gap ▲; 5 mm gap +).

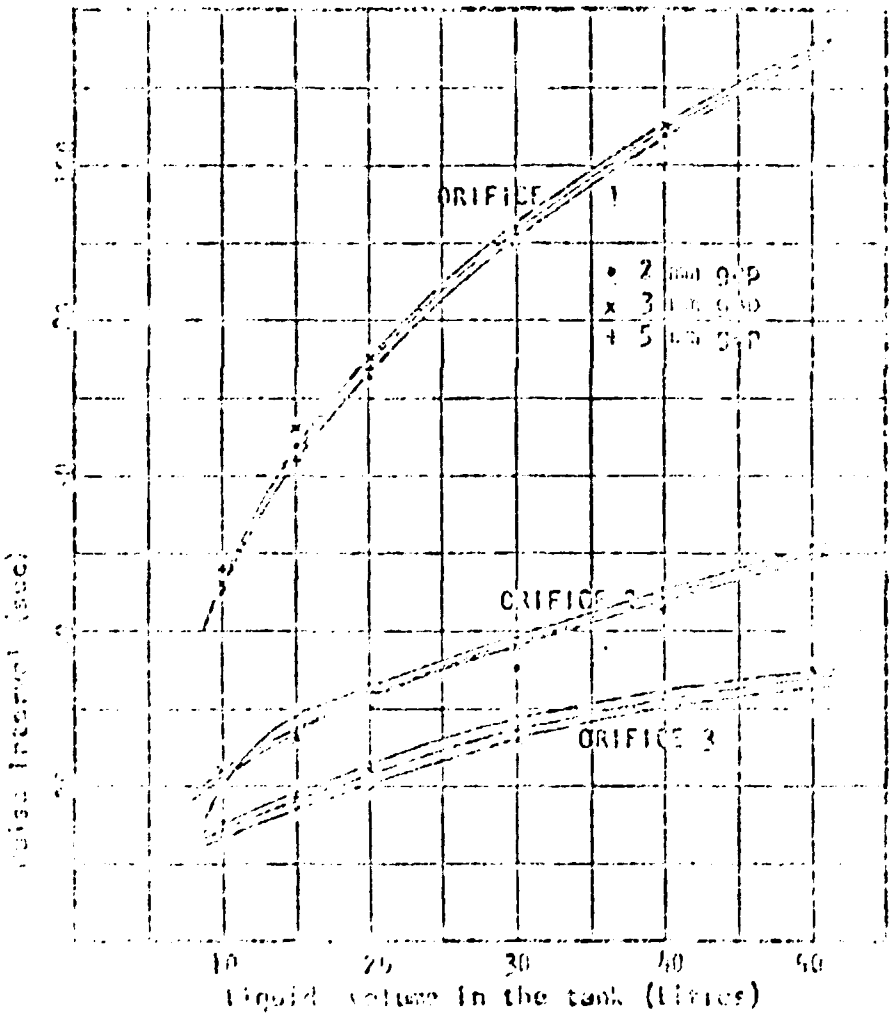


Figure 4 – Pulse intervals as a function of volume

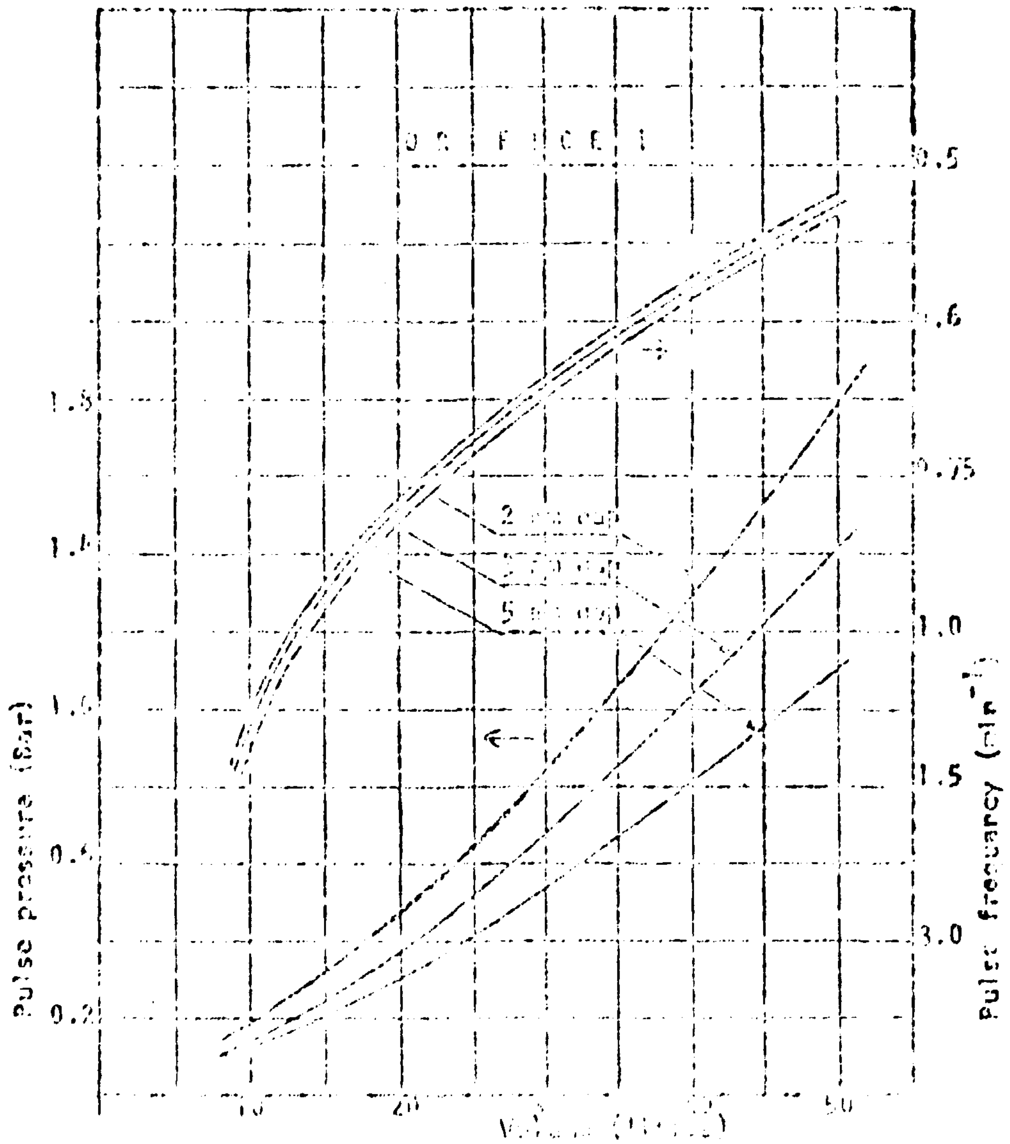


Figure 5 – Pulse pressure and pulse frequency as a function of volume of liquid in the tank (Pulse width = 3 sec)

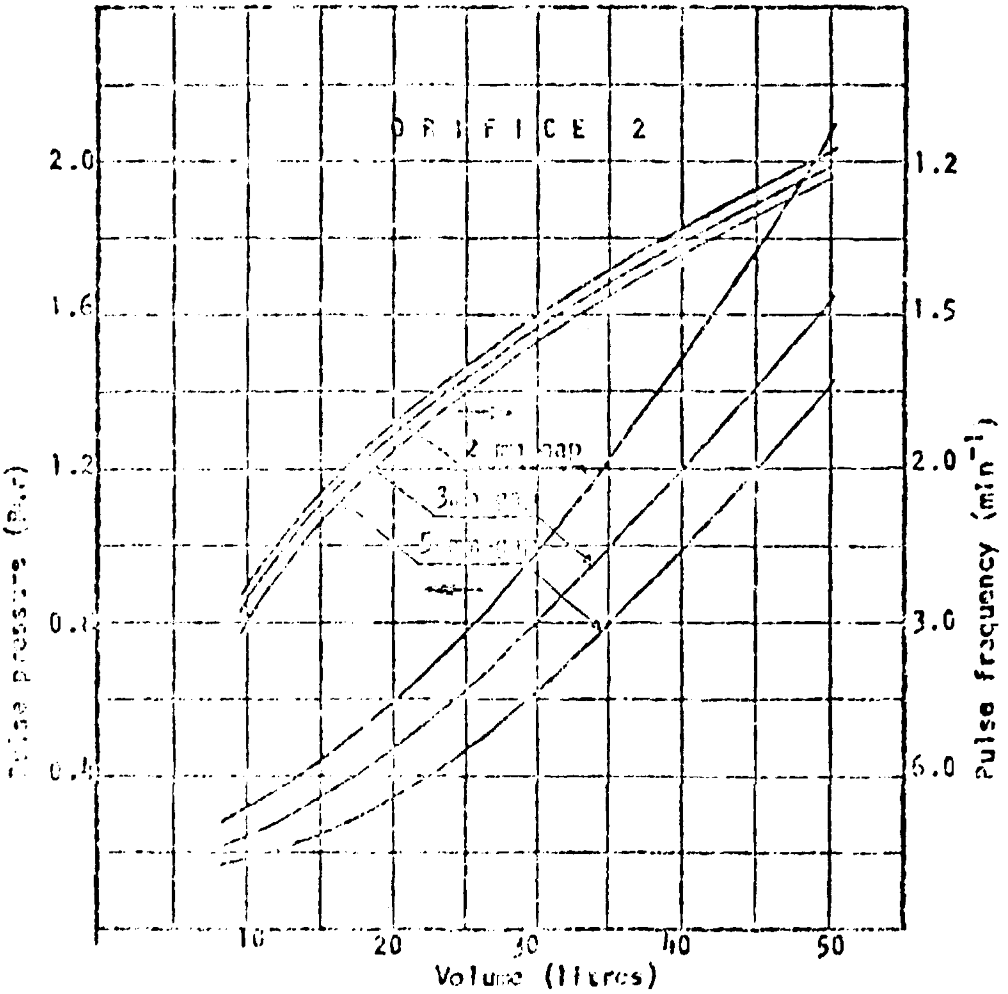


Figure 6 – Pulse pressure and pulse frequency as a function of volume of liquid in the tank (Pulse width = 3 sec)

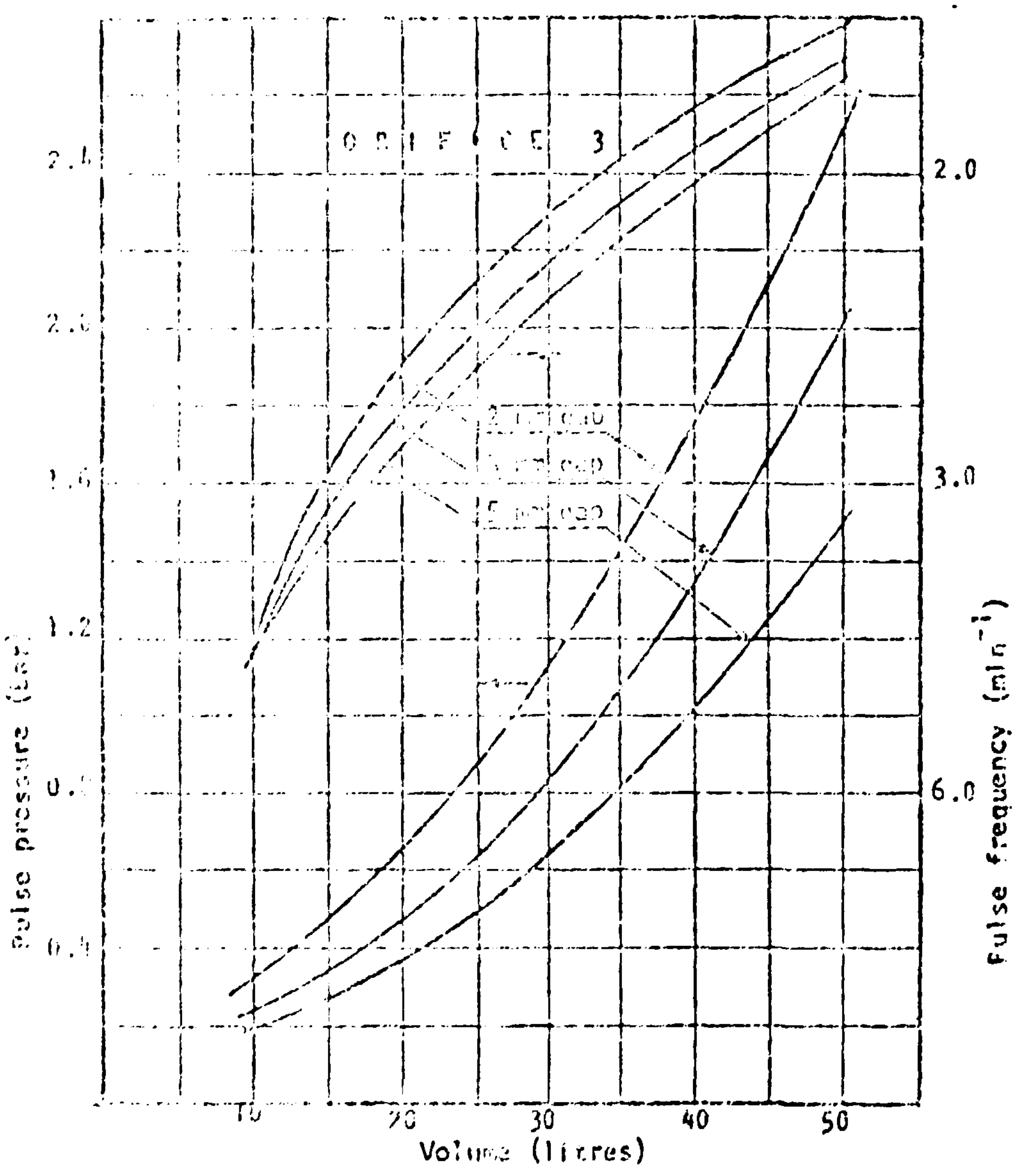


Figure 7 – Pulse pressure and pulse frequency as a function of volume of liquid in the tank (Pulse width = 3 sec)

These last three curves are used as rulers for control of the pulsation column.

To measure liquid homogeneity, samples will be withdrawn from three different heights in the liquid during agitation and the concentration of the tracer in each sample compared with the average concentration.

Air-Pulse Generator

The air-pulse generator must supply compressed-air to the pulsation column at fixed time intervals, with preset air pressure and volume. We have decided to use a pneumatic timer to control the air-pulse generator. Among many tested configurations, that shown in figure 8 presented the best results with regard to reproducibility, reliability and control sensitivity. The tests made with the candidate air-pulse generators were: changes in pulse duration and pulse interval after hundred thousands accumulated pulses, response to changes in control knobs and sensitivity.

2.3. Liquid Level Indication

The use of dip tube to measure liquid level in waste tanks is widespread because it is reliable, has no moving parts, do not depend on electronic or electrical equipments to be installed in the tank and is simple and cheap.

A differential manometer indicates pressure drop between liquid surface and the tip of the dip tube sunken in the liquid near the tank botton. As the density of the liquid influence the readings in the manometer and the liquid density can vary over time and for different wastes an indication of the liquid density is necessary to correct the level measurements. Another dip tube is so installed at a different and accurately known height and the pressure drop between the tips of the two dip tubes gives the density of the liquid through the formula $\Delta p = \rho \times g \times h$ where Δp is the measured drop, g is the local acceleration of gravity, h is the height difference between the two dip tubes and ρ is the desired density value. In figure 9 the level indication system is schematically shown.

In the present case, one pair of dip tubes is enough to measure the liquid level and density with acceptable accuracy.

The laboratory work shows that the density can be determined to an accuracy of ± 0,05g/cm^3 and that the liquid level can be determined to an accuracy of ± 0,5 mm, when the pressure drops are read with a precision of 1 mm and when the height difference of the dip tubes are known with an accuracy of 1 mm.

2.4. High-Level Alarm for the Tank

Although the level indication instrument warns high level conditions it is advisable to forsee a device which alarms by means of buzzers and light signals those conditions.

We choosed a pneumatic-operated system based on the principle of dip tube. In this case the pressure drop in the dip tube is converted to a pneumatic signal that is amplified and converted to optical and sound signals by appropriate transducers.

We have designed the instrument shown in figure 10 and tested it as regard to its repetitiousness and to its sensibility to decreasing liquid heads. Results show that the instrument is reliable and is sensitive to at least 2 mbar (20 mm water gauge).

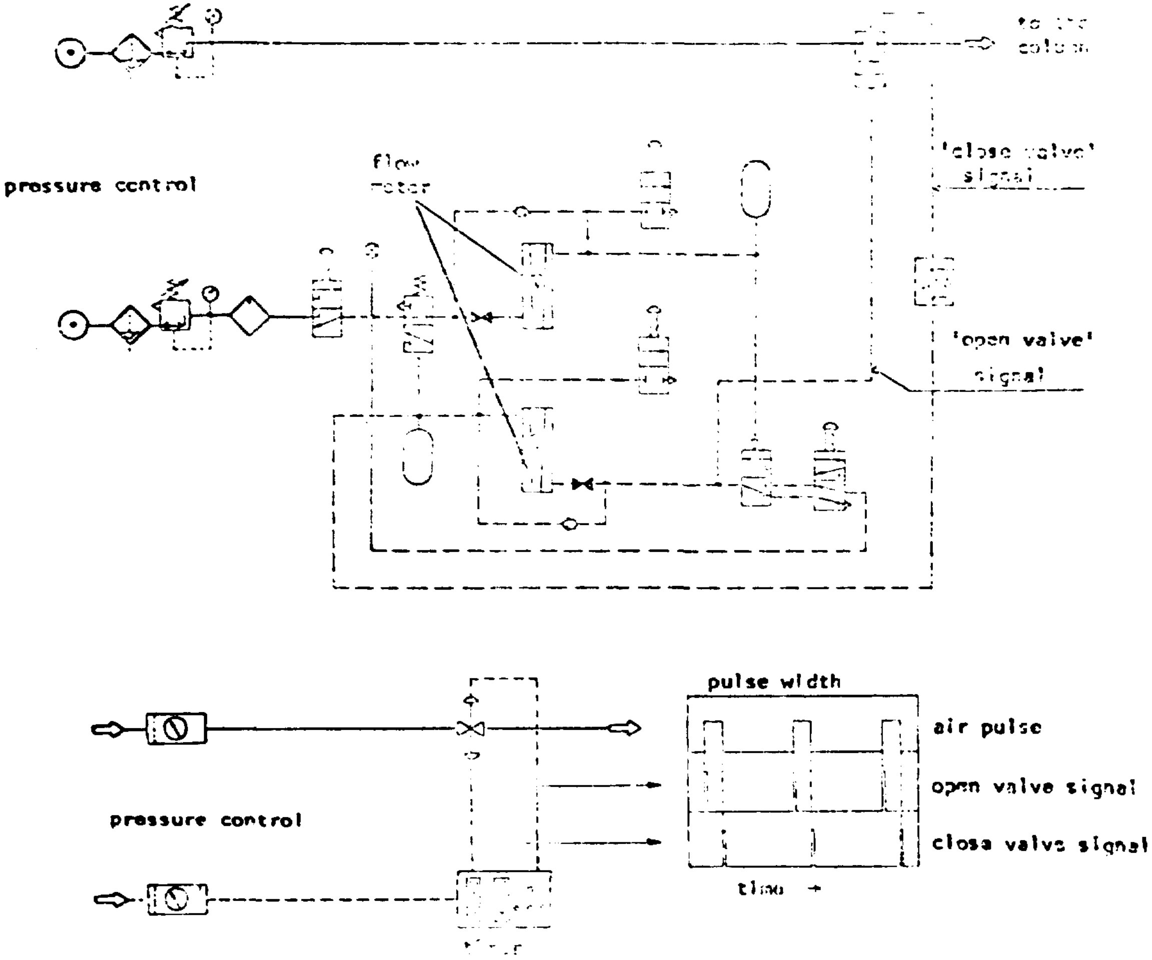


Figure 8 — Pneumatic - control timer (air pulse generator)

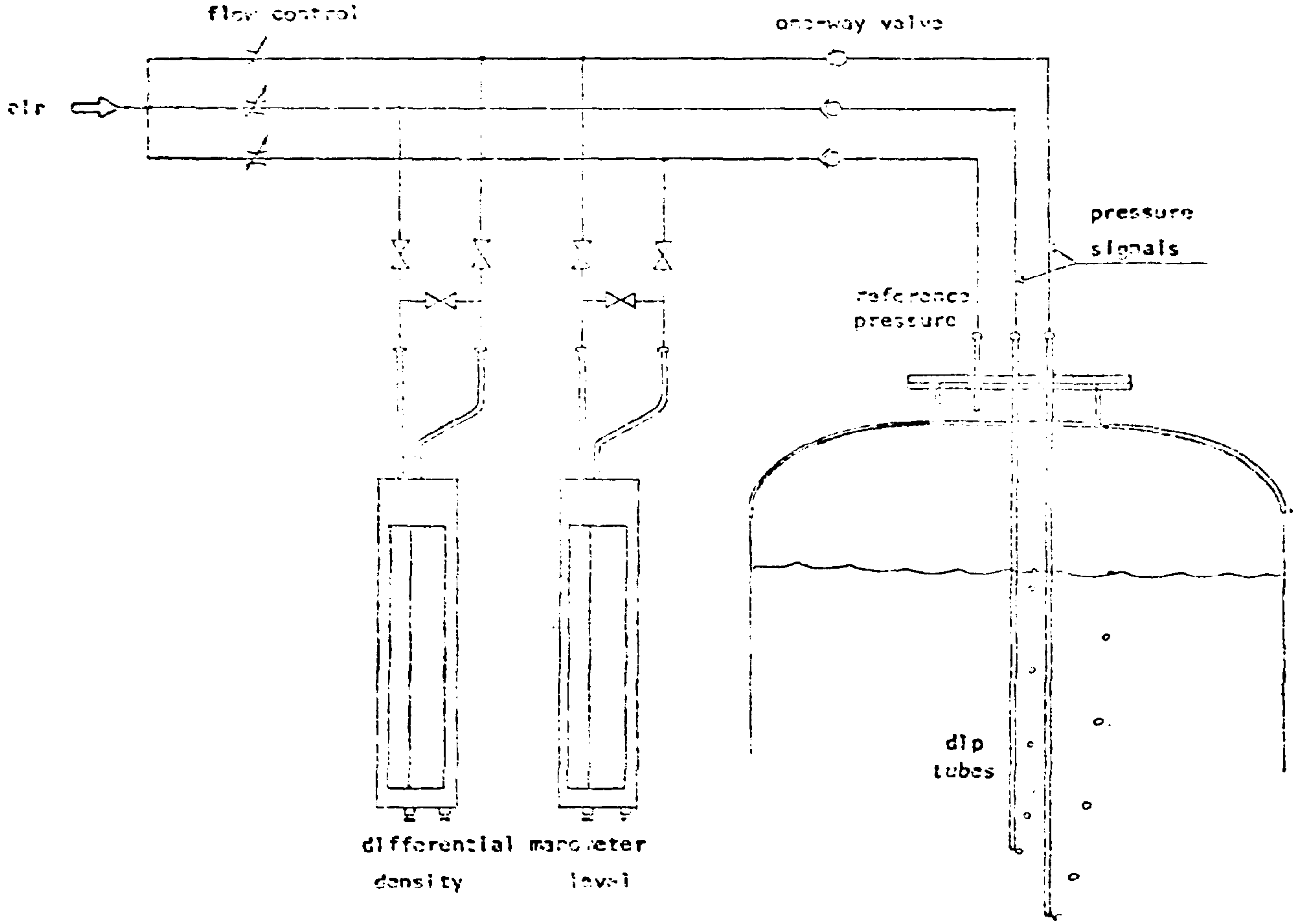


Figure 9 – Liquid level indicating system.

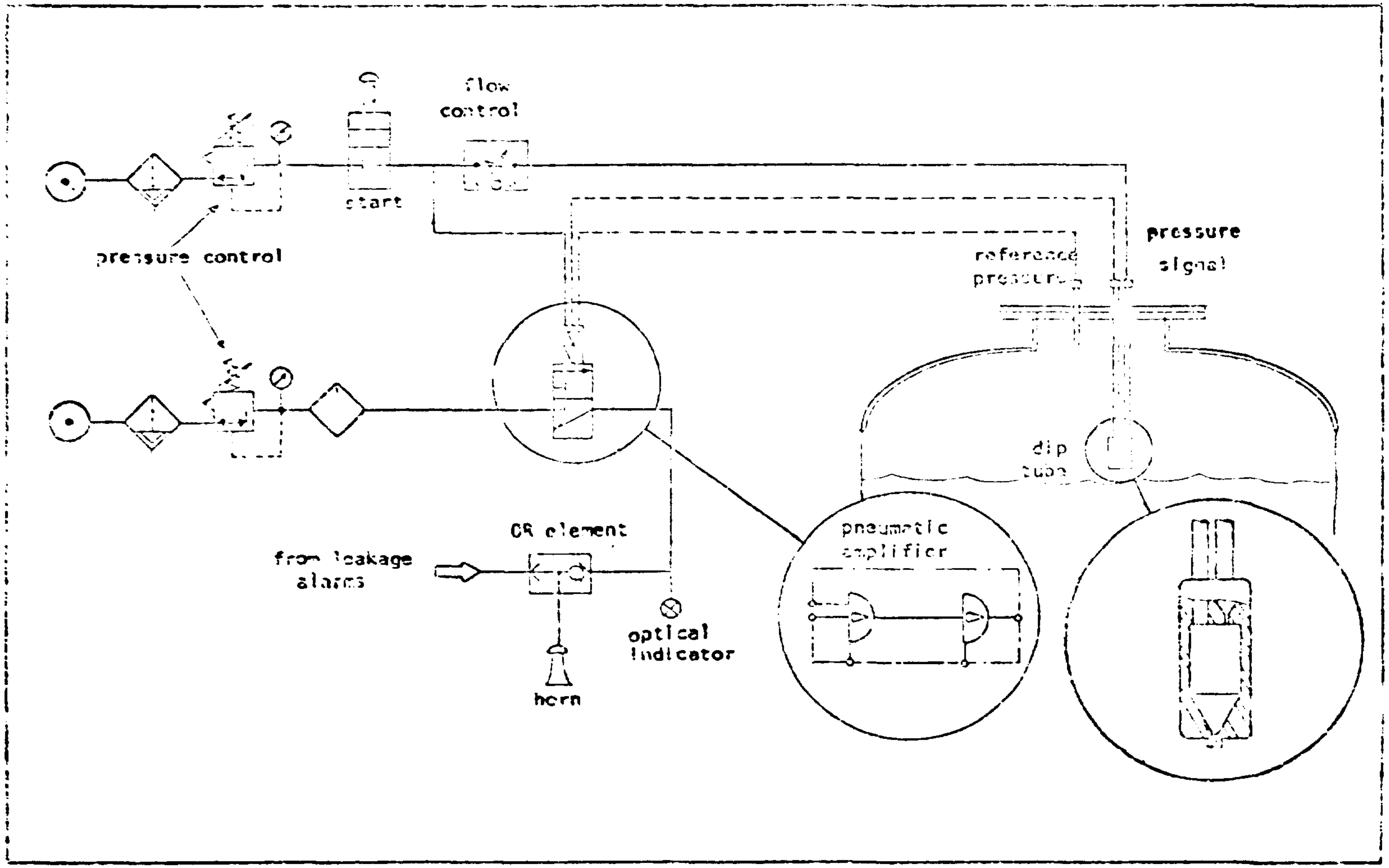


Figure 10 — High liquid level alarm - schematic diagram.

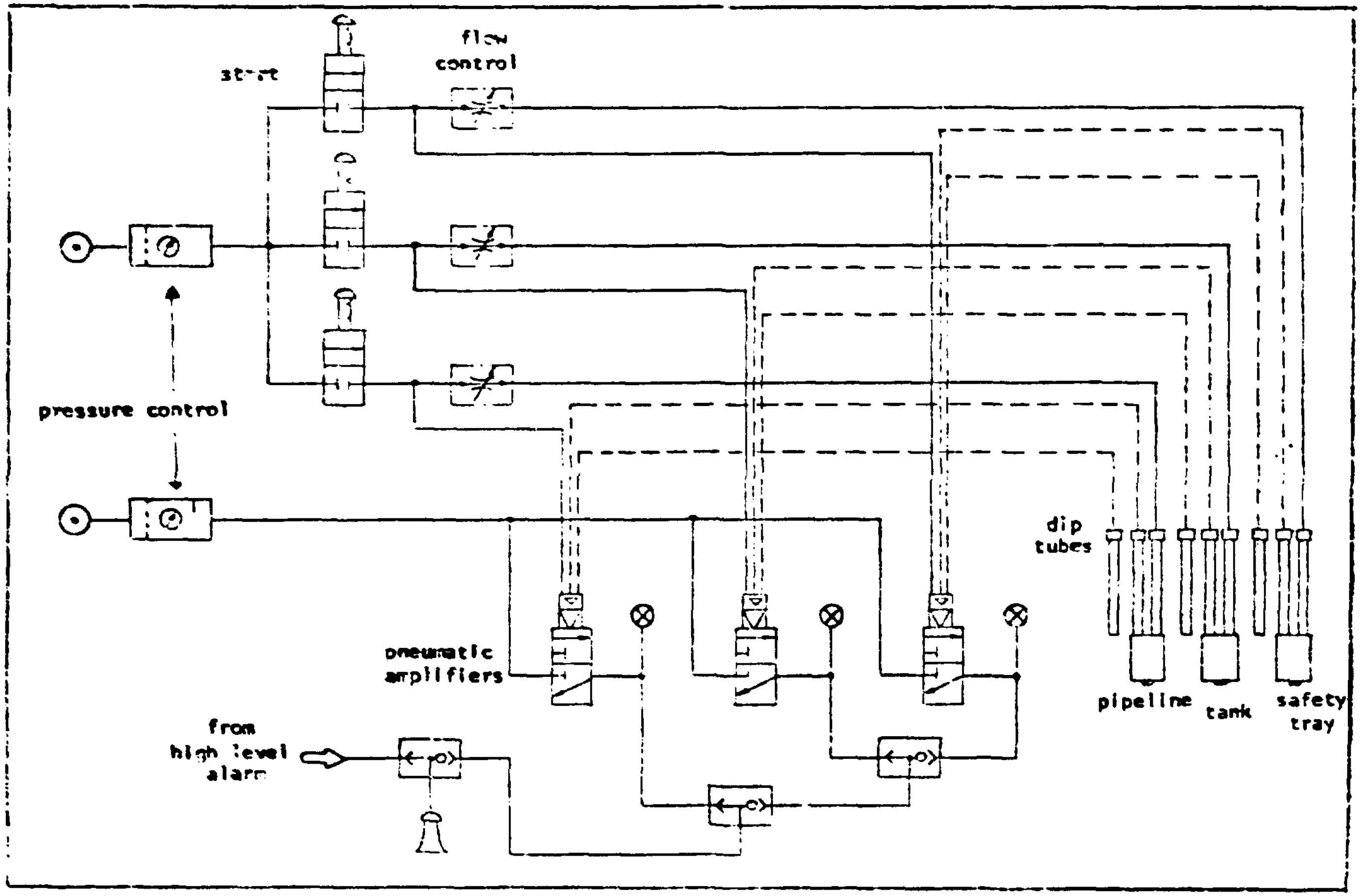


Figure 11 – Schematic diagrams for the liquid leakage alarms.

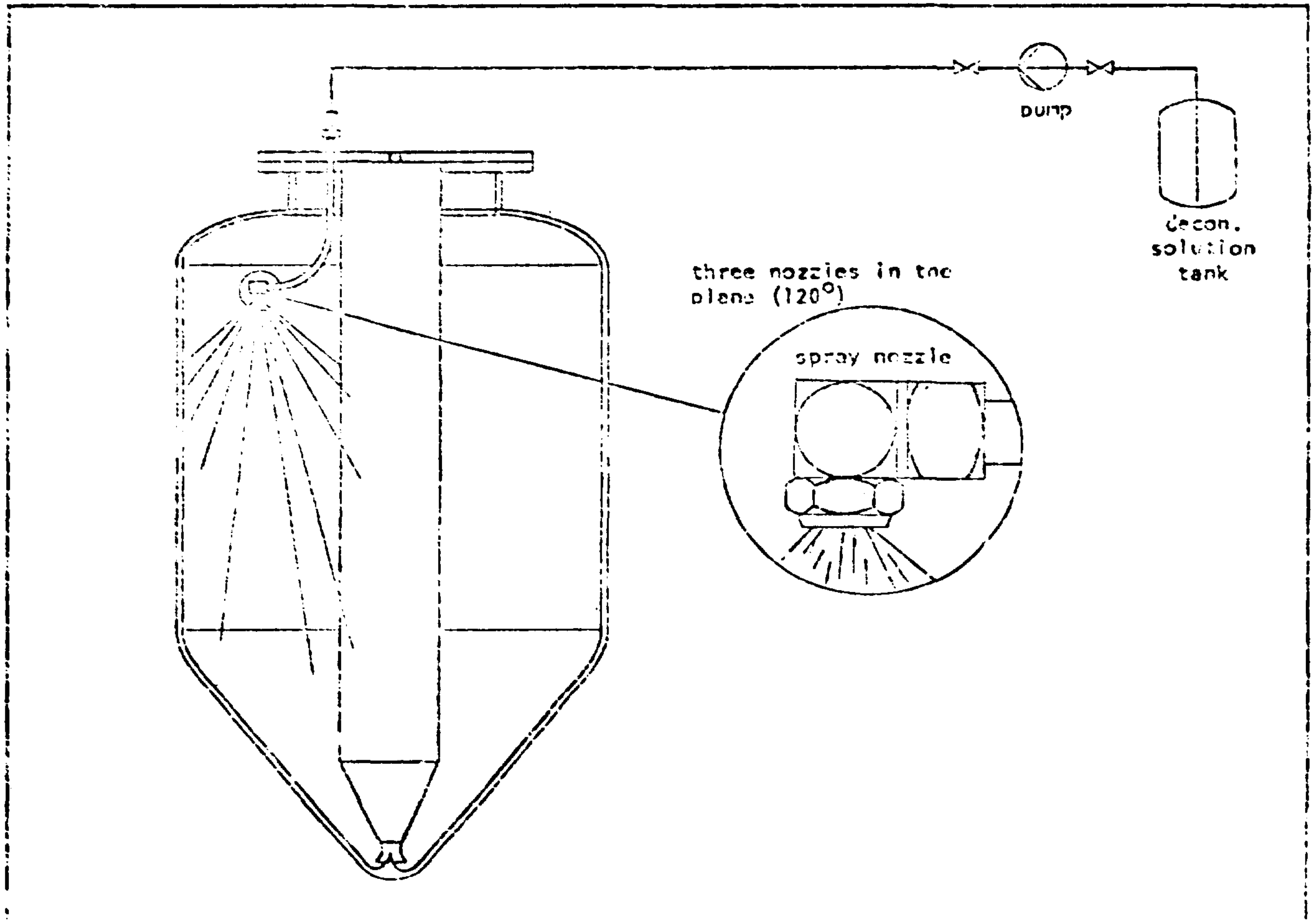


Figure 12 – Tank decontamination system.

2.5. Leakage Alarm

The leakage alarms to be installed in the pipeline, in the tank safety jacket and in the vault safety tray are all besed in the same instrument used for high level alarm. The three devices are shown in figure 11.

2.6. Tank Decontamination

When maintenance or replacement of tank parts are necessary the waste must be transferred to another tank or handling facility and the tank must be decontaminated to an extent that allows personnel to intervene in the tank vault. The decontamination solution must be selected according to the chemical nature of the waste and to the likely settled material which remains in tank walls. In any case the installation of spray nozzles aids in removing residual active material.

We selected the spray nozzles shown in figure 12 for the tank model. Three spray nozzles in a plane 100 mm above maximum liquid level height in a ring 250 mm diameter could rinse all surfaces.

3. CONCLUSION

Laboratory work undertaken served to give clues to design waste storage tanks and to demonstrate to some extent the viability of chosen instrumentation.

As a general conclusion, we can state that tanks designed in compliance with the specifications of this work can be operated safely and reliably for intermediate level wastes, even if it is corrosive and has solids in suspension.

Any explosive gases generated by radiolysis or explosive vapours arising from organic solvents eventually present in the waste are diluted to safe concentrations in the exaust duct by the air stream from the pulsation column and from built in instruments.

DEVELOPMENT AND DESIGN OF A CEMENTATION PROCESS

Roberto Vicente

ABSTRACT

The conceptual design of a facility for the immobilization of intermediate-level liquid wastes in cement is presented. The cementation process adopted a vibration assisted mixing process. The solidified waste is packed in 200 litres drum with barite concrete lining. The waste package is classified as Type A package for transport.

INTRODUCTION

Cement has been used as matrix for immobilization of low-and intermediate-level wastes since early times of nuclear industry, and although other materials became available, safety, low cost and simplicity of cementation processes keep up this matrix widespread used. The performance of cement-based waste forms is being improved in consequence of research aiming at improving materials (alternative types of cement, aditives etc.) and cementation processes.

We are engaged in such a kind of work: that of developing waste matrices and process equipment to immobilize low-and intermediate-level liquid wastes and that of developing techniques for quality assurance program.

In this paper it is presented part of the work carried out to develop the conceptual design of a facility for immobilization and encapsulation of low-and intermediate-level liquid wastes, and the laboratory work carried out to develop a cementation process for that facility.

The work done can be divided in three steps in order of precedence: definition of waste container and package; choice of the cementation process; and lay-out of the facility. This sequence represents to a some extent the steps which were followed in approaching the final goal of conceptually designing the facility.

Selection of Immobilization Container

A 200 liters drum was adopted as outer package for the immobilized waste. The waste package should complies with the especifications for a Type A package, non special form of the regulations for the safe transport of radioactive materials, and would be, preferably self shielded. For such type of packages the limits of content activities are very stringent. The consequences of such limitation is that, for intermediate-level wastes with, for example, 3.7×10^{10} Bq/litre (1 Ci/litre) the volume of liquid waste which can be immobilized in the drum can be as low as a few litres, if the waste contain certain radionuclides with low limits of allowable activity. From this it follows that immobilization containers can be as small as a few litres.

Two commercially available cans were selected as candidate immobilization containers for the laboratory work aiming at developing a process for liquid waste cementation: 3.4 litres (1 gallon) and 20 litres. With such containers and with a barite concrete lining in the drum, the radiation field in the package surface and the activity content limitations can be both fulfilled.

The proposed containers and package are shown in figure 1.

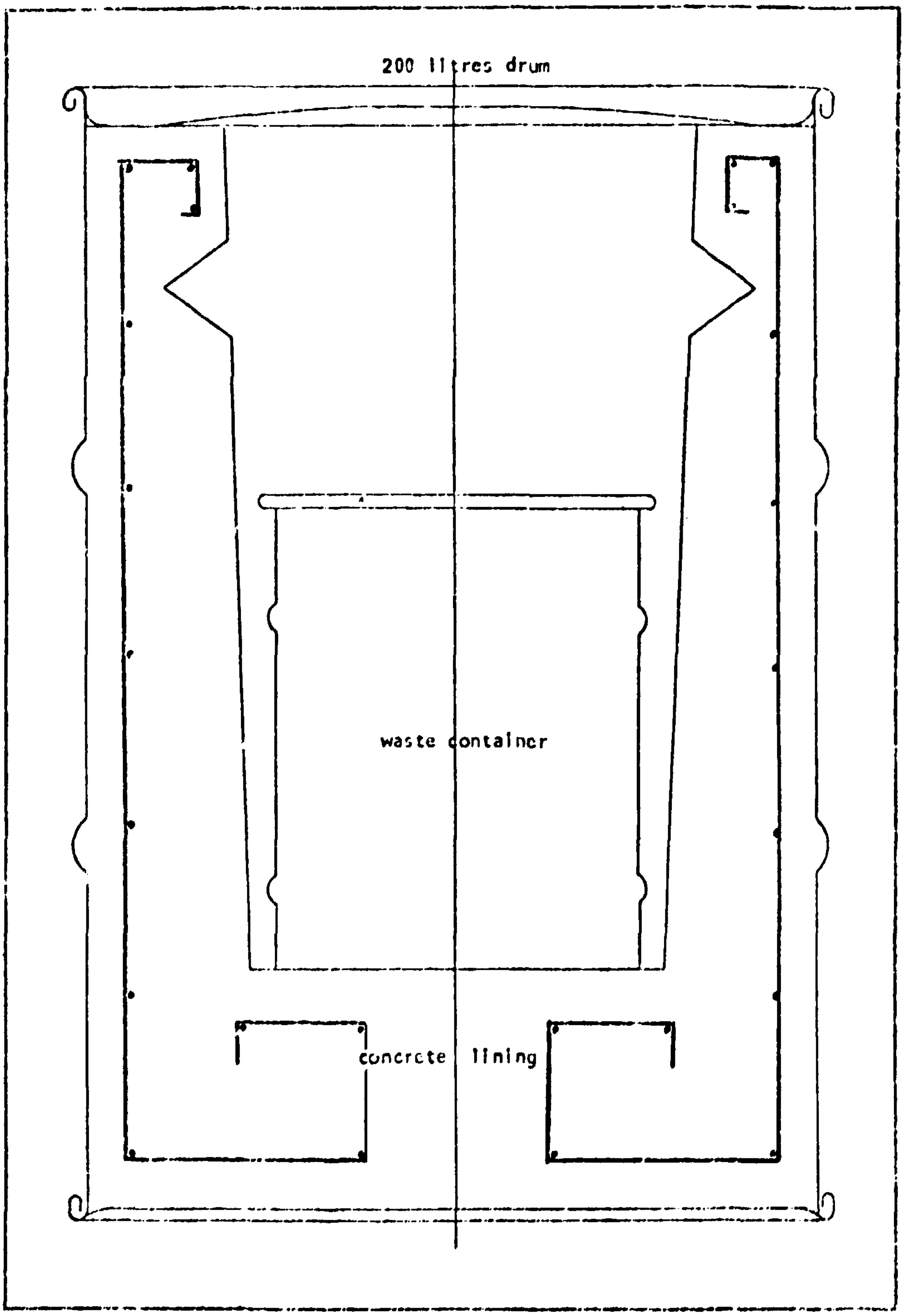


Figure 1 – Container and package for the immobilized waste

Selection of Cementation Process

Having defined the immobilization container, we searched for a process that took advantage of their small size. Another premise was adopted in this step of the work: the waste volumes to be treated would be tens or a few hundred litres and so the selected process would be batchwise. The candidate processes were: paddle mixer with disposable blades, paddle mixer with removable blades, roller mixers and vibration mixers. After some trials, a vibration mixer was chosen for further improvements. It was selected because it shows some advantages over the alternative processes: cleanliness, efficiency and simplicity of equipment.

The vibration mixing, actually a vibration assisted mixing process, consists in to place the container with the waste and cement in a vibration table and allow the two phases to mix by effect of a slow rotation of the container about its own axis in a slanting position. The vibration lowers the viscosity of the mix, accelerating the process of homogenization. One or two blades made of metal sheet fixed to the can lid further improve the mixing. Phase separation is avoided by appropriate water/cement ratios and by the rotation of the container. The schematic drawing of the apparatus used in the laboratory test is shown in figure 2.

At the beginning of the tests it was noticed that some of the cement blocks obtained by this process showed regions of little hydration and a markedly high air bubbles concentration all over the specimen. These results indicated that air was entrapped in the cement powder during the feeding of the waste. It was found that the more the compactness of the cement powder before waste feeding the greater those deleterious effects. Thus, it was developed a method of conditioning of the cement so as to improve the process. The better results arose when the container filled with the cement was quickly turned back and forth about its axis. Feeding the liquid waste in the botton of the can through a pipe aided in achieving better results.

The whole process is shown in figure 3. Schematic drawings of the model used in the laboratory to perform the aeration is shown in figure 4. Those models are the starting design for a more detailed project that could be built for active operation. (figure 2 and 4).

Conceptual Design of the Cementation Facility

The objective of the facility is to immobilize intermediate-level wastes and pack them for transport to final disposal sites. The designed facility has the following characteristics:

- the cementation is accomplished by a vibration assisted mixing process;
- the chemical adjustment of the waste before immobilization is realized in batches;
- active lines, vessels and parts of the system that can become contaminated by contact with the waste are placed in a hot cell;
- the system operation is manually controled by means of a control panel and master slave manipulators.

The cementation process diagram is shown in figure 5.

In figure 6 a lay-out of the facility with relevant process and control components is presented.

Four alternatives have been compared as regard to cost and to operational aspects. Two different shielding materials, concrete and lead, were considered; two hot cell concepts examined. The use of concrete has a positive impact on the cost but negative one if constructive, operational and decommissioning aspects are looked over. Negative aspects of use concrete arises mainly from the thicker walls needed.

The two concepts of hot cell examined are schematically shown in figure 7.

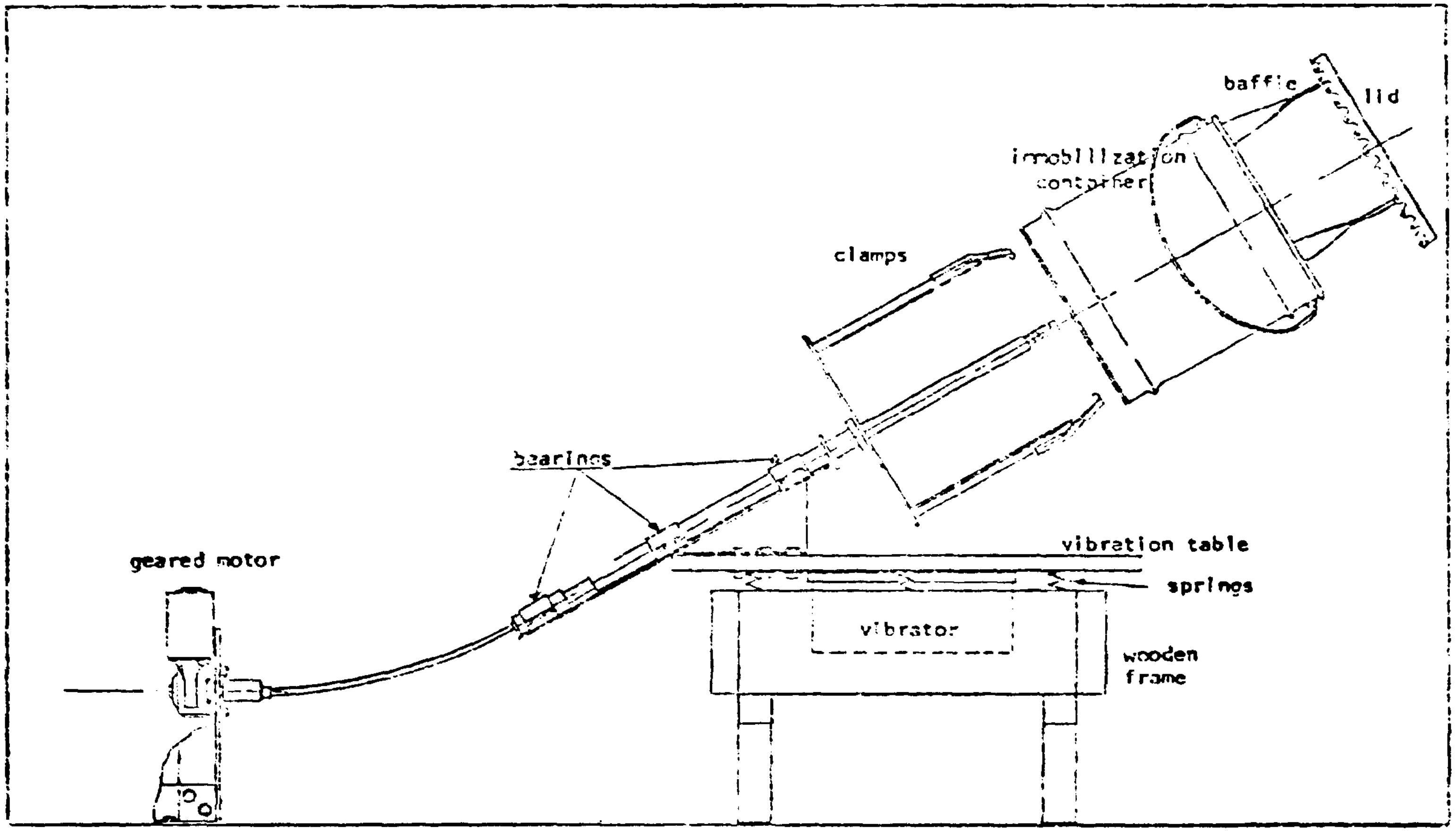


Figure 2 – Schematic drawing of vibration apparatus.

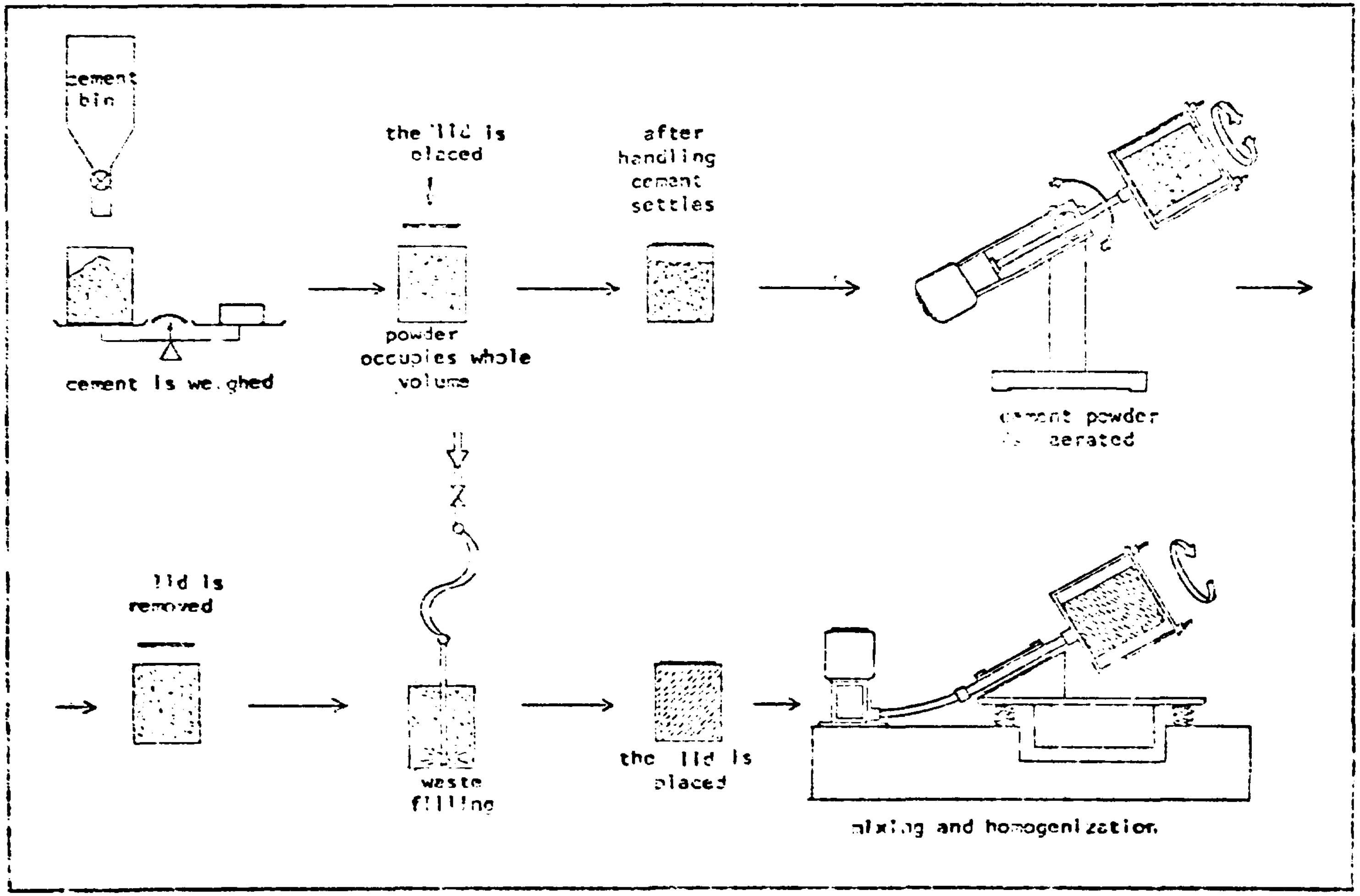


Figure 3 – Cementation process.

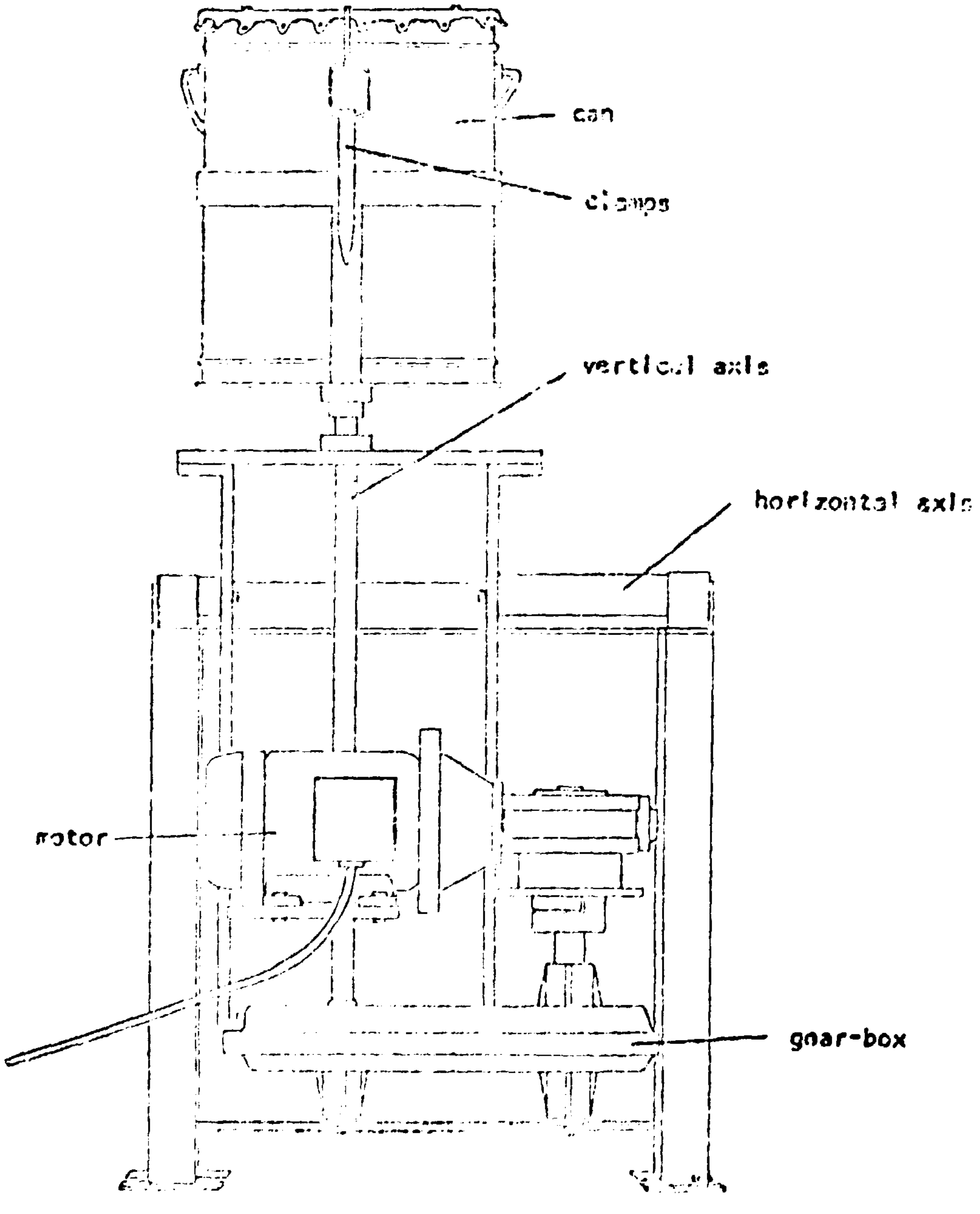


Figure 4 – Schematic drawing of the aeration apparatus

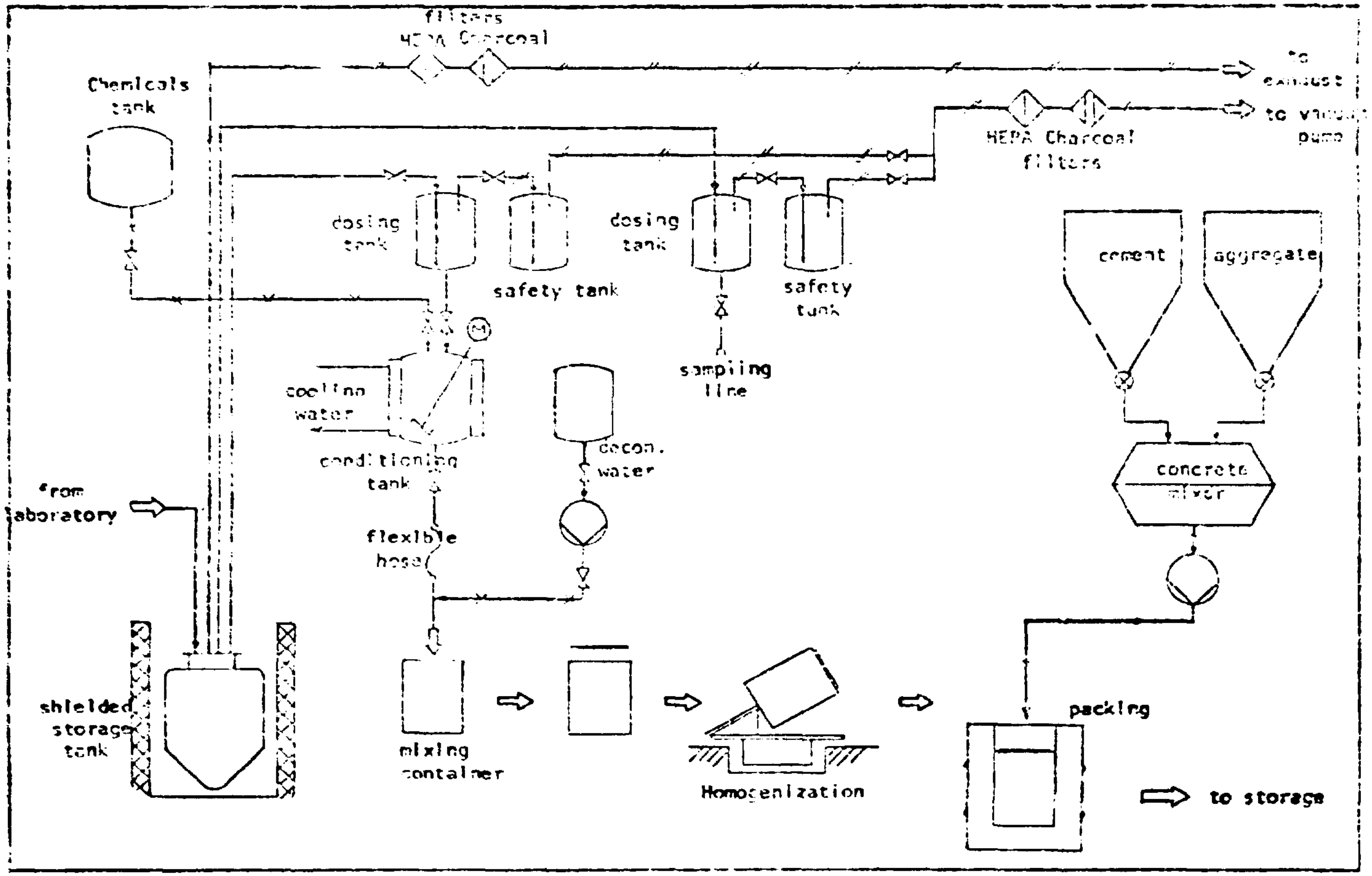


Figure 5 – Cementation process flow diagram

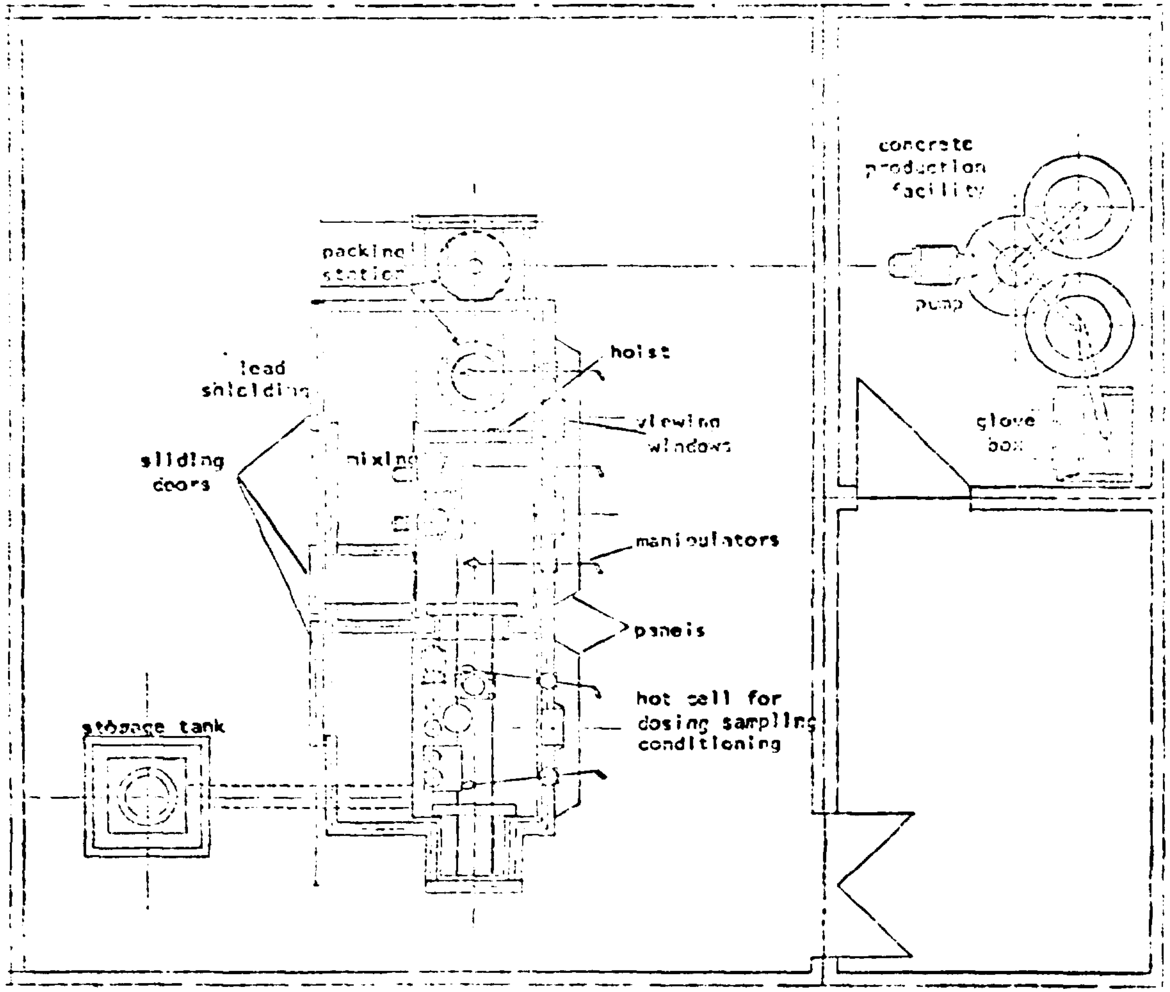


Figure 6 – Cementation facility lay-out.

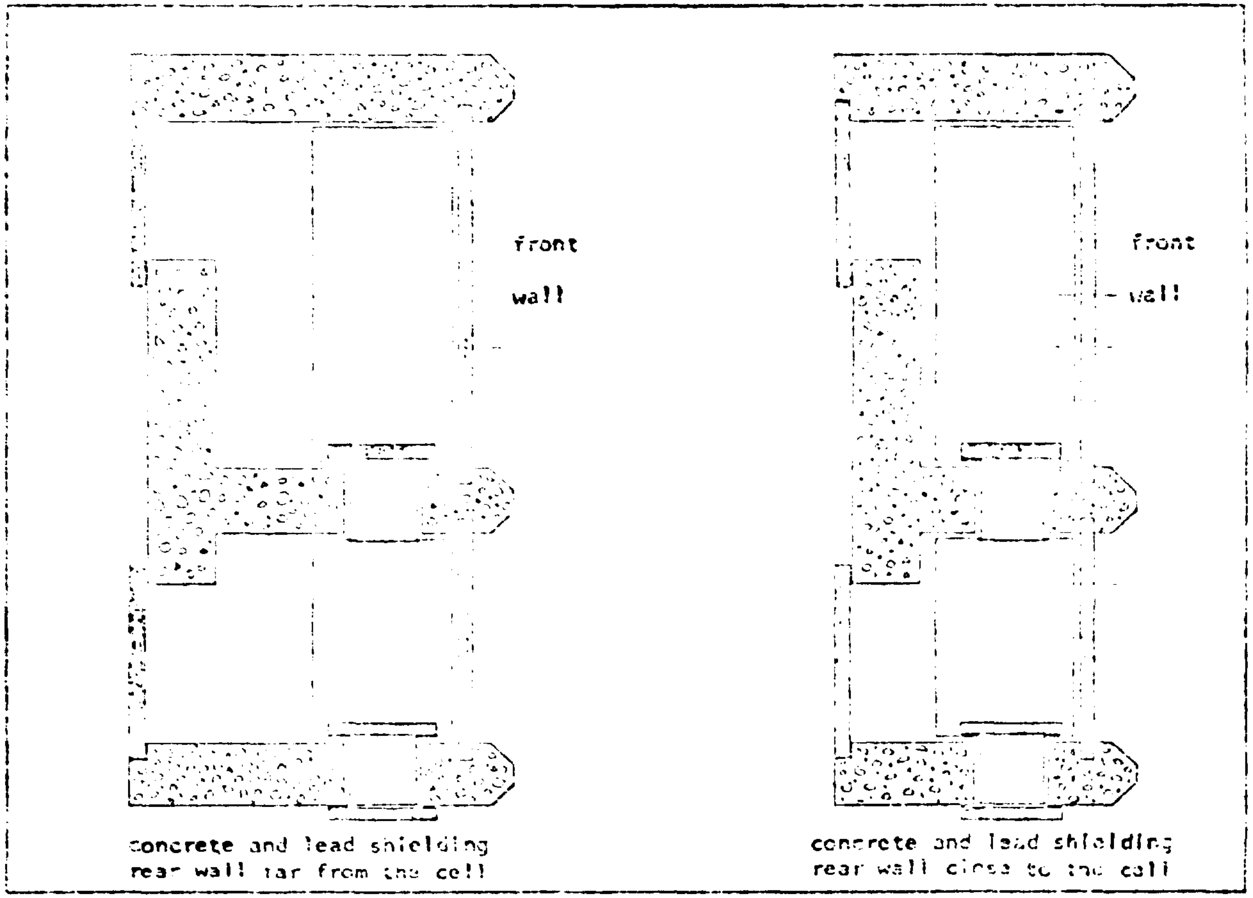


Figure 7 – The two concepts of hot cell examined: rear wall close to and far from the cell

METHOD FOR QUALIFICATION OF CEMENTATION PROCESSES AND ITS APPLICATION TO A VIBRATION MIXER

R. Vicente, B.M.Rzyski, A.A. Suarez

ABSTRACT

In this paper the definition of homogeneity is discussed and methods to measure the "degree of heterogeneity" of waste forms are proposed. These measurements are important as aids for mixing process qualification, and as tools in quality assurance procedures and in the development of waste management standards.

Homogeneity is a basic quality requirement for waste forms to be accepted in final disposal sites. It do not depend on the matrix of immobilization, rather it is one mean to qualification of the immobilization process.

The proposed methods were applied to a vibration assisted mixing process and has proved to be an useful mean to judge process improvements.

There are many conceivable methods to evaluate homogeneity of waste forms. Some were selected as screening tests aiming at quickly reaching a promising set of process variables. Others were selected to evaluate the degree of excellence of the process in respect to product quality. These envisaged methods were: visual inspection, the use of cement dye as tracer, scanning of radioactive tracers, and measurements of variations of density, water absorption, porosity and mechanical strength across the waste form sample.

The process variables were: waste-cement and water-cement ratios, mixer geometry, mixing time and vibration intensity. Some of the apparatus details were changed during the experimental work in order to improve product quality.

Experimental methods and results were statistically analysed and compared with data obtained from samples prepared with a planetary paddle mixer, which were adopted as the homogeneity standard.

INTRODUCTION

The objective of cementation is to convert radioactive waste into a homogeneous stable monolithic form, which minimizes the probability of radionuclide release to the environment during interim storage, transportation and final disposal. The solidified waste leaving the immobilization plant should be of such chemical, mechanical, thermal and radiolytic stability that its integrity can be assured over the time required for the decay of the contained radionuclides to an acceptable level. To attain such characteristics in a long term a property which is prerequisite for all of them is the homogeneity.

Usually this property is not considered, except for a qualitative statement where there is gross phase separation. Specially in the setting up of receipts for waste immobilization in laboratory scale the homogeneity condition is assumed without objection. However, in the scale up of the immobilization process this condition is mandatory.

In laboratory scale procedures emphasis is given to the necessity of well homogenize the mixture in order to obtain an uniform and reproducible product. In counterpart this is not always done in the process itself. It is assumed that the process will delivers a homogeneous product and a simple visual inspection is enough to assure it.

In some few cases compressive tests are realized in samples taken from the real size product. However none reliable statistical test is usually applied to them.

In this paper is discussed the necessity of apply a complete set of tests before the homogeneity property be attributed to the waste form.

Statistical evaluation of the experimental data arising from the tests has to be made in order that the decision about homogeneity be objective.

This criterion has been applied to a vibration mixing process and shows be useful in order to improve the convergence of process variables as well to qualify it.

QUALIFICATION OF AN IMMOBILIZATION PROCESS

When developing a process to immobilize nuclear wastes a doubt occurs: What are the criteria to be applied to the waste form and what the process envisaged has to fulfil ?

In spite of the fact that a long list of properties are recognized have to be met by the waste form only few of them are required by national or international organizations.

Usually they are only: low leachability, long-term chemical stability, mechanical strength, radiation resistance and low water content.

More than this rather few organizations venture to give numbers to the established criteria leaving for the waste manager the charge of develop a "reasonable" waste form product for afterwards fix the rules.

Precise definitions of terms as well as definitive descriptions of the tests to be applied should be available in order that uniform standards and criteria for judging the adequacy of immobilized low-and intermediate-level waste forms be practiced.

Large difference of opinion exists concerning the relative importance of the properties considered desirable in immobilized waste forms. Anyway it is generally agreed that the main objective of the waste form is to retain the radioactive nuclides for long periods to ensure that the radioactivity has decayed to acceptable levels before reach the environment. How to arrive to such condition is function of several circumstances. The requirements and constraints imposed by the storage facility and disposal site can influence the choice of type and composition of the waste form. Many physical, chemical, physico-chemical and radiological properties have to be considered in order to attain such ideal waste form. As is not possible to satisfy all constrains at the same time or to achieve all the specified tests a small group of properties is required to be fulfilled.

The selection of that group of properties has to be made carefully in order that each selected property stands for a class of correlate properties. For instance, it is well known that the porosity affects mechanical strength as well the leach rate. No matter which property be considered it has to be uniform along the waste form. The criterion used to consider a waste form as homogeneous in terms of a given property is described in other paper of this seminar (1).

The selected group of properties have to be measured along the waste form following statistical criteria and afterwards analysed properly. Once those statistical criteria be satisfied the waste form can be considered homogeneous with regard to the selected properties. However, this is not enough. It must be necessary to establish the range of values for those properties in order that the waste be acceptable for a given waste management operation.

To fulfil these requirements is necessary to promote wider and frequent exchange of informations in order to foster the establishment of uniform standards and acceptance criteria for waste forms.

The laboratory scale test are used only to characterize the matrix product and not the process itself. For that purpose samples should be taken from the real or simulated waste form in order to measure the group of properties selected. The sampling have to be statisticaly valid and must show an uniform distribution along the waste form otherwise the immobilization process cannot be qualified.

HOW TO QUALIFY THE VIBRATION PROCESS ?

The vibration process is been proposed because it shows some attractive characteristics such as the cleanliness of the mixing process that reduces the external surface contamination of the container and of the inner side of the cementation cell. In this process the cement and waste to be immobilized are placed in the container, covered and put to vibrate in an apparatus as described in another paper presented in this seminar(2). Two kinds of containers are being assayed: one with 3.6 L and other with 20 L capacity.

Because the process is still in development and because the matrix samples produced are of real size type the chemical homogeneity investigation was adopted to verify if the process is on a good way of development. The chemical homogeneity criterion is also used because it is known that if the mixture of cement and waste is chemicaly homogeneous the other physical, chemical and physico-chemical properties have more chance of to be homogeneous. During the initial steps of development a cement dye was used as a tracer indicating the eventual improvements obtained in the process due to some applied modifications.

Figure 1 shows the results of a mixture vibrated using only cement dye as tracer in the early stage of the process development. Some parameters as rotation speed, vibration frequency, intensity, etc were varied resulting in a mixture which can be considered homogeneous by visual inspection.

As a second homogeneity test it was used uranium and thorium compound as tracers following the technique described elsewhere(1). It is possible to observe in figure 2 that the results are not yet satisfactory. Adjustments in the vibrator machine must be made. Other modifications will be applied to the container where baffle will be introduced to promote better mixture of the components using a combined effect of rotation and vibration.

Water absorption and pore volume was also determined and a heterogeneity of the mixture is observed between the top and the bottom of the vibrated samples. Table I shows the results obtained with paddle mixer and the vibration process.

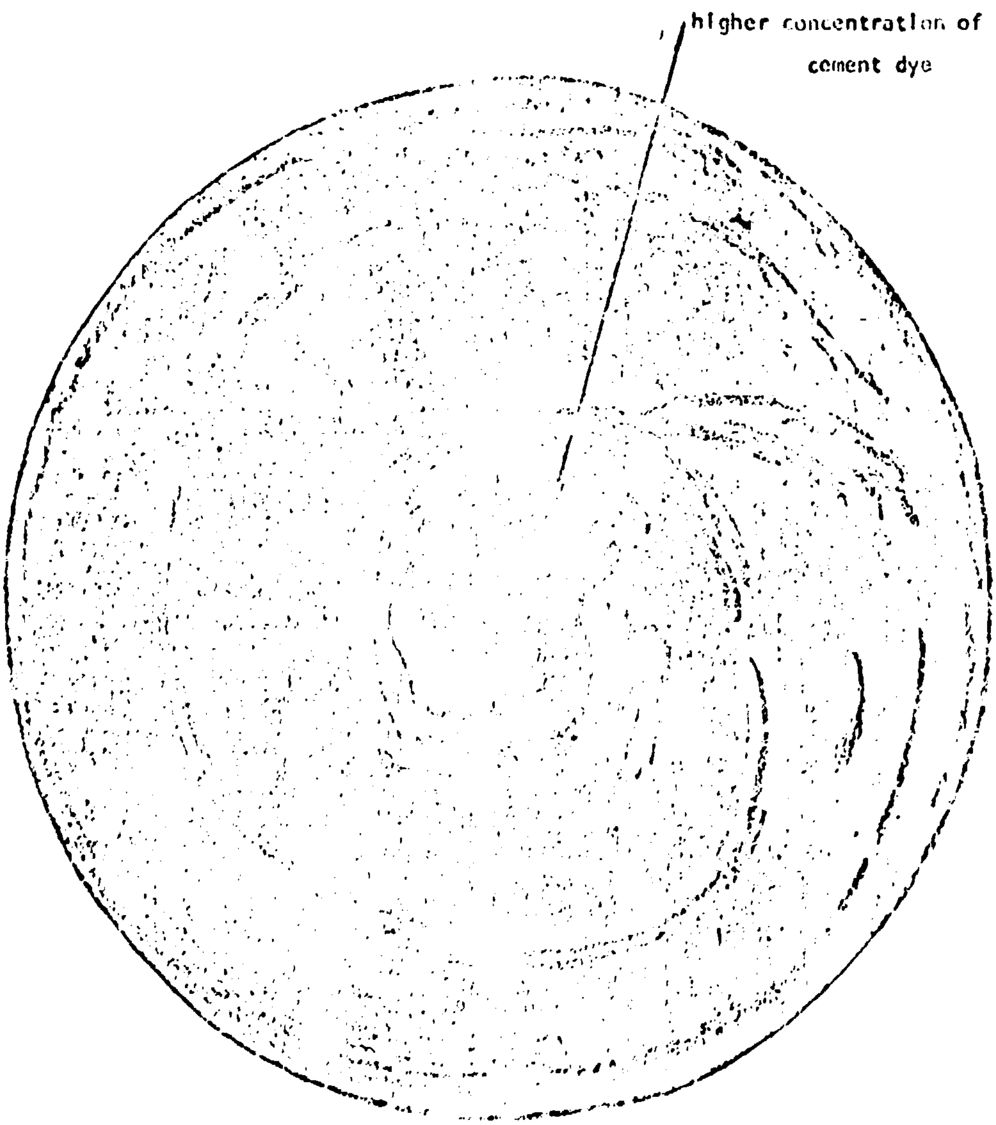
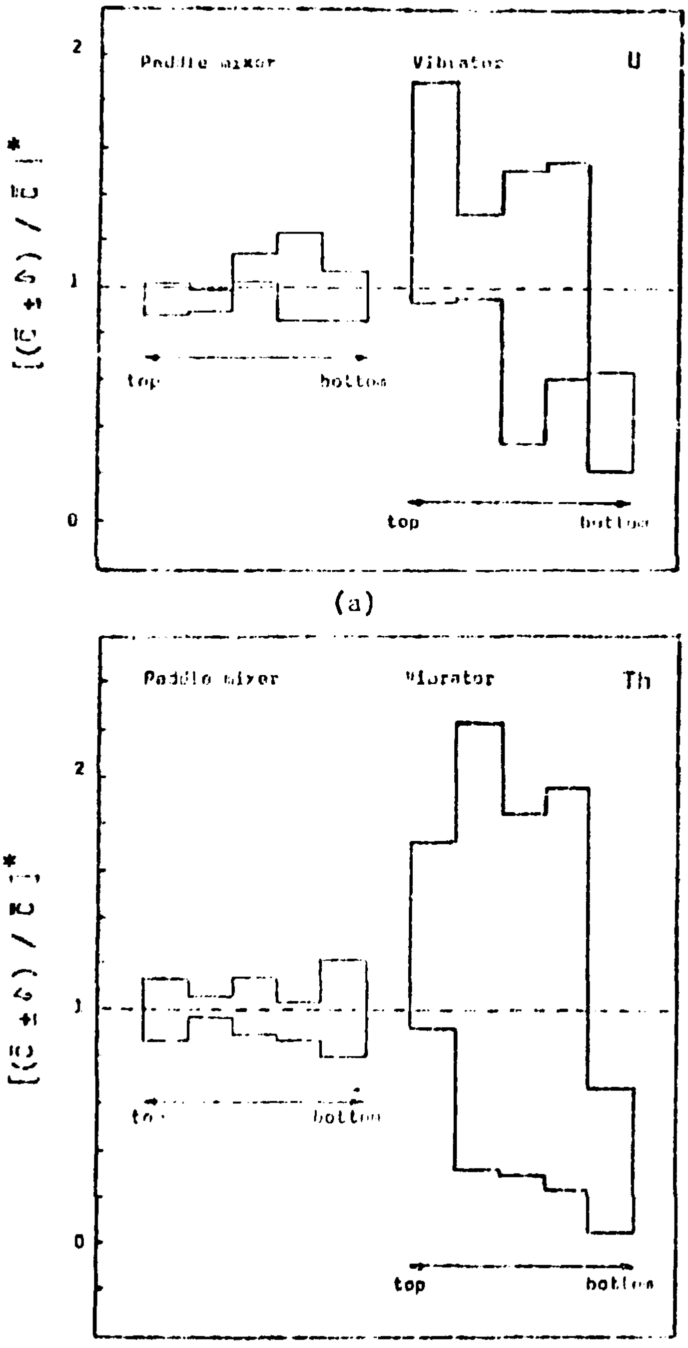


Figure 1 – Mixture of cement and cement dye by vibrated process



(a)

(b)

Figure 2 – Normalized concentration distribution along the sample matrix which contains (a) Uranium and (b) Thorium tracers for paddle mixer and vibrator mixing processes.
* $\bar{C}$ = slice average; σ = standard deviation; $\bar{\bar{C}}$ = full matrix average.

Table I

Comparison between samples processed by paddle mixer and vibrator.

Mixing process	Water absorption (mass %)	Pore volume (%)	Observations
Paddle	16.10 ± 0.01	30.15 ± 0.05	on the top
	15.96 ± 0.16	30.05 ± 0.10	on the bottom
Vibrator	18.97 ± 0.11	32.42 ± 0.28	on the top
	15.09 ± 0.21	27.84 ± 0.28	on the bottom

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2. VICENTE, R. Development and design of a cementation process. In: INTERNATIONAL ATOMIC ENERGY AGENCY. *Management optious for low and intermediate level wastes in Latin America: seminar. . . held in Rio de Janeiro, Oct. 13-17, 1986.* (Preprint)

FACILITY FOR LOW-LEVEL SOLID WASTE TREATMENT

R. Vicente, H. Miyamoto

ABSTRACT

A facility for low-level solid waste compaction, encapsulation and storage is described. Solid wastes are compacted in 200 ℓ drums and stored over concrete platforms covered with canvas, for decay or for interim storage before transport to the final disposal site.

INTRODUCTION

Low-level solid wastes arise from the research laboratories and radioisotopes production facilities in the Instituto de Pesquisas Energéticas e Nucleares - IPEN (Energy and Nuclear Research Institute).

The IPEN also receives wastes from other institutions, mainly radioisotopes users in hospitals.

These wastes have in most cases short-lived radionuclides, with half-lives of about weeks or a few months; long-lived radionuclides that can be present in the wastes are tritium, natural uranium and thorium and eventually small amounts of fission products and activation products from the research reactor IEA-R1.

These solid wastes are mainly made up of papers, plastics, rags, glass, air filters and small pieces of equipment discarded from laboratories.

Until 1978 those wastes were buried in shallow trenches, in selected sites within IPEN boundaries. Because of increasing wastes amounts and activities, decreasing space for disposal and aiming at getting expertise on waste treatment techniques it was decided to start studies for the construction and commissioning of a waste treatment facility. It was decided too, that the wastes should be stored in 200 liters drums without any treatment until the facility became operational.

The construction of the waste treatment facility was started in 1980 and in the middle of 1983 it begun operation.

The treatment adopted was volume reduction by compaction and storage for decay or interim storage before transport to final disposal.

Although compactible solid wastes were the urgent problem, provisions were made for future expansions to handle other wastes, like combustible wastes, liquid wastes requiring treatment and immobilization etc.

DESCRIPTION OF THE FACILITY

The waste treatment is located in a fenced area measuring 30 by 60 meters. The site is supplied with water, eletricity, telephone and sewer. The planned installations are shown in figure 1.

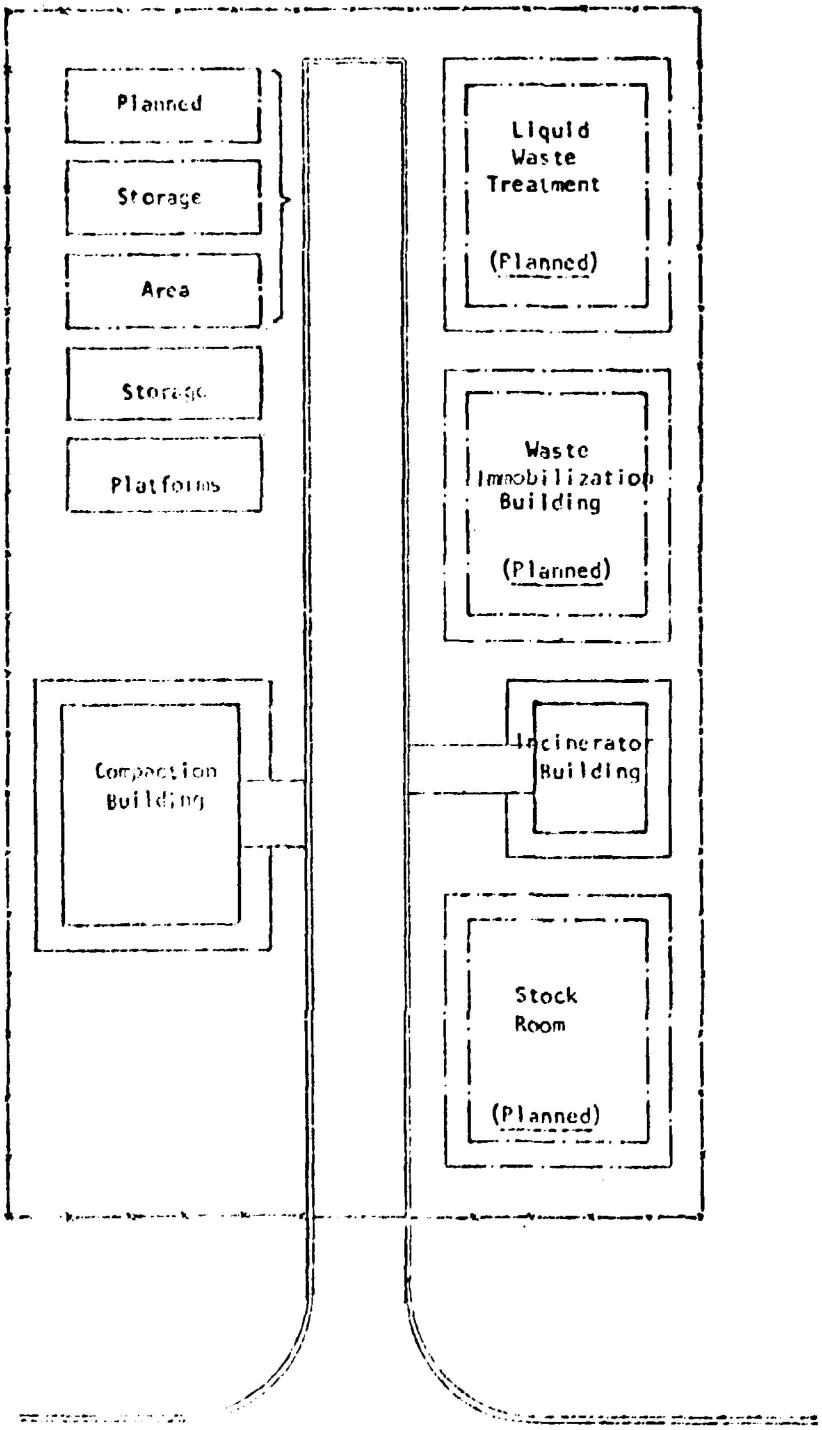

Figure 1 – Waste treatment facility

In the first stage the compaction facility and the storage place were built. Figure 2 is a lay-out of the building that shelters the waste compaction equipment. This is a commercially available 10 tons hydraulic press in which some modifications were made to adapt it for this work. It is shown in figure 3.

An exaust duct takes away gases, vapours and aerosols coming from the wastes being compacted. HEPA filters and charcoal filters prevent the release of radioactive materials to the environment. The air filtration system is shown in figure 4.

The building is fitted out with a space for the storage of uncompacted wastes that are transported from the laboratories in paper and plastic bags. The storage place is shielded with concrete walls.

A glove-box is provided for eventually necessary waste sorting and to dismantle, to cut or to saw pieces that not fit the drum or that could damage it. The glove-box is assigned mainly for the dismantling of replaced HEPA filters. It is shown in figure 5.

The floor under the press and under the glove-box and in the storage area is coated with an epoxi-resin-based paint for easy decontamination.

A trolley conveys unfilled and filled drums between the places of drum filling and compaction and to outside of the building.

A radioactive sewer drains water used in decontaminations of the storage area and the press and brings it to an interim storage tank placed underground and outside of the building. This is a 1 m^3 concrete tank, internally coated with epoxi-resin paint and fitted with a pump which can discharge the liquid to the sewage system or to a mobile tank. An alarm for high liquid level and a sampling line are provided in the tank.

The drums used as packaging for the wastes are made of 18 U. S. gauge tinplated iron sheet with two bracing rings and a removable lid. They are coated with epoxi-resin-based paint. This package complies with the requirements for LSA materials set forth in the regulations for the safe transport of radioactive materials.

A few percent of the solid waste volume can be classified as non-compactible waste. It is mainly metal scrap, debris from dismantling operations and contaminated tough materials.

This wastes are simply packed in the drums, or depending on the activity and nature of radioactive material, they are, after placed in the drums, embedded in fresh cement mortar.

The drums are stored over concrete platforms and are covered with canvas. In figure 6 the storage facility is shown.

Some modifications in the compaction and storage facilities were done later where the experience in operating the installation showed necessary. The observed problems and their solutions are presented in another paper (Experience on the operation of a low-level solid waste treatment facility).

SAFETY OF THE FACILITY

Some safety related aspects were considered in the design and the erection of the facility. Attention was paid for:

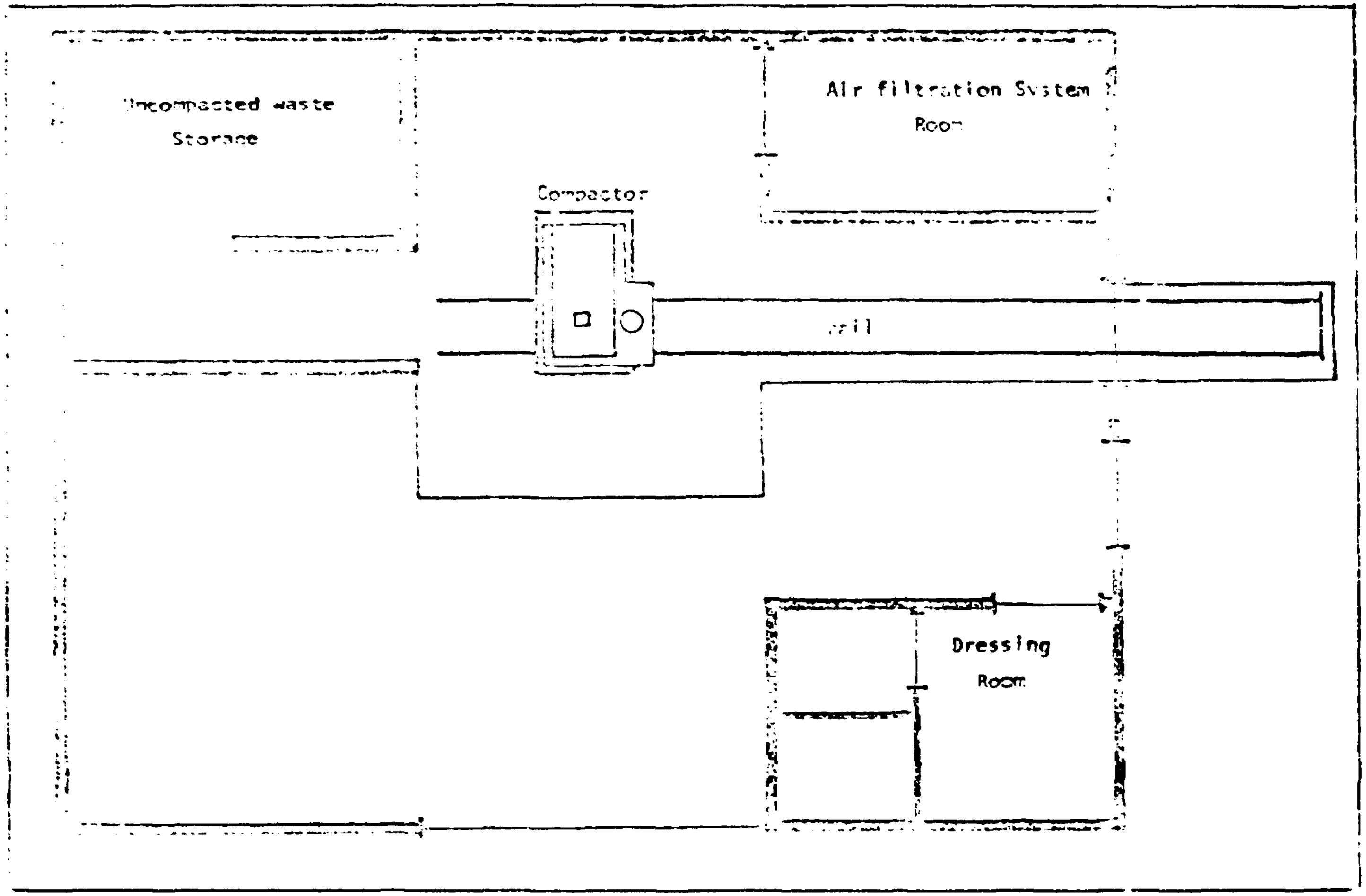

Figure 2 — Waste compaction building

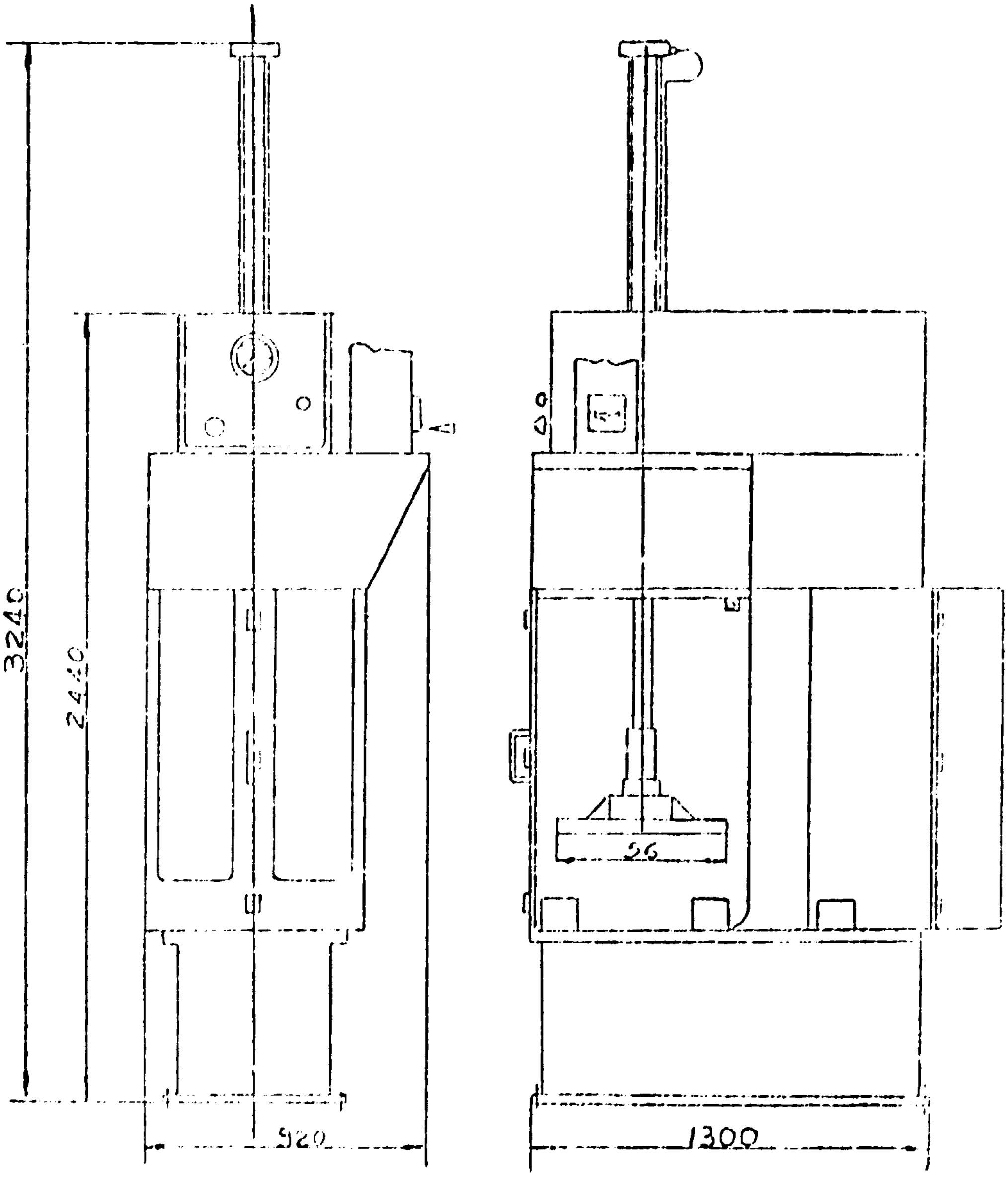

Figure 3 – Hydraulic press

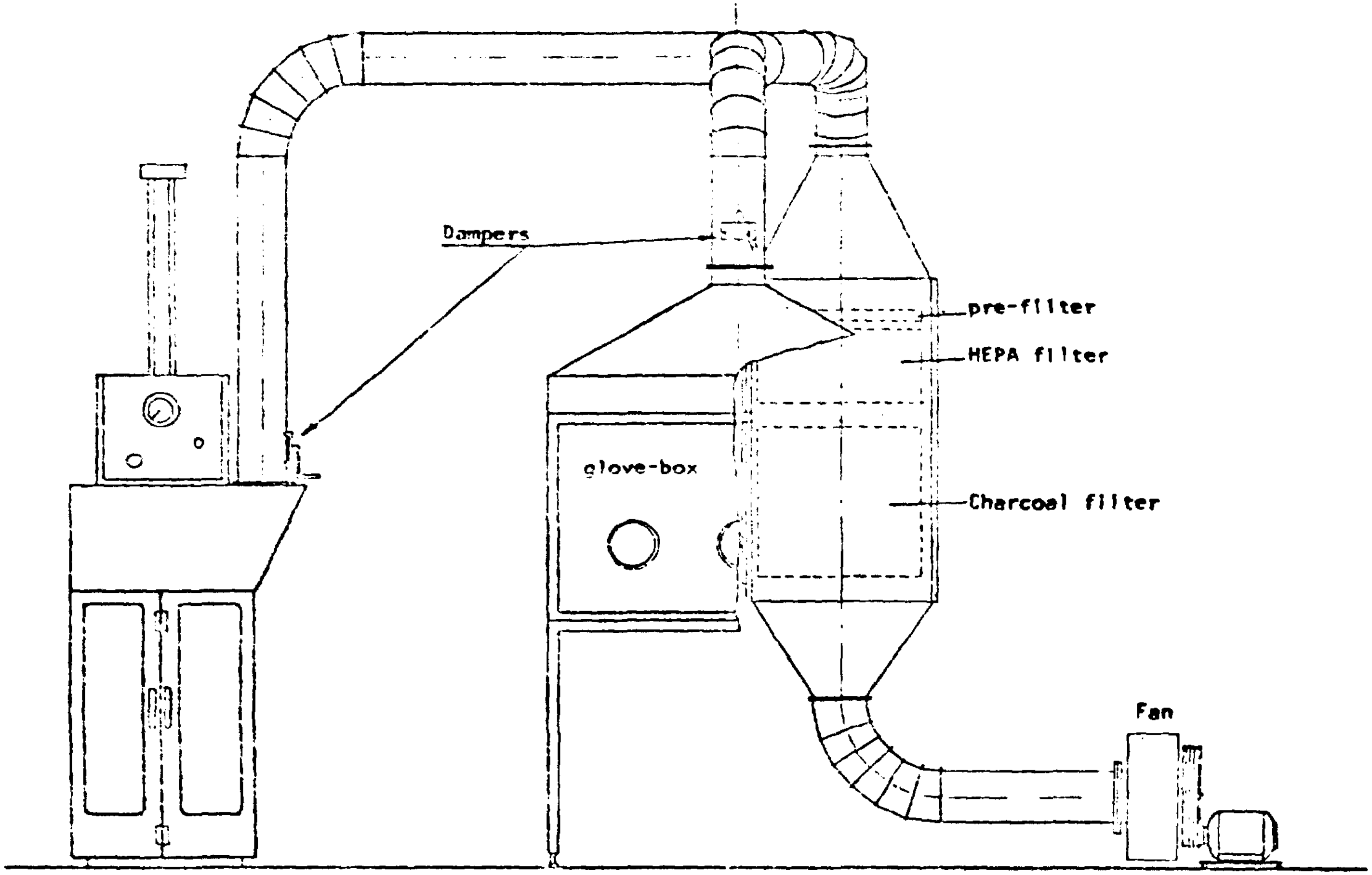

Figure 4 — Air filtration system

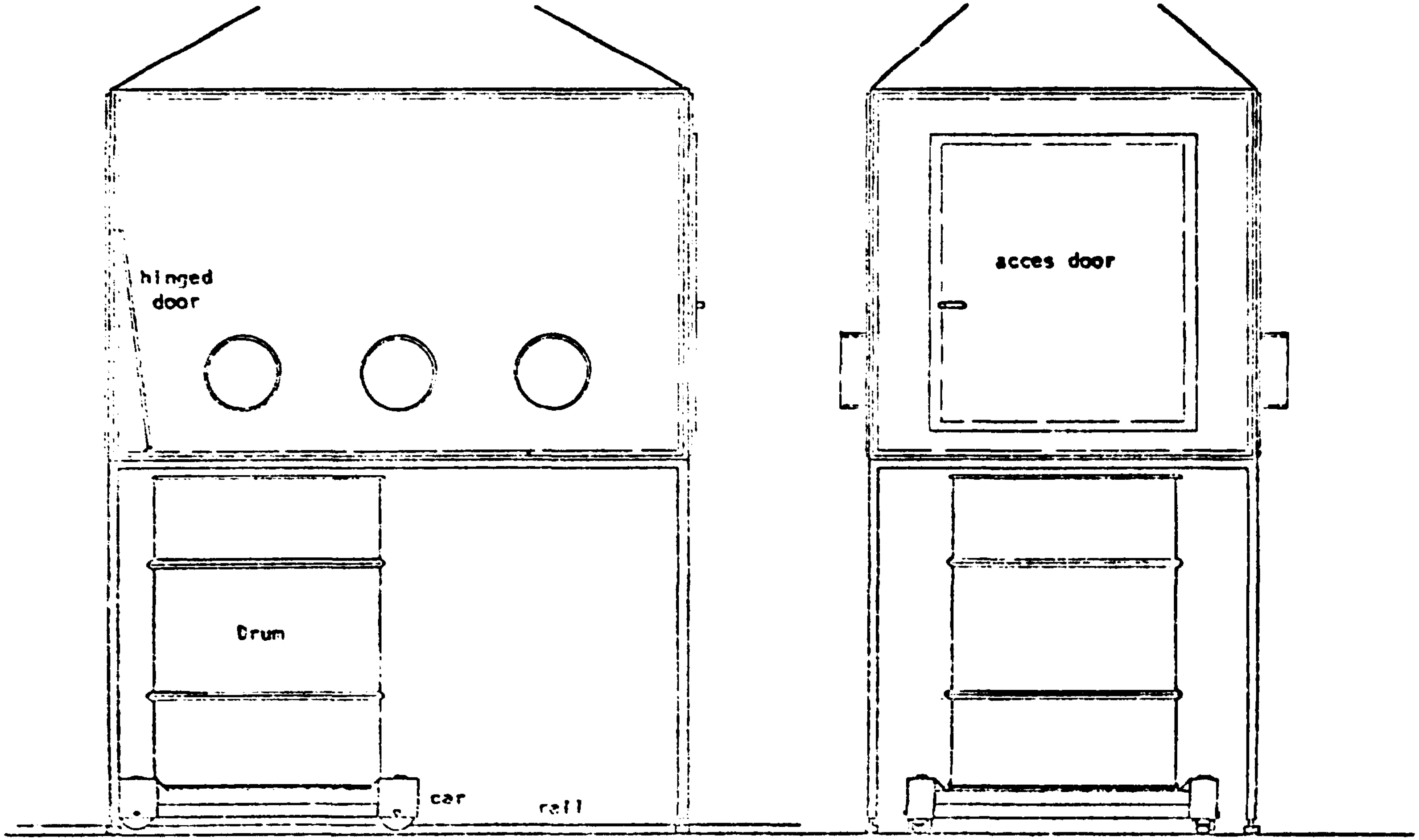

Figure 5 — Glove-box for sorting and dismantling

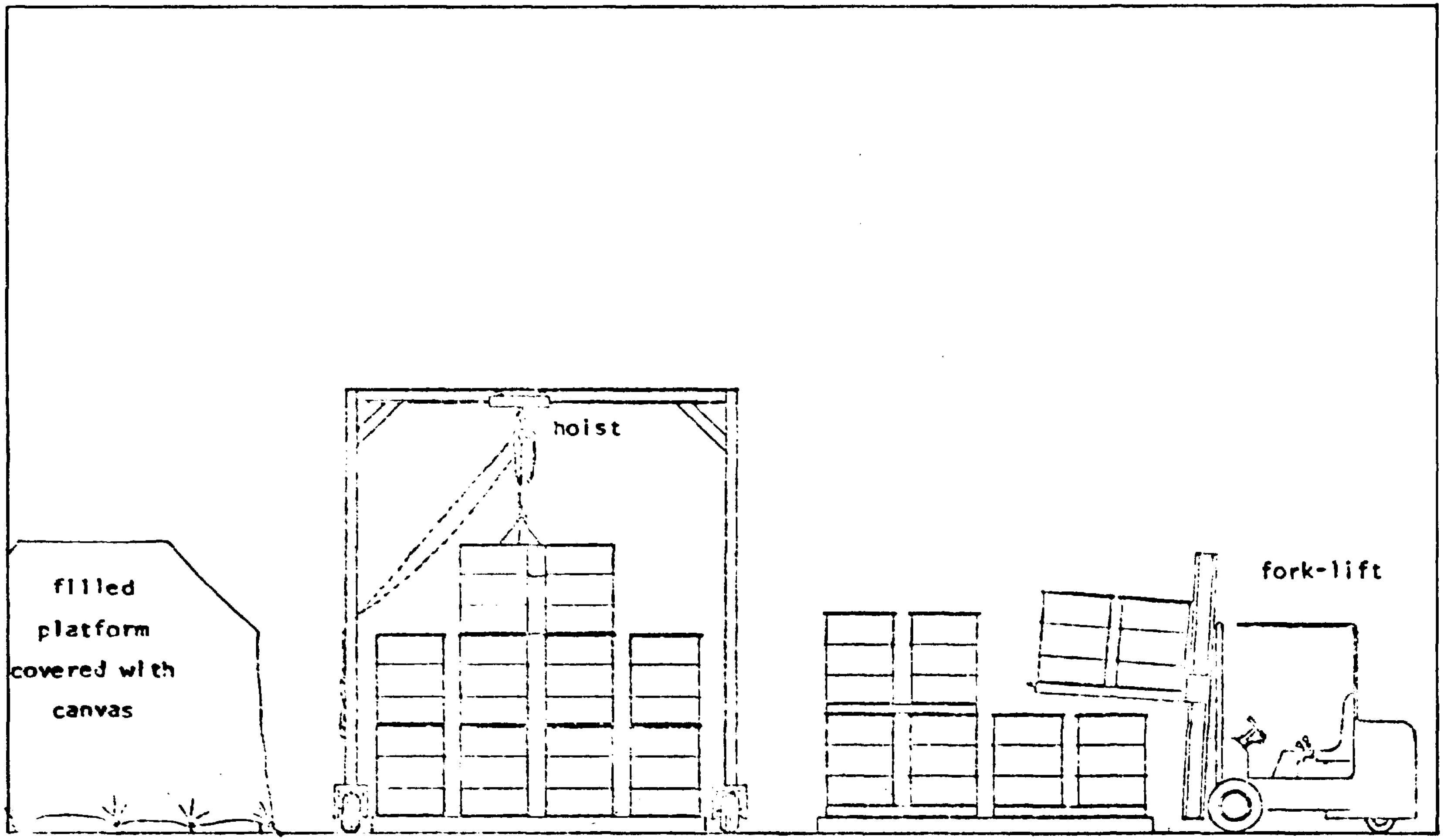

Figure 6 – Storage area

- Radiation doses in the personnel and public during normal operation;
- Radiation doses due to accidental releases;
- Countermeasures in accident conditions;
- Detection of fire and fire-extinguishing;
- Inadvertent intrusion.

In normal operation three irradiation paths were considered: irradiation by the gamma field; airborne radioactive material released from the air exhaust system; and radionuclides in liquid efluents released from the interim storage tank. It is, at least, difficult to know the radiochemical inventory of the solid wastes stored in the facility at any time. However estimates based on activities handled in the laboratories and facilities and on conceivable releases to the solid waste and from these to the environment upon compaction show that doses in general public are negligibly small. Also, the doses in the workers are well bellow dose limits for workers as shown by personnel dosimetry.

The doses incurred from accidents were calculated by simplified models and with a conservative scenario. Eventhough all the inventory were released in the air (as a consequence of fire, for instance) the doses in the critical group would be negligible. The possibility of flooding is unconceivable because the site is in a hill.

Small accidents that are possible to ocurr are the drop of a filled drum or a rip in a waste bag. In these cases the release of radioactive material is small and limited to the facility. To repack the waste and to decontaminate the floor are the measures forseen to handle the incident.

In any case, care is taken to do not include in the solid wastes: pyrophoric, explosive, putrescible or low flash-point materials.

The storage area for waste bags in the building is provided with a sprinkler system. At 60° C a shower is activated over the storage.

The facility is fenced with a 2,5 m fence with barbed wire in the top and hanging warning signals.

SITE CHARACTERIZATION

Two wells were dug in the site for ground water investigations. The water table lies between 6.5 m and 11 meter depth, and the horizontal water flow velocity varies between 9.3 and 42 cm/day.

The vertical water flow velocity was determined by tritium tracer; three months after injection of the tracer, the rain water infiltration was 134,9 cm.

The soil is clayed with silt and sand, were predominate caulite and ilite. The ion exchange capacity is low (0.7 to 2.0 meq/100 g).

Data of 1978 and 1979 indicates an average annual rainfall of 1399 mm; wind velocities less than 1 m/s predominate (about 30%) and maximum average velocities are about 10 m/s.

In figure 7 the population in the neighborhood of the facility are shown. Between 0 and 2 km in west, north and east directions that population is composed mainly by the workers and students of the University of São Paulo, IPEN and other public institutions. In the other regions the population is made up of dwellers.

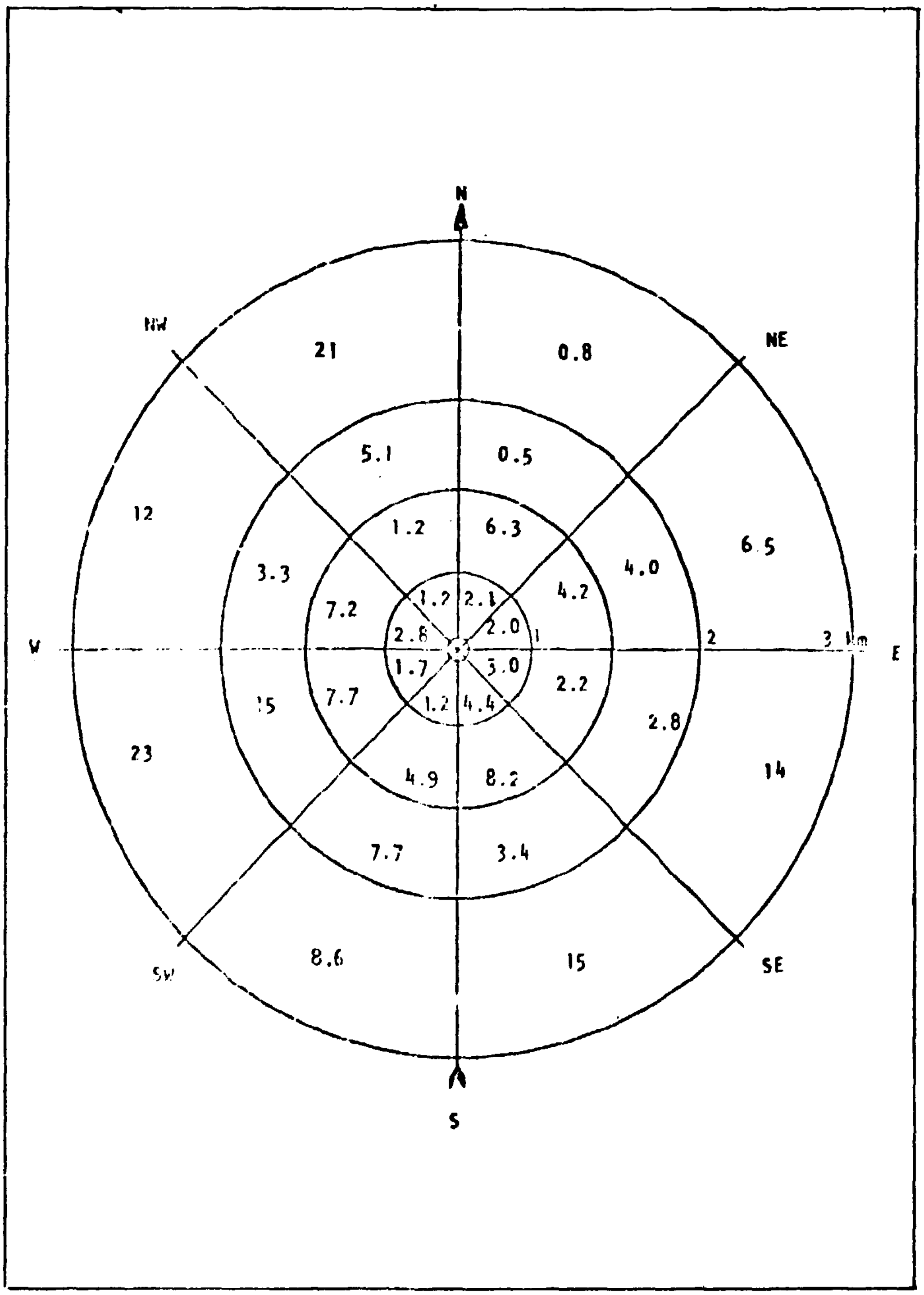

Figure 7 – Population near facility. (In thousands)

LEACHING STUDIES ON ION EXCHANGE RESINS IMMOBILIZED IN BITUMEN MATRIX

C. E. Grosche Filho, J. P. Villalobos, U. Chandra

ABSTRACT

To study radionuclide leaching from bitumen waste forms, many samples of bitumen mixed with ion-exchange resin labelled with ^{134}Cs were prepared. The resins used in the tests were nuclear grade mixed cationic/anionic bead resins. Different bitumen types were assayed: two distilled and two oxidized bitumens. Laboratory to scale samples, with surface/volume ratio (S/V) = 1, were molded to 5 cm diameter and 10 cm height. The composition of the mixtures were: 30, 40, 50 and 60 % by weight of dried resin with bitumen. The leachent was deionized water with a leachant volume to sample surface ratio of about 8 cm. Leached fractions were collected according to the recommendation of ISO method, with complete exchange of leachent after each sampling. The volume collected for analysis was one liter. Marinelli beckers were used for counting in a Ge(Li) detector. Up to now, results of 250 days have been accumulated. Samples prepared with distilled bitumen have shown a diffusion coefficient of the order of 10^{-14} cm^2/sec and those prepared with oxidized bitumen yielded a diffusion coefficient of the order of 10^{-12} cm^2/sec. Mathematical models of transport phenomena applied to cylindrical geometry were employed to fit experimental data.

INTRODUCTION

The comparation of various compositions of the radwaste bituminized products was studied by preparing different specimens of waste/bitumen while using two distilled and two oxidized bitumens.

The leaching experiments were conducted according the IAEA and ISO Standard recommendation.

Mathematical models of transport phenomena for cylindrical geometry were applied to fit the experimental data. This aims to explain leaching mechanism and to allow prediction of the long time behavior of the immobilized radionuclides. Three methods were used to investigate their ability to fit the experimental data: the first method was based on the time square- root versus leached fraction relationship; the second method tried was the log-log relationship; and the third method was based on diffusion law with concentration dependent dissolution rate.

EXPERIMENTAL PROCEDURE

Nuclear grade granular ion-exchange resin, in H^+ and OH^- form (IRN-150), loaded with Cs-134, was used for the study[4].

The resins were loaded by batch method, i. e. 4.5 kg of pure resins were equilibrated with 4.5 liters of distilled water containing 3.2 mCi of Cs-134. The labelling efficiency (> 99%) was determined by measuring the activity content in the solution after equilibrium. After loading, the resins were dried at 120° C for 16 hours.

Table I

Mathematical equations of transport mechanism

Medium	Transport mechanism	Equation for release of the mobile species (cumulative fractional release)
semi - infinite homogeneous chemically inert	diffusion	$\left(\frac{\Sigma a_n}{A_o}\right)\left(\frac{V}{S}\right) = 2\left(\frac{Dt}{\pi}\right)^{1/2}$ (EQ. 1)
semi - infinite uniform initial concentration	diffusion+ concentration-dependent dissolution	$\left(\frac{\Sigma a_n}{A_o}\right)\left(\frac{V}{S}\right) = (KD)^{1/2}\left[(t+1/K)\ \mathrm{erf}\ (Kt)^{1/2} + (t/\pi K)^{1/2}\, e^{-Kt}\right]$ (EQ. 2)

$\frac{V}{S}$ = Volume surface ratio; t = time; D = effective diffusivity constant; K = dissolution rate constant; erf = error function

Specimens for leach tests were prepared by mixing bitumens with 30, 40, 50 and 60 wt% of dry resin. Bitumes used were: T 50/60 and V 85/100 distilled type and T 75/25 and VB 65 oxidized type. The range of surface/volume ratio of the specimens was of 0.86 - 1.2.

Leaching was carried out at room temperature in polyethylene bottles with 1.6 ℓ distilled water as the leachant. The sampling frequency adopted was as per recommendation of ISO(2). The activities released in the leachant were determined by counting 1 liter of the leachant in Marinelli flasks using Ge-Li detector.

RESULTS

The experimental results presented take into account leaching time of 1000 days(4). Figures 1, 2, 3, and 4 show the cumulative leached fractions versus elapsed time for each bitumen type as a function of the waste composition.

For a better comparison, on the other hand, figures 5, 6, 7 and 8 show leaching from differents types of bitumen containing the same quantity of wastes load percents.

Mathematical models(3) based on transport phenomena applied for semi-infinite medium were used to fit the experimental data. Table I shows the matematical equations corresponding to these models.

Tables II and III show the values of diffusion constant D (cm^2/seg) and dissolution constant K (seg^{-1}) for a leaching interval of 250 days calculated by means of equations 1 and 2.

Equations 1 and 2 were used to predict the leaching fractions for a accumulative leaching time of 1000 days. The results so obtained are presented in the table IV.

DISCUSSION

Analysis of experimental and predicted data could not establish any definite correlation between cumulative leach rate and the waste loads (ion-exchange resins) in the bitumen products for either type of bitumen employed. The D and K values obtained (tables II and III) are of the same magnitude and are independent of the waste load and bitumen type. In the case of distilled bitumen specimens, this behavior could be explained due to the fact that such wastes products present a greater trend of deformation, which changes the surface to volume ratio. On the other hand oxidized bitumem specimens did not suffer deformations but for the significant swelling of the resin due to water absorption.

50% resin-bitumen products obtained with either type of bitumen showed better matrix integrity.

Experimental data of distilled bitumen specimens showed better fitting to mathematical models than oxidized bitumen specimens. In the case of oxidized bitumen specimens the estimated error in some cases were greater than the values of K and D.

Table II

Value of effective diffusivity (cm^2/s) calculated by transport equation 1

Ion-resins	Bitumen type			
(%)	T 50/60	T 75/25	V 85/100	VB 65
30	8.04E(-14) 3.75E(-13)	– –	– –	4.69E(-13) 6.72E(-13)
40	7.07E(-13)	4.12E(-13)	2.85E(-13)	1.11E(-12)
50	3.63E(-13) 2.50E(-13)	2.27E(-13) 1.31E(-13)	3.18E(-13) 1.81E(-13)	3.29E(-13) 1.78E(-13)
60	2.36E(-13)	4.58E(-14)	5.49E(-13)	1.18E(-13)

Table III

Value of effective diffusivity, D(cm^2/s) and dissolution rate constant, K(s^{-1}) calculated by transport equation 2

Ion-resins	T 50/60		T 75/25		V 85/100		VB 65	
(%)	D(cm^2/s)	k(s^{-1})	D(cm^2/s)	k(s^{-1})	D(cm^2/s)	K(s^{-1})	D(cm^2/s)	k(s^{-1})
30	2.39E(-14)	2.70E(-7)	–	–	–	–	*	*
	1.17E(-14)	3.48E(-6)	–	–	–	–	*	*
40	8.14E(-14)	8.76E(-7)	2.64E(-13)	6.73E(-8)	1.79E(-13)	1.46E(-7)	9.20E(-13)	1.23E(-8)
50	6.56E(-15)	6.11E(-6)	2.09E(-13)	7.28E(-11)	2.98E(-13)	8.45E(-9)	*	*
	7.35E(-14)	2.78E(-7)	1.47E(-13)	1.64E(-11)	4.19E(-14)	3.76E(-7)	1.34E(-13)	3.23E(-8)
60	1.66E(-14)	1.17E(-6)	3.33E(-14)	4.49E(-8)	4.76E(-14)	1.19E(-6)	*	*

* Equation 2 was unable to fit the data

Table IV

Comparison of the amounts leached from different bitumen after 1000 days to those predicted by equation 1 and equation 2

Bitumen type	Ion-resins (%)	Experimental data (cm).E(-3)	Predicted by	
			EQ.1 (cm).E(-3)	EQ.2 (cm).E(-3)
T 50/60	30	6.0	5.8	7.1
		9.8	6.4	17.5
	40	5.1	8.8	23.2
	50	3.6	6.3	17.3
		4.2	5.3	12.6
	60	2.2	5.1	12.1
T 75/25	40	4.0	6.7	12.5
	50	2.2	5.0	4.8
		2.1	3.7	4.0
	6.0	5.8	2.2	3.7
V 85/100	40	4.8	6.6	14.5
	50	2.7	5.9	7.0
		2.2	4.4	11.0
	60	12.2	7.8	20.6
VB 65	30	3.8	7.2	–
		4.8	5.6	–
	40	4.9	11.0	4.3
	50	2.4	6.0	–
		10.8	4.6	4.0
	60	6.4	3.6	–

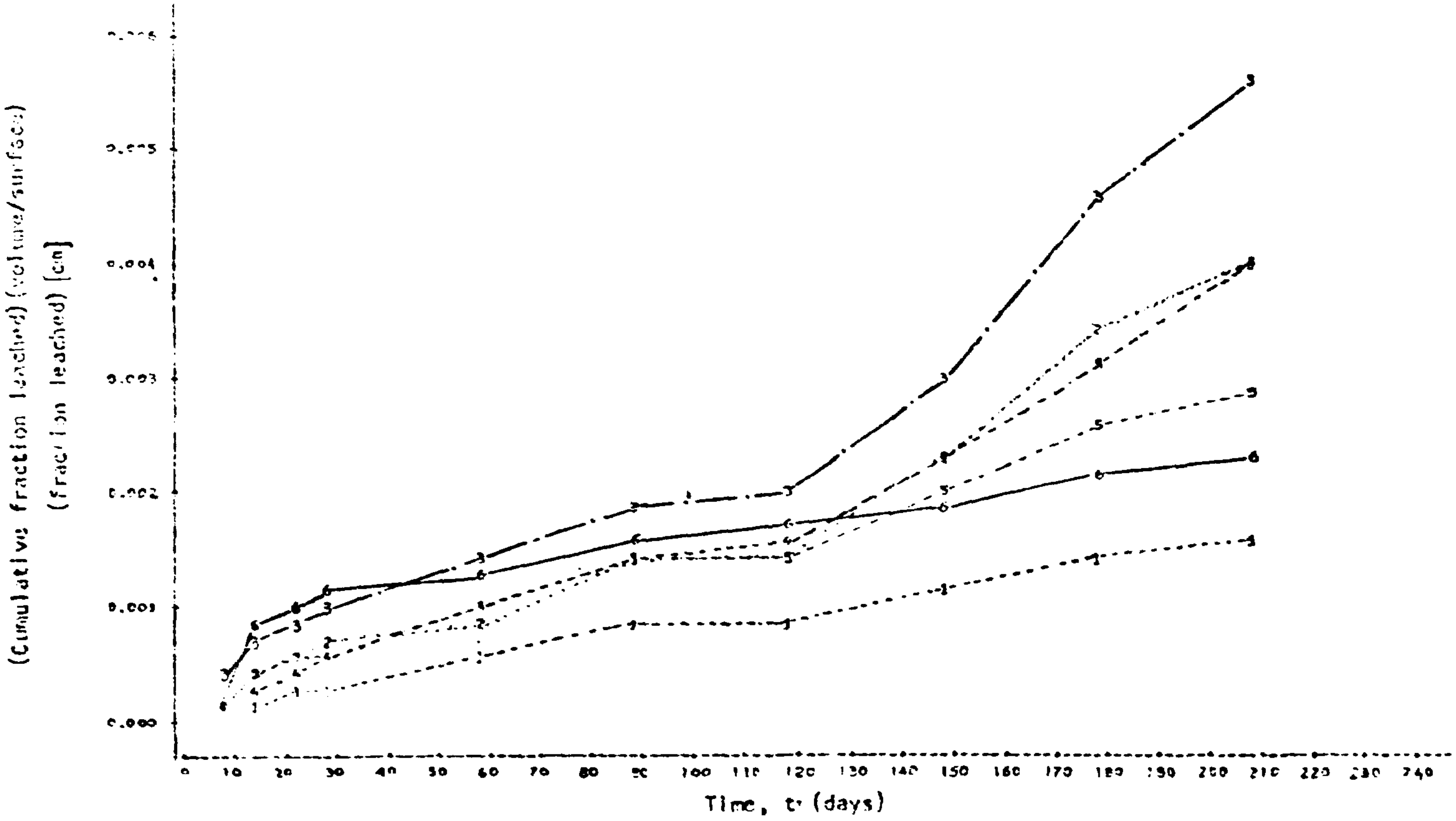

Figure 1 — Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the distilled bitumen type T 50/60

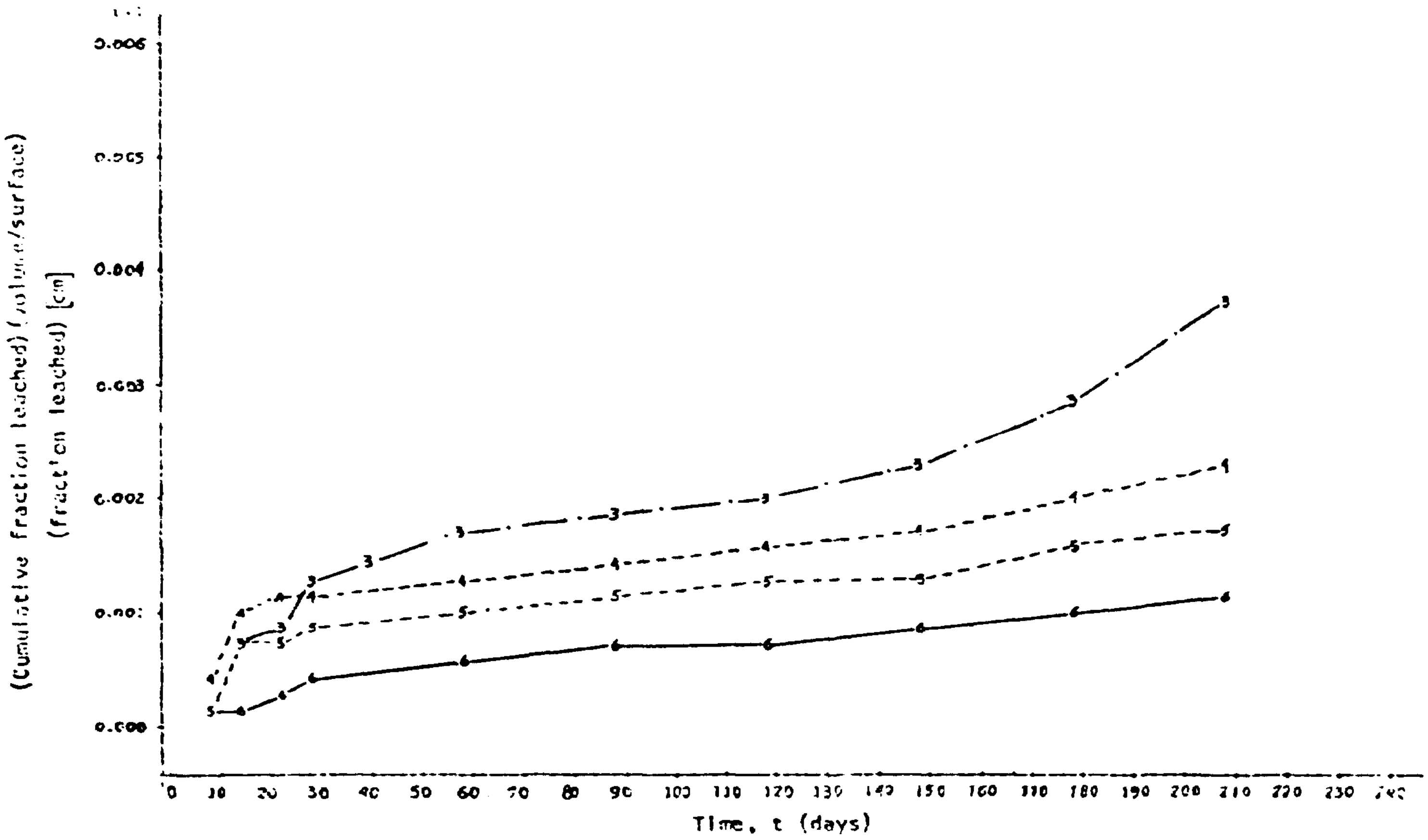

Figure 2 – Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the oxidized bitumen type T 75/25

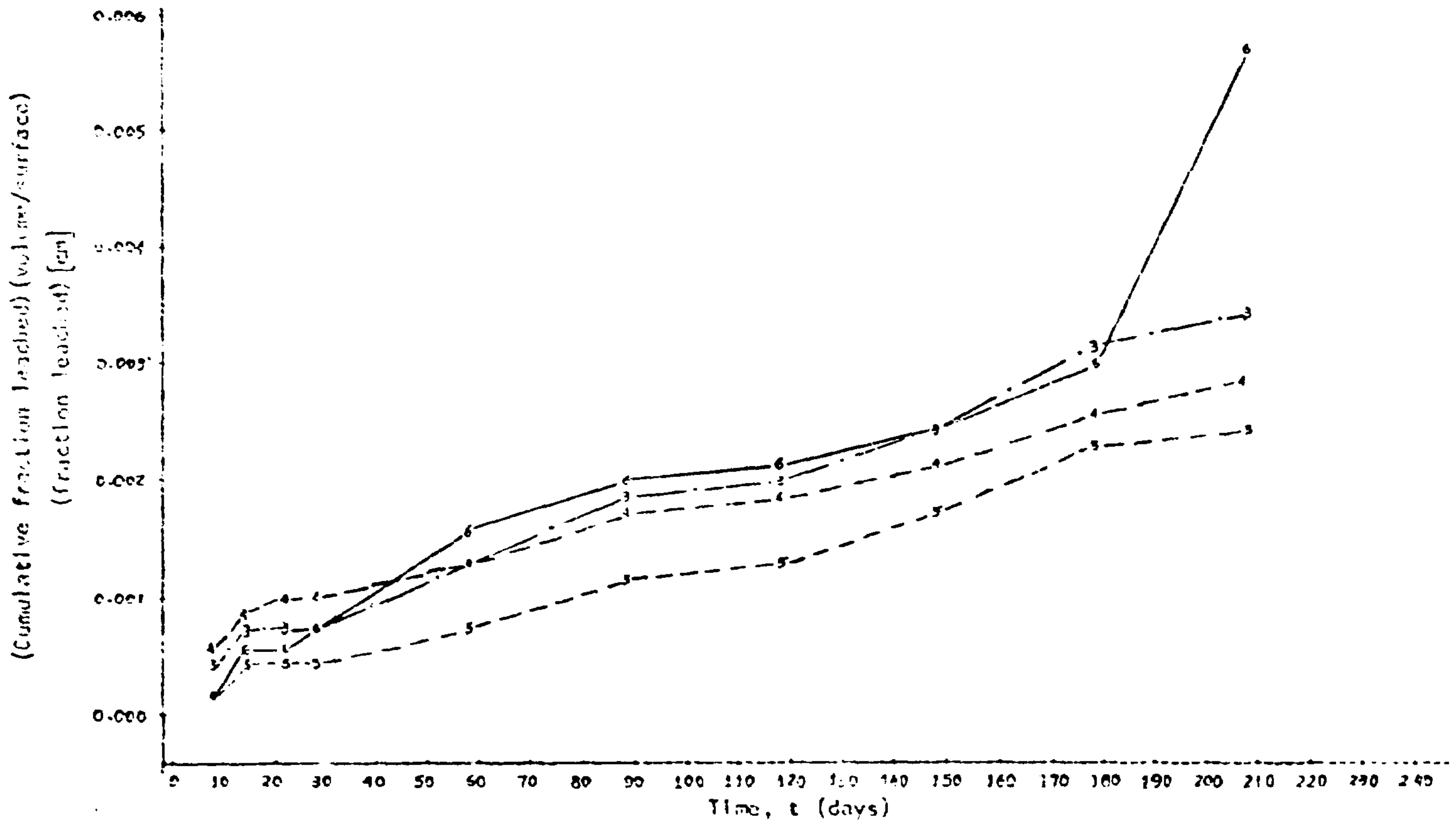

Figure 3 – Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the distilled bitumen type V 85/100

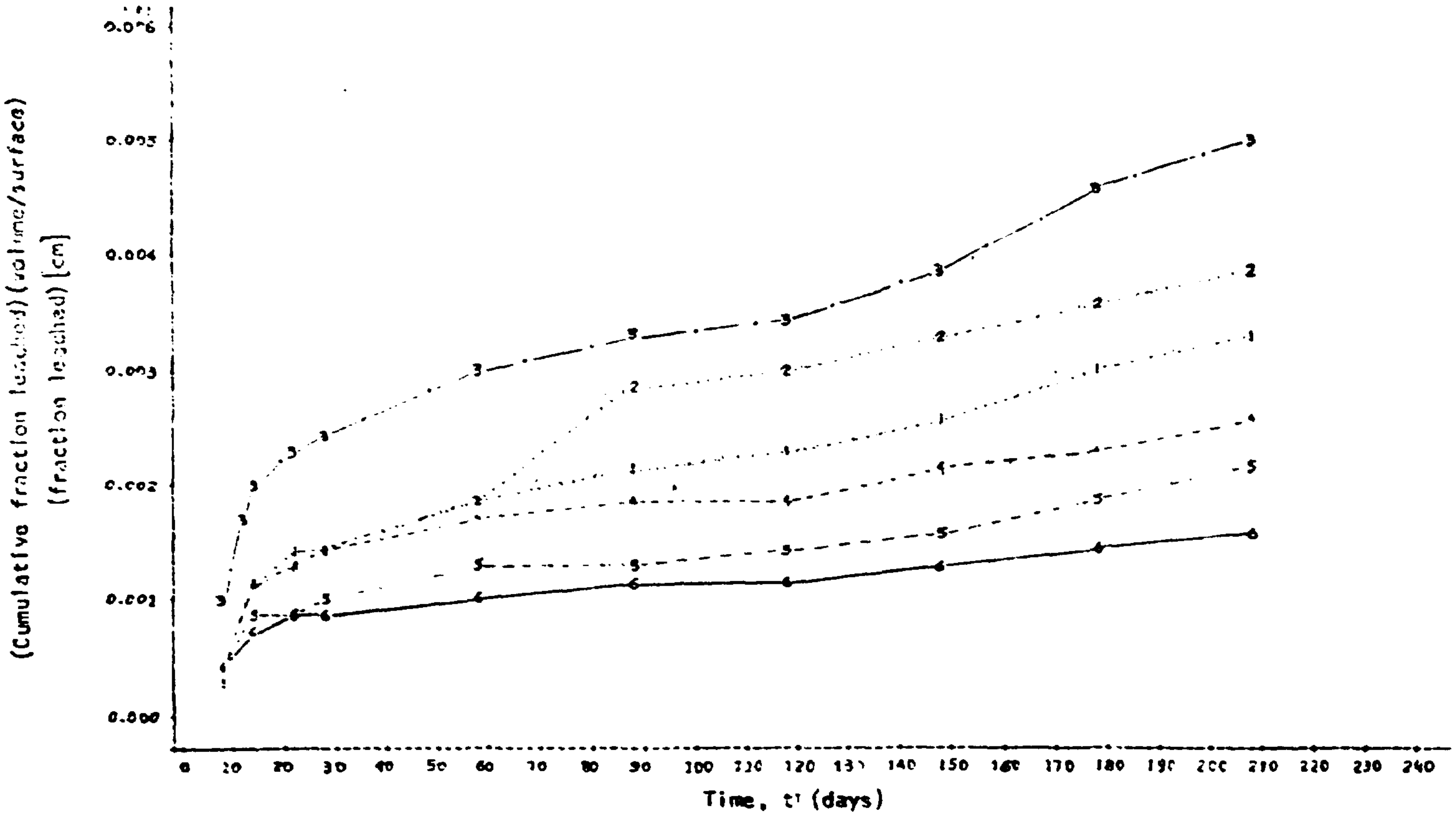

Figure 4 – Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the oxidized bitumen type VB 65

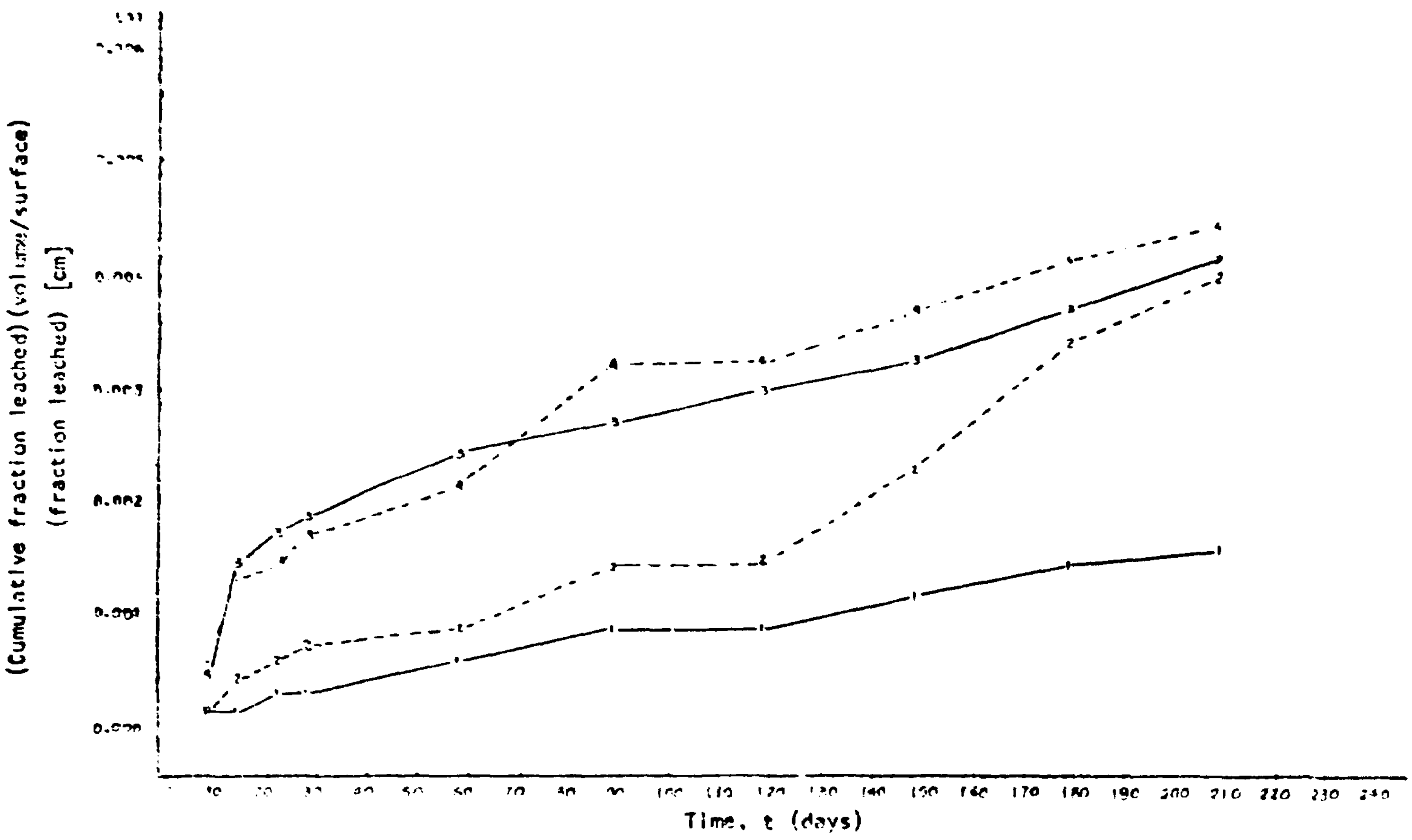

Figure 5 – Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the different type of bitumen studied containing 30% by weight of the ion exchange resin.

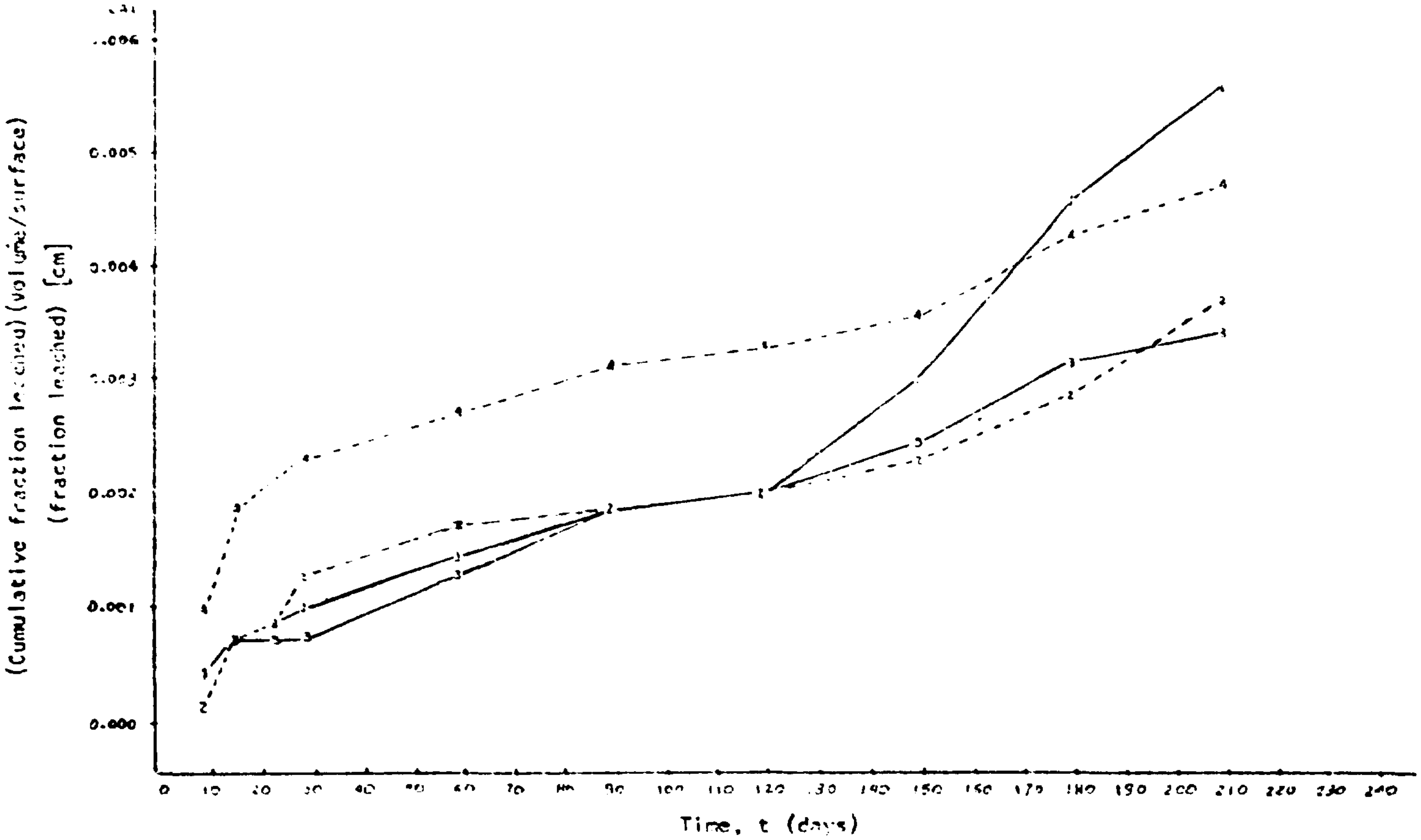

Figure 6 – Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the different type of bitumen studied containing 40% by weight of the ion exchange resin.

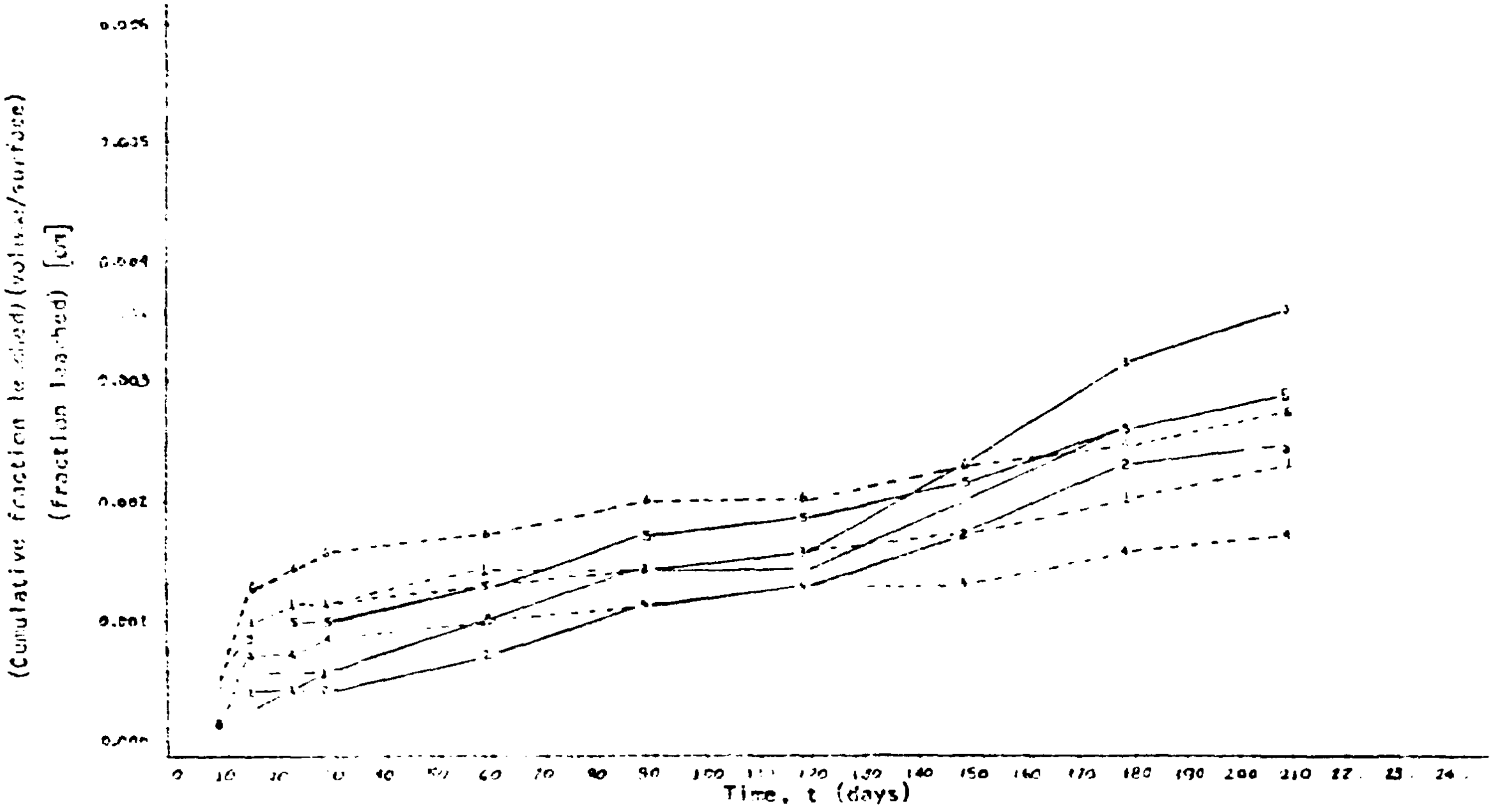

Figure 7 – Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the different type of bitumen studied containing 50% by weight of the ion exchange resin.

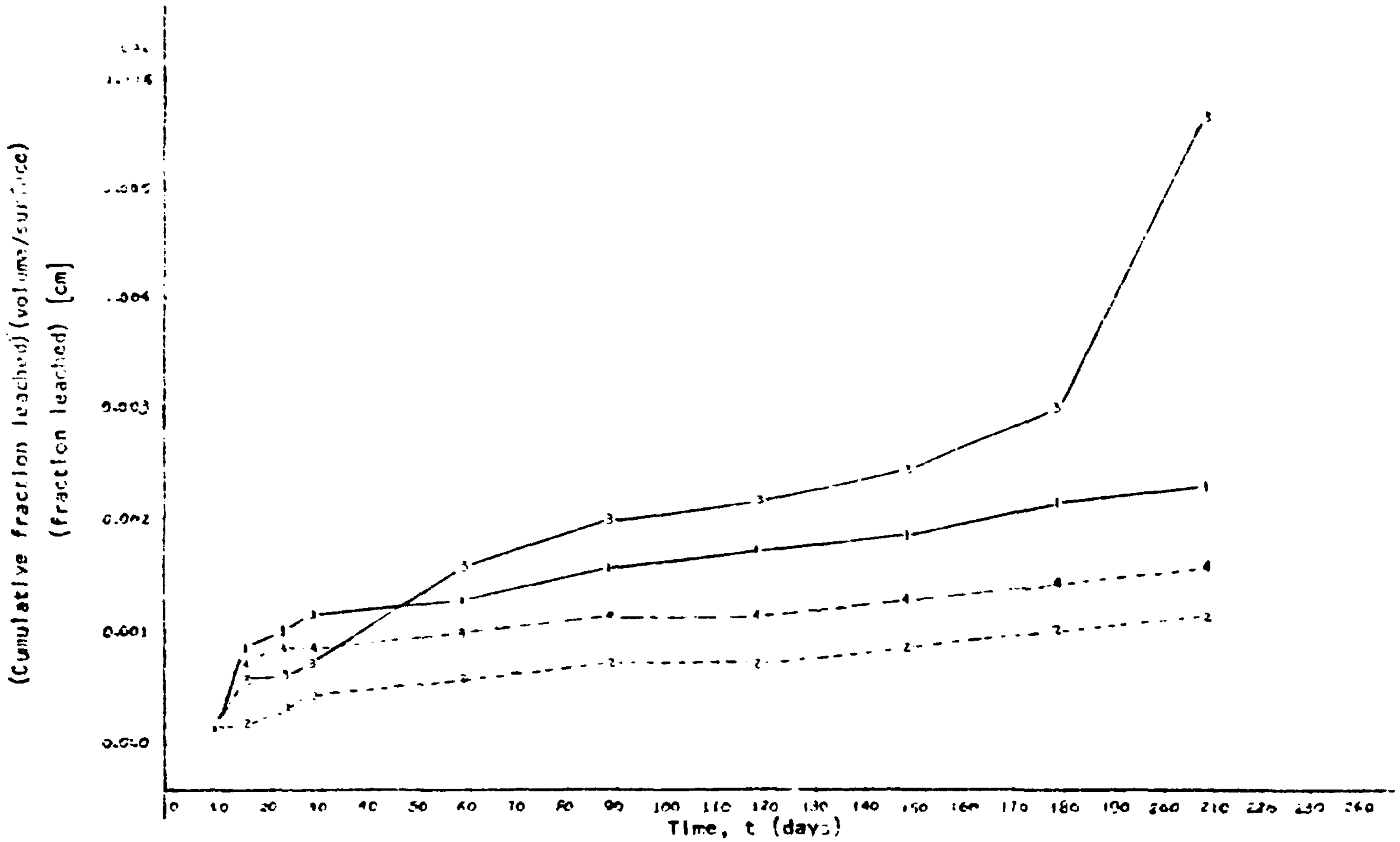

Figure 8 – Cumulative fraction leached multiplied by the volume-surface ratio plotted against time for the different type of bitumen studied containing 60% by weight of the ion exchange resin.

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BITUMINIZATION OF SIMULATED WASTE - SPENT RESINS, EVAPORATOR CONCENTRATES AND ANIMAL ASHES BY EXTRUSION PROCESS

C. E. Grosche Filho, U. Chandra

ABSTRACT

The results of the study of bituminization of simulated radwaste - spent ion-exchange resins, borate - evaporator - concentrates and animal ashes, are presented and discussed.

Distilled and oxidized bitumen were used. Characterization of the crude material and simulated wastes-bitumen mixtures of varying weight composition (30, 40, 50, 60% by weight of dry waste material) was carried out. The asphaltene and parafin contents in the bitumens were also determined.

Some additives and clays were used with an aim to improve the characteristics of solidified wastes.

For leaching studies, granular ion-exchange resins were loaded with Cs - 134 and mixtures of resin-bitumen were prepared. The leaching studies were executed using the IAEA recommendation and the ISO method.

A conventional screw-extruder, common in plastic industry, was used to determine operational parameters and process difficulties. Mixtures of resin-bitumen and evaporator concentrate-bitumen obtained from differents operational conditions were characterized.

INTRODUCTION

Two distilled bitumens and three oxidized bitumens samples were selected to characterize the locally available commercial bitumens. This choice was made by comparing them with the international bitumens, using the physical properties as selection parameter.

Analysis of the asphaltenes and parafins was carried out for better characterization and interpretation of wastes-bitumen behaviour.

Simulated PWR wastes - spent granular ion-exchange resins, borate-evaporator-concentrates and animal ashes were utilized for the bituminization studies. According to IAEA classification these wastes belong to the category of low-and intermediate-level radioactive wastes ($A \leq 3,7 \times 10^{13}$ Bq/m^3).

CHARACTERISTICS OF THE BITUMENS

The characteristics of the distilled and oxidized bitumens selected for the bituminization process are indicated in the table I and II(3).

The physical properties were determined using the standard ASTM tests.

The asphaltene and parafin contents were determined using the method of "Rostler-Sterberg".

Table I

General bitumen properties (average values)

	Bitumen				
	Oxidized			Distilled	
Properties	T 75/25	TB 90	VB 65	T 50/60	V 85/100
Penetration (0.1mm)	25	10	25	54	86
Softening point (°C)	82	84	72	52	46
Flash point (°C)	254	286	234	288	250
Specific density at 25°C (g/cm^3)	1.02	1.02	1.08	1.02	1.04
Burning point (°C)	340	–	–	360	–

Table II

Asphaltene and parafin contents of the bitumen samples

Bitumen	Fractions %	
	Asphaltenes	Parafins
T 50/60	34.3	10.9
T 75/25	31.0	20.1
TB 90	33.3	8.0
V 85/100	37.8	13.1
VB 65	23.2	7.2

Physical properties of the some bitumens utilized in various countries are given in table III[1].

RADWASTES UTILIZED

The simulated wastes considered were:

- Nuclear grade IRN-150 (H^+, OH^-) ion-exchange resins, granular type.
- Evaporador borate-concentrates of the following weight composition:

H_3BO_3	70%
NaOH	12%
Na_2SO_4	12%
Na_2PO_4	2%
NaCl	2%
$Fe_2(SO_4)_3$	2%

- Animal ashes

Table III

Properties of various bitumens

	Distilles			Oxidized	
	Mexphalt 10/20	Mexphalt 40/50	Mexphalt 50/60	R 90/40	R 85/40
Softening point (°C)	65 - 75	59 - 69	50 - 58	85 - 90	80 - 90
Penetration at 25°C (0.1 mm)	10 - 25	20 - 30	50 - 60	35 - 40	35 - 45
Flash point (1°C)	>250	>250	>250	250	240
Density at 25°C (g/cm^3)	1.02 - 1.07	1.01 - 1.06	1.01 - 1.05	1.01 - 1.05	1.01 - 1.05

SIMULATED RADWASTE-BITUMEN MIXTURES

The mixtures prepared in the laboratory scale consisted of:

- Resin/bitumen with 30, 40, 50, 60 wt% of the dry waste material;
- Resin/bitumen/additive, with 50 wt% of the dry resin and 2 wt% of the additive by bitumen weight;
- Evaporator borate-concentrates/bitumens with 30, 40, 50, 60 wt% of the dry waste material;

- Evaporator borate-concentrates/bitumens/additive with 40 wt% of the dry concentrate and 2 wt% of the additive by bitumen weight;
- Animal ashes/bitumen with 40, 50, 60, wt% of the ashes in the dry material.

The evaluation of the twin screw extrusion process, was made while using the 40 and 50% by weight mixtures and different operating parameters of the extruder.

The characteristics of the extruder were as following:

- screw (twin, coupled, with no auto-cleaning and without extra gas/vapor outlets).

  diameter - 50 mm
  length - 1000 mm
  pitch - 12 → 8 mm (variable)
  depth - 10 → 2 mm (variable)
  rpm - 0 ~ 60

- Heating,
  three heating zones with temperature range between 0°C - 230°C

- Motor,
  7,5 HP/1750 rpm.

LEACH TESTS

For leaching studies resin -bitumen specimens were prepared. Cesium-134 was used for labelling the resin.

The dimensions of the cylindrical specimens were: diameter 5 cm, height 10 cm - with volume/surface ratio ≃ 1. The resin-bitumen mixtures studied contained 30, 40, 50 and 60 wt% of the dry material. 1.6 ℓ of distilled water was used as leachant.

RESULTS

The laboratory scale results of the resins, evaporator-concentrates and animal ashes are shown in tables IV, V and VI.

The operational conditions of the extruder tests are indicated in table VII and some characteristics of the bituminized products are shown in table VIII. The results of the leaching studies are shown in table IX.

DISCUSSION

The data in the tables I to IX demonstrate the compatibility of the bitumens used by us in comparison to the bitumens used in other countries. The characteristics of the bitumen, however, depend on the origin of the petroleum and processing in the refineries.

The bitumens studied showed easy workability in the temperature range between 150°C and 180°C. In this aspect distilled bitumen demonstrated better workability as compared to the oxidized bitumen.

Table IV

Physical properties of resin/bitumen mixtures in laboratory specimens

Granular resin (%)	Flash point (°C)	Softening point (°C)	Penetration at 25°C (0.1mm)	Bitumen
30	228	88	13	T 75/25
40	228	88	12	
50	220	93	11	
60	197	108	9	
70	120	113/119	(*)	
30	234	61	32	T 50/60
40	234	66	29	
50	222	67	20	
60	208	85	7	
70	116	90/117	(*)	
30	238	52	53	V 85/100
40	232	54	43	
50	230	59	29	
60	230	69	17	
70	136	115/130	(*)	
30	232	76	17	VB 65
40	232	79	12	
50	216	89	10	
60	182	126	5	
70	136	128/131	(*)	

(*) Values not determined

Table V

Physical properties of borate – concentrates/bitumen mixtures in laboratory specimens.

Borates concentrates (*)	Flash point (°C)	Saftening point (°C)	Penetration at 25°C (0.1 mm)	Bitumen
30	238	91	(*)	T 50/60
40	258	88	(*)	
50	210	105	13	
60	273	90	(*)	
30	202	95	20	T 75/25
40	174	102	(*)	
50	288	102	(*)	
30	218	122	(*)	T 90
40	208	116	(*)	
50	218	107	(*)	
30	215	56	44	V 85/100
40	215	77	(*)	
50	227	91	(*)	
30	178	80	21	VB 65
40	200	84	12	
50	186	86	(*)	

(*) Values not determined

Table VI

Physical properties of ashes/bitumen mixtures in laboratory specimens

Ashes (%)	Flash point (°C)	Softening point (°C)	Penetration at 25°C (0.1 mm)	Bitumen
40	263	95	15	T 50/60
50	287	105	8	
60	211	129	7	
40	218	97	17	T 75/25
50	230	115	14	
60	225	142	10	
50	264	116	5	TB 90
60	252	137	5	

Table VII

Operational conditions in the extruder for bituminization

Waste	Sample test (nº)	Water content (wt%)		Speed (rpm)	Temperature (°C)			
		Waste	Product		Heating zone			Product
					1	2	3	
Resin	1	40	–	–	–	–	–	–
	2	70	25	20	150	150	190	110
	3	0	0	20	150	150	180	160
	4	78	22	30	250	150	150	140
	5	80	1.7	48	200	200	150	125
	6	78	2.6	54	180	180	180	125
	7	78	3	54	180	180	180	115
Borate - concentrates	1	17	–	–	–	–	–	–
	2	0	0	20	150	150	190	165
	3	0	0	60	170	170	230	195
	4	64	7	60	170	170	230	–
	5	69	1.4	20	150	200	200	125
	6	75	2.2	48	200	200	180	120
	7	63	0	54	180	200	200	125
	8	70	5	54	180	180	180	110
	9	61	8	54	180	180	180	120
	10	60	4	54	180	180	180	110

Table VIII

Physical characteristics of some resin and borate concentrates bituminized by extruder process

Waste	Sample test (nº)	Physical characteristic		
		Flash point (°C)	Softening point (°C)	Penetration at 25°C (0.1 mm)
Resin	2	302	72	16
	3	302	77	46
	4	286	64	30
	6	300	62	48
Borate Concentrates	3	285	70	57
	6	280	60	33

Table IX

Results of leach test (250 days)

Bitumen type		(Cumulative fraction leached). (volume/surface) (cm) . 10 [illegible]
Distilled	T 50/60	1.5 – 5.5
	V 85/100	2.2 – 6.0
Oxidized	T 75/25	1.2 – 6
	VB 65	1.5 – 5.5

Characterization of the bitumen waste mixture by penetration test is not an adequate procedure. More than 50% by weight resin-bitumen mixture demonstrate an uneven surface thus invalidating the penetration test.

Due to the anionic resin (amine) content in the resin-bitumen products the flash point decreased with the increase of the resin quantities. The borate-concentrate bituminized products, however did not show any such behaviour of the flash point.

The results of the 250 days leaching tests performed in distilled water, showed no significant difference among the various bitumen types. The range of the diffusion coefficients are between 10^{-14} - 10^{-12} cm^2/sec. The long-term studies are necessary for understanding the leaching behaviour of the bituminized resin.

The extruder showed better product performance in the temperature range between 150-180°C and 50-60 rpm range. Plugging in the extruder was observed with the evaporator-concentrates because of the geometrical characteristics of the screw and limit of the maximum attainable rpm. In the extrusion of resin-bitumen no such difficulty was observed. It was possible to obtain homogeneous products with small residual water contents. With extra gas/vapor outlets, the final products could be obtained without trapped moisture.

CONCLUSION

The preliminary experiments executed in the laboratory and with a conventional industrial extruder demonstrate that bituminization by extrusion is a satisfactory process for immobilization of granular ion-exchange resins, evaporator borate-concentrates and animal ashes. The process results in waste forms with good characteristics.

ACKNOWLEDGEMENTS

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DESIGN OF AN EVAPORATOR/CRYSTALLIZER FOR RADIOACTIVE LIQUID WASTE TREATMENT

C. E. Grosche Filho, J. C. Dellamano, L. Dalaqua Junior

ABSTRACT

This paper describes a design of an evaporation/crystalization unit for liquid waste treatment. The project is intended to waste treatment of the I 131 production plant.

The waste composition is: Te isotopes, MoO_4^{2-} ions and I 131 residual in 5M H_2SO_4 solution.

At present the waste is stored to allow complete decay, but the risk of accident in handling, transport and storing associated to the space necessary to this, justify the study.

This project was developed in parallel with the study of chemical treatment techniques that search for a more secure storage form.

The design of the unit was made in a versatile form to be applied not only to this waste but also to other types of liquid wastes existing at IPEN-CNEN/SP.

One evaporator/crystallizer unit with 7 ℓ/h capacity was designed as a "two body unit" with two external heating system.

Special attention was given to the system of collecting the solid product. It was designed to act as a container of the solid waste.

In addition it avoid the contact of waste with cement or other package used.

This part of the evaporator/crystallizer unit is attached by a locked pneumatic system that guarantees the unit to be watertight.

When necessary, the design foresee the substitution of this part by a fixed valve to collect concentrated products.

Considerations about waste composition, waste collection, construction materials and process security necessary to establish a reliable project are made. The flow-sheet process and drawings of the evaporator/crystallizer unit are also presented.

INTRODUCTION

Among the liquid waste treatment techniques one of the most versatile for a high salt content is evaporation.

The choice of evaporation was based in a high decontamination factor achieved in the solvent which is, in most cases, the major waste fraction.

In some specific cases, like this, the evaporation can be made until salt crystallization.

WASTE FEATURES

The waste to be treated is produced during a I 131 hot-cell operation.

A more detailed explanation about waste arising was given in "Conditioning liquid waste from I 131 production by natural TeO_2 neutron irradiation" presented in this publication

The I 131 production is made in batch generating 1 ℓ waste/batch with about 259 GBq (7 Ci) in hot-cell outlet.

The waste composition is basically Te isotopes, residual I 131 activity and MoO_4^{2-} ions in 5M H_2SO_4 solution.

TREATMENT CONSIDERATIONS

Actually a common procedure is to store this waste in 30 ℓ polyethylene bottles inside 200 ℓ shielded drums, to decay about five years, followed by controled environmental dispersion.

The contamination risk in accident is high enough to justify a study aiming at a more secure form for conditioning waste.

The immobilization or evaporation without previous treatment was not considered due to volatile I 131 presence and high H_2SO_4 concentration.

The option followed was waste alkalinization until pH 11 to prevent I 131 volatilization and equipment corrosion.

The alternatives for neutralized waste treatment would be:

a) cement immobilization: in the neutralized waste there is a high concentration of Na_2SO_4 which is incompatible with cement where the limit for SO_4^{2-} incorporation is about 25 g/ℓ waste.

b) betuminization: a betumen matrix is limited for neutralized waste incorporation due to the hygroscopic salt content of the waste. The concentration limit is about 40% in weight and the obtained product is not secure form in water presence.

Thus, both techniques led to an increased waste volume that is not the objective of low activity waste management.

Considering the above exposed facts the option was the water evaporation until salt crystallization followed by storage in sealed containers for decay.

PROCESS DESCRIPTION

After each I 131 batch production the waste will be treated by the procedure presented in figures 1 and 2 which is described as follow.

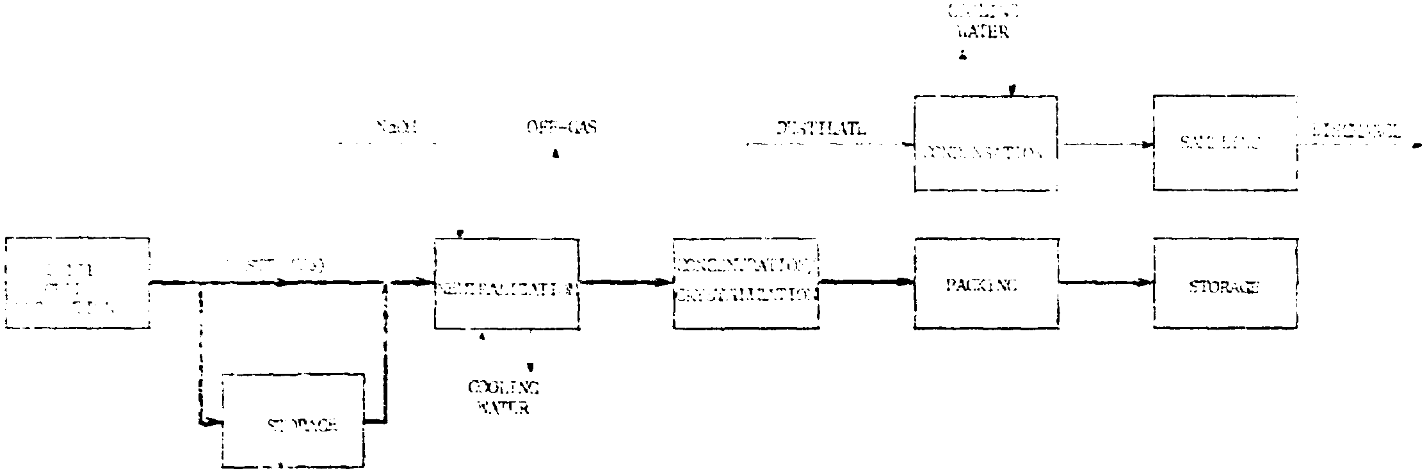


Figure 1 – Process diagram of the tellurium waste treatment

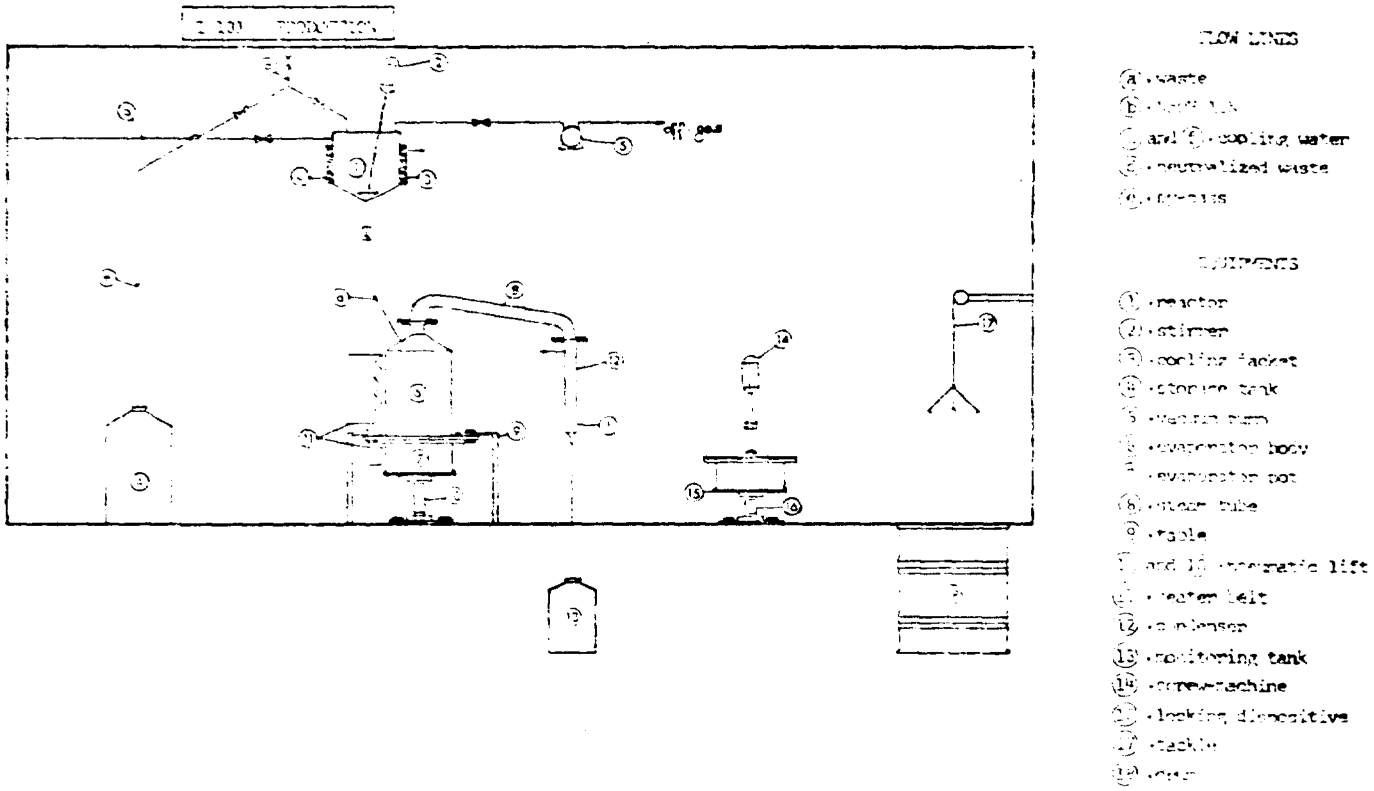


Figure 2 – Lay-out of a batch pilot installation for tellurium waste treatment

NEUTRALIZATION

The waste (a) produced by one batch, 1 ℓ, is sent to a reactor (1) with 5 ℓ capacity, where a 10M NaOH solution is added (b) to waste neutralization and alkalinization.

This reactor (1) is provided with a helicoidal mechanical stirrer (2). Since the H_2SO_4 neutralization is an exothermic reaction the reactor has an external cooling jacket (3) with water circulation (c).

After the reaction completion the waste (d) which volume is about 2 ℓ with 750 g salt content is sent to the evaporation unit.

There is a by-pass (e) to a storage tank (4) that may be used when the waste can not go directly to the evaporation unit. The turn back to normal operation is provided by a vacuum pump (5).

EVAPORATION/CRYSTALLIZATION

This procedure will be carried out with a 14 ℓ batch of neutralized waste in an evaporator at a 7 ℓ/h evaporation rate, until complete salt crystallization.

The evaporator has two main parts, made in AISI 304 stainless steel, named body (6) and pot (7).

The body has a cylindrical geometry with 20.4 cm of diameter and 45.0 cm height with two edge joint-flanged to connect the steam tube (8) and the pot to collect the crystallized salt.

This unit is supportted by AISI 304 stainless steel table (9).

The pot is a cylindrical container with the same diameter of body and 13.0 cm height. For each batch a new container is connected to the evaporator and pressed by a pneumatic elevating mechanism (10) until sealing with an "o" ring.

The necessary heating to 7 ℓ/h evaporation rate is given by two external eletrical belts (11) that act independently one of another.

The heating source control is made using two thermocouples, one for each resistance.

CONDENSATION

An heat exchanger (12) shell and tube type using water at room temperature as coolant fluid (f) makes the steam condensation.

The condensed liquid is then collected in a monitoring tank (13) and, after spectrometric analysis, is released to sewage.

SEALING AND STORAGE

At this point a crystallized salt, containing all waste activity, in about 50% of initial volume is ready to be sealed and stored.

The sealing system consists of a metallic (AISI 304 stainless steel) cap pressed against an "o" ring in a flanged joint pot.

This sealing is made with a screw-machine (14) and locking device (15) over a pneumatic elevating machine (16).

The sealed pot is then placed inside a 200 ℓ shielded drum (18) using a tackle(17) to do it.

A 200 ℓ shielded drum can receive 6 pots to storage and decay.

COMMENTS

This evaporation unit was designed to be applied in the treatment of wastes coming from the I 131 production. With minor modifications it is possible to use it for other liquid wastes.

INCINERATION FACILITY FOR COMBUSTIBLE SOLID AND LIQUID RADIOACTIVE WASTES IN IPEN - CNEN - SÃO PAULO

I. Z. Krutman, C. E. Grosche Filho, U. Chandra, A. A. Suarez

ABSTRACT

A system for incinerating the combustible solid and liquid radioactive wastes has been developed in order to achieve higher mass and volume reduction of the wastes generated at IPEN-CNEN/SP or received from other institutions. The radioactive wastes for incineration are animal carcasses, ion-exchange resins, contaminated lubricant oils, cellulosic materials, plastics, etc.

One of the advantages of the incineration process is that the resultant ashes are highly insoluble. This fact provides an extra security against contamination risk during subsequent handling and conditioning/immobilization step.

The optimization of the process was achieved by considering the following factors: selection of better construction and insulating material, dimensions, modular design of combustion chambers to increase burning capacity in future, applicability for various types of wastes, choice of gas cleaning system. The present design features include flexibility by use of multichamber for combustion and operation with controlled air (pyrolysis) and excess air. With this it should be possible to burn material more than the present capacity of 5 kg/h.

The electric heating adopted provides additional operational safety compared to gas or oil fired furnace, resulting further in the reduction of volume of off-gases to be treated.

The off gas system utilizes dry treatment and consists of one cyclone, electrostatic precipitador, condenser, activated carbon filter followed by one HEPA filter. For exhaust, a vacuum pump of 300 m^3/h flow and 1000 mm-H_2O pressure drop was selected. The operation is designed to function with a negative pressure of about 30 mbar to avoid escape of radioactive gases. In the case of failure of exhaust, accidental release is avoided by the adequate height of the chimney which provides the required depression.

The radionuclides most frequently found in the burnable wastes of IPEN are ^{131}I and ^{99m}Tc. The activated carbon filter provides a retention efficiency of 99.9 % for gaseous ^{131}I. The solid wastes under consideration are of IAEA category 1 with exposure rate of <200 mR/h.

The incineration facility has been installed and operational tests for the determination of, heating and cooling rate; the time required to reach the operating temperature (1000° C) and burning capacity, have been concluded. Inactive tests using animal carcasses (rats) resulted in a burning rate of 2.2 - 2.7 kg/h.

Isokinematic sampling devices as specified by the Environmental Protection Agency - US have been installed to analyse particulate material in the off-gases.

The facility is expected to be ready for semi-continuous operation in the second semester of 1986.

INTRODUCTION

Among the radwastes generated from activities in IPEN - CNEN/SP, about 80 to 90 wt % as for example, cellulosic materials, plastics, cotton, small animal, organic liquids, paper, rubber and others can be considered suitable for the incineration treatment. At. present, the procedures being implanted are: storage until radioactive decay, for the wastes containing short-lived radionuclides and compaction into 200 liters steel drums. In the first case, after decay, wastes are released to the environment as normal waste and in the second they are stored.

In order to provide a better treatment aiming at higher volume reduction factors and improving the safety of the stored wastes, a waste incineration system has been worked out since 1983. Wastes transformed into ashes are more easily and safely stored and transported. Incineration is an adequate pre-treatment for combustible wastes contaminated with long-lived radionuclides or having high radiotoxicity and for wastes requiring immobilization.

In the present work the technical and economical considerations which led to the design of the facility are described.

WASTE MANAGEMENT SCHEME

Before the start of operation of the incineration facility it is intended to implant a careful sorting of the solid and some liquid wastes.

The following criterias were adopted to select the wastes which will be incinerated:

1. Wastes contaminated with long-lived radionuclides (T 1/2 > 60 days);

2. Materials of high chemical toxicity;

3. Wastes having large volumes and susceptible to combustion;

4. Perishable wastes;

5. Wastes of such a nature that the release of which is hindered by environmental requirements.

CONCEPT OF THE FACILITY

The incineration facility consists of the following equipments (figure 1):

1. Two modular furnaces: the first is the primary waste burner and the second is a post-burner for the off-gases;

2. Off-gas treatment system: a high efficiency cyclone, an electrostatic precipitator, a gas cooler/condenser, a HEPA filter and a charcoal filter.

3. Exhaust system: a vacuum pump and a stack.

The furnaces used for primary combustion and post-burning are of mechanical modular type. This main characteristic permits an horizontal or vertical furnace installation as a simple way to increase the burning capacity.

The projected throughput for this facility with a typical waste composition is 5kg/h.

The modular type furnaces shown in figure 2 consists of:

- a cylindrical combustion chamber of 280 mm inner diameter and 1200 mm height made in refractory steel AISI 310.
- thermal isolation by special fibrous material type Fiberflex. It permits that the outside wall temperature be at 50°C (maximum).
- incinerator chamber shells in carbon steel.
- furnace body of carbon steel (AISI 1020) cylinder with 980 mm diameter and 1200 mm height.
- electric heating using silicon-carbide elements at 17 kV for operational temperature range of 900 to 1000°C. The power supply is controled by thermo-couple elements.
- two gates located in the furnace body to permit the inspection of the thermal isolation and the carbide elements.
- two primary air inlets located in the bottom of the combustion chamber below of the grate.

The furnace is loaded from the top by means of a manual batch feed system consisting of a carbon steel chamber (2 liter capacity) with double door to protect the operator from flame, thermal radiation and radioactives releases.

A cylindrical discharge pipe is located in the upper part of the post-burner furnace. This allows the gaseous effluents to enter tangentially to external secondary air in the cyclone in order to promote cyclonic motion.

The ashes are collected in as intermediate cooling chamber with double security doors to prevent release of radioactive ashes.

The operational conditions of the feeder and ash collector permit a semi-continuous process. The off-gas cleaning is executed before the liberation to the environment. For this purpose a dry off-gas system was utilized. The first stage consists of a cyclone separator. This provides an economical pre-treatment to reduce the particulate loading specially for the larger particles (ϕeq > 10μm). The design value of the efficiency of the particulate removal is 98%.

The second stage consists of an electrostatic precipitator to remove aerosols from the off-gas. The ionization of the particles is realized by charged stainless steel needles and the collection occurs in an aluminium plate. The design value of the collection efficiency for submicron particles is 99.9%

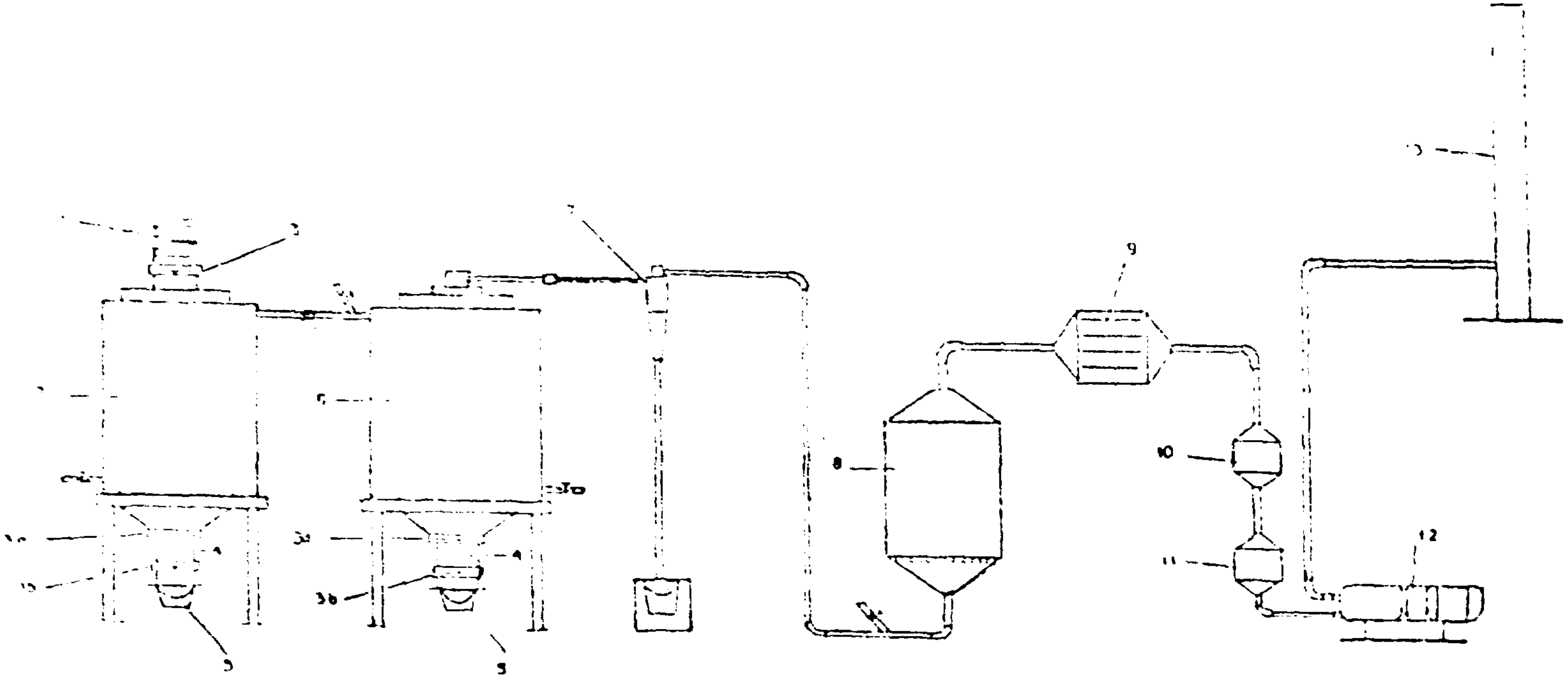
With these equipments the particulate from the off-gas are collected almost completely.

A condenser is provided to remove the moisture and eventual tritiated vapor from the off-gases. The dry off-gases are then directed to a HEPA filter (dimentions 600 x 600 x 350mm). It is used as an additional particulate collector to guarantee even higher filtration efficiency.

The volatile radionuclides, specially iodine, are removed by sorption in an activated carbon bed (600 x 600 x 350 mm) with > 99% efficiency. A secondary purpose of this bed is elimination of the odours.

The exhaust for the incineration process is executed by a vacuum pump. It creates a depression (-30 mbar), in the combustion chamber in the off-gas line and in the treatment train. The power rating of the pump is 7.5 HP and output 300 m^3/h.

The final liberation to the environment is made through a stack of 300 mm diameter and 16m height. The height of the stack was designed to guarantee enough depression in the incineration system even when there is a fault in the exhaust system.



1- Feeder
2- Incinerator furnace
3- Air-proof valve
3(a)-Air-proof valve-ashe collector
3(b)-Air-proof valve-ashe discharge
4- Intermediate cooling chamber
5- 20 L ashe drum
6- After-burning furnace
7- Cyclone separator
8- Eletrostatic precipitator
9- Condenser
10- HEPA filter
11- Active carbon bed
12- Vacuum pump
13- Stack

Figure 1 — Incineration system for the treatment of combustible wastes: solids and liquids

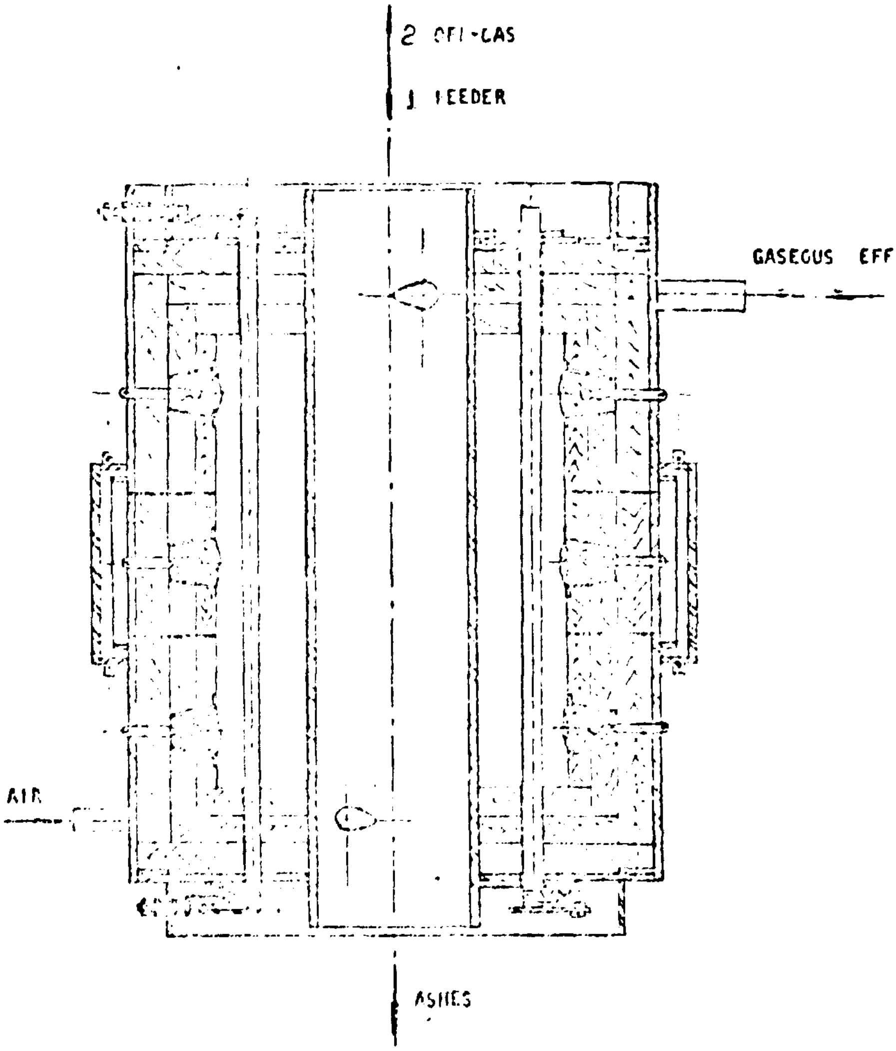


Figure 2 – Modular furnace (cross section)

COMMENTS

The design proposed for the incineration facility at IPEN-CNEN/SP for the burnable radwaste aimed to attend the following requisites: greatest safety, flexibility of the process, high combustion and off-gas, cleaning, efficiency and use of nationally available materials and equipments.

The facility was designed to minimize the off-gas generation by electrical heating which also enhances the safety.

The modular furnace coupling permits a higher flexibility insuring complete combustion or pyrolysis with higher burning capacity.

The double door system for the feeder or ashes collector guarantees operational safety conditions.

In addition to the immediate benefit of the incinerator facility i. e. a considerable reduction of the combustible wastes generated in IPEN, the most important benefit of it will be to serve as experimental incinerator for training, to design, construct and operate incinerator facilities for other nuclear installations in the country.

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VOLUME REDUCTION AND TOLUENE RECUPERATION FROM LIQUID SCINTILLATION WASTE

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ABSTRACT

The crescent application of liquid scintillation counting, in radioimmunoassay with typical composition of PPO, POPOP, toluene and a surfactant, has led to development of a treatment technique aiming to reduce waste volume.

The water soluble ^3H sample is homogeneized in a 1:10 ratio with the organic cocktail.

After counting the mixture, considered then waste, it is usually stored for further treatment.

Because of the inherently small radiological hazard from tritium the current method of disposal, when in small quantities, is dilution and dispersion. However in this case due to the presence of toluene, a carcinogenic agent, the most used technique of treatment is incineration which is quite costly.

This paper describes the experimental work undertaken to evaluate the recuperation of the liquid scintillator by NaOH treatment and also a distillation process to recovery only the toluene.

Recovered toluene was compared with scintillation grade toluene in terms of efficiency and background counts at the IPEN - CNEN/SP laboratories as well in a radioimmunoassay laboratory. Both of them certified the quality, of the recovered toluene for radioimmunoassay.

The volume reduction attained decreases significantly the costs of treatment for the remaing wastes beyond the profit of the toluene recovered.

All procedures including equipment drawing and results are reported.

INTRODUCTION

The growing use of liquid scintillators for radioimmunoassay hormonal tests, which make use of ^3H, ^{14}C or ^{125}I as radioactive tracers, are producing an increasing volume of radioactive waste.

Besides widely composition almost every solution uses toluene, dioxane or xylene as solvent in about 65% vv of the total scintillation cocktail. After sample dilution and counting the cocktail is discarded as waste.

The average annual volume of used scintillation liquid received at IPEN is around 3000 ℓ with 74 MBq (2 mCi) of total activity. About 1950 ℓ of them are toxic and rather high cost chemical reagent.

Because of the small radiological hazard, the current method of treatment, when in small quantities, is dilution and dispersion taking care that chemical and radiological discharge to the environment are observed.

However, when great volumes are involved, the most used technique is the incineration when it is available. Special tricks have to be used in order that the long - lived radionuclides be trapped in the filters or high dillution be practiced. Anyway this treatment procedure is costly. Both applied techniques led to reagent loss.

Having this in mind a study was conducted aiming to decontaminate and recycle the cocktail reagents.

Besides the large use of liquid scintillation cocktails, there is only few papers (1,2) in the litterature describing treatment techniques aiming cocktail or solvent recovery.

WASTE COMPOSITION

The scintillation cocktail composition used in several radioimmunoassay labs were verified and the average composition utilized is:
PPO (5 g), POPOP (0.5 g), toluene (666 ml) and triton X - 100 (333 ml).

The aqueous sample containing the radioisotope (e. g. ^3H) is then diluted in a 1:10 ratio with the scintillation cocktail, counted and afterwards discarded as waste. Usually in polyethylene bottles.

EXTRATION PROCEDURES

The first assayed technique was NaOH addition to the waste aiming at total scintillation solution recovery by phase separation.

This was made as follow:

- solid NaOH addition to the wasted liquid scintillation cocktail.
- magnetic stirring by 20 minutes.
- rest of the solution to phase separation.
- collect the organic phase for decontamination factor evaluation.

Different amounts of NaOH were used with 50 ml of scintillation samples. The obtained results are presented in table I.

Based on those results a new step was introduced in the procedure in order to enhance the decontamination factor. The organic phase was percolated through a silica-gel column to retain the aqueous phase. That experiment was done using only 1.5 g of NaOH with the 50 ml scintillation sample. The obtained results are shown in table II.

In spite of the decontamination factor increase this procedure was not used anymore because the organic phase becames cloudy after each percolation probably due to emulsion breaking.

DISTILLATION

The distillation of the cocktail was also assayed as a technique to recover the solvent.

The tests were made with simple distillation, reflow and bi-distillation followed by condensation, phase separation and toluene grade evaluation.

Table I

Decontamination factor and efficiency as a function of NaOH quantity

NaOH (g)	0.5	1.0	1.5	2.0	2.5	3.0
D. F.	1.66	1.76	1.91	1.48	1.43	1.38
% Ef	39.68	43.27	47.67	32.59	30.25	27.29

Table II

Decontamination factor and efficiency obtained with silica - gel percolation

	Before silica - gel percolation	Number of percolations			
		1	2	3	4
D. F.	1.91	2.31	2.55	2.89	*
% Ef	47.67	56.64	60.77	65.44	*

* The scintillation solution became cloudy

Table III

Data obtained with scintillation solution prepared using the recovered and A.R. toluene

Toluene used in scintillation solution	Counts (cpm)
A. R. toluene	31.9
recovered by simple dist.	38.0
recovered by reflow dist.	32.5
recovered by bi - dist.	36.3

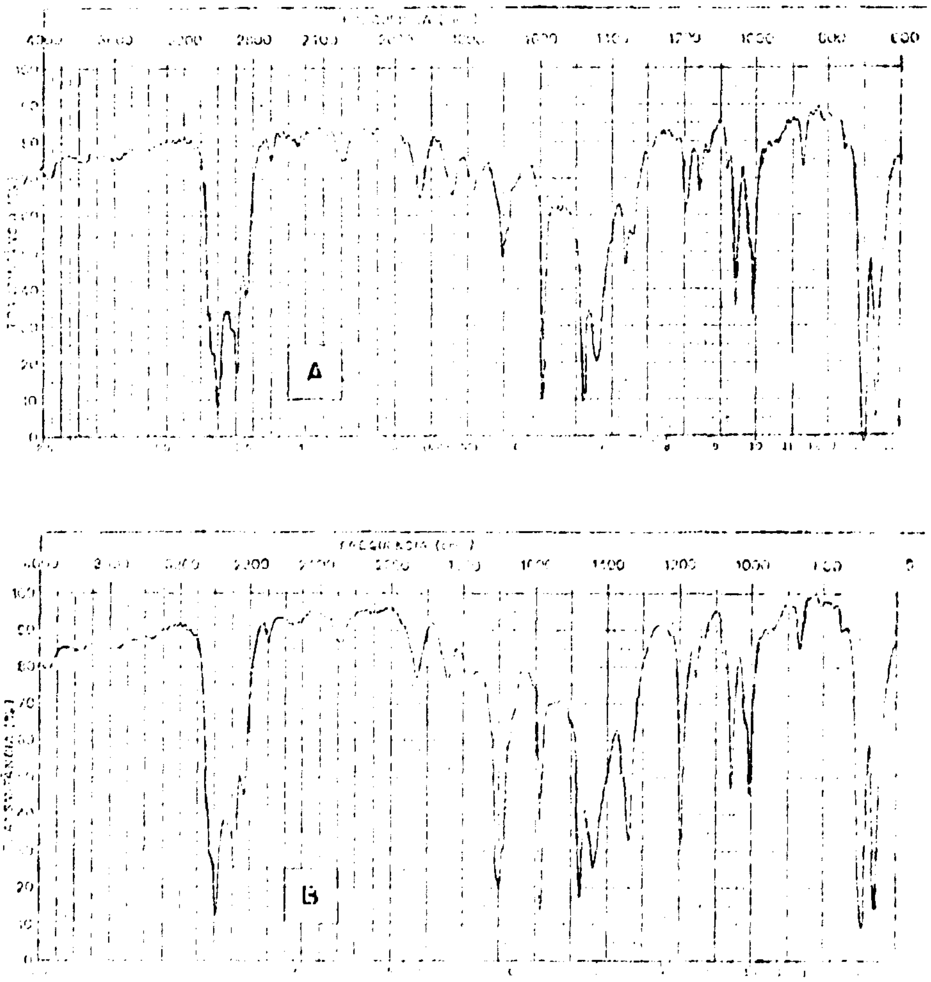

Figure 1 – (A) Standard toluene infrared spectrum
(B) Recovered toluene infrared spectrum

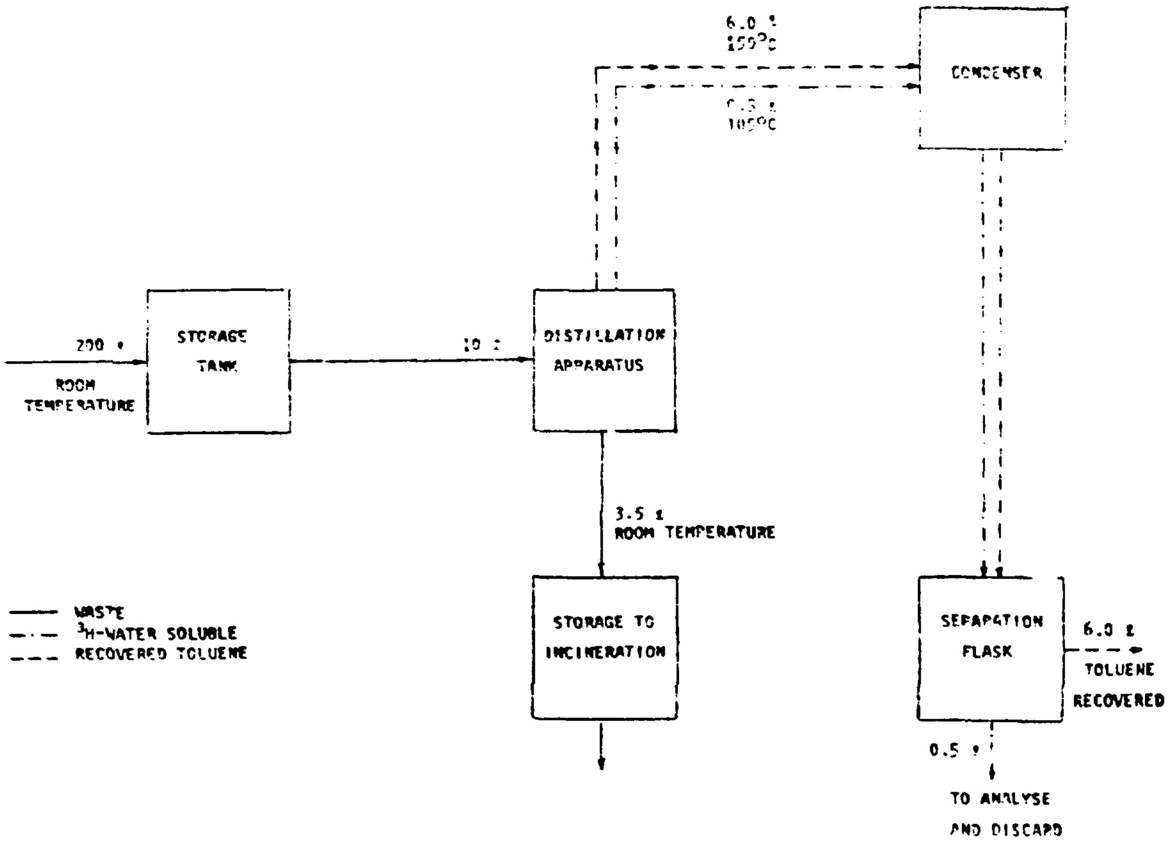

Figure 2 – Process flow diagram of the toluene recovery

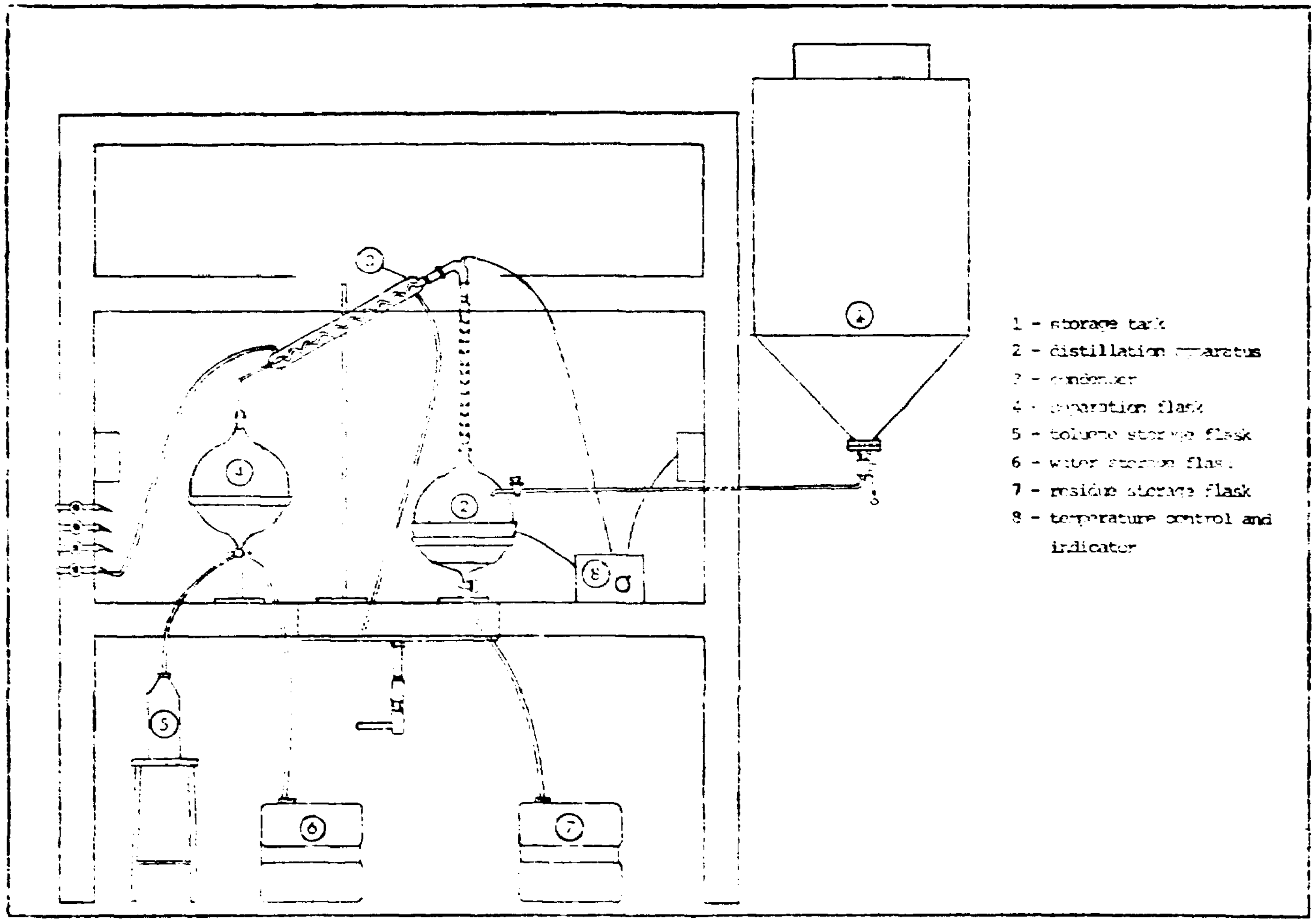

Figure 3 – Lay-out of a 20 ℓ batch pilot plant for toluene recovery

The phase separation is necessary due to the azeotropic mixture formed between toluene and water at 85° C. The recovered toluene was compared with standard scintillation cocktail using Analytical Reagent (A.R.) grade toluene and the obtained results are presented in table III.

Among the three distillation procedures the reflow distillation was considered the best one because of easy operation and contamination hindrance.

The toluene chemical purity was assured by infrared spectrometry analysis. Figure I shows the recovered and A. R. grade toluene infrared spectra. The recovered toluene was also used by radioimmunoassay labs which certified the quality of it for that purpose.

Figures 2 and 3 show the process flow diagram and the lay-out of a 20 ℓ batch pilot installation for toluene recovery.

CONCLUSION

The distillation technique used for toluene recovery is shown as an attractive pre-treatment to be used with liquid scintillation wastes. Besides, the toluene recovery reduces the amount of radioactive waste to be treated and reduces also the scintillation cocktail cost.

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CRITERIA AND EVALUATION OF SHALLOW LAND BURIAL SITES

Umesh Chandra

ABSTRACT

The criteria for shallow ground disposal of radioactive wastes as developed by Nuclear Regulatory Comission-US and *International Atomic Energy Agency* are discussed. Emphasis has been given to: elaboration of various basic components of a national strategy for radioactive waste disposal; development of technical requirements; environmental standards, consensus among scientific community and public confidence.

Aspects of site selection, evaluation/characterization and site suitability are dealt with. Some results of characterization of a site for temporary storage of low level solid radioactive wastes in Instituto de Pesquisas Energéticas e Nucleares - IPEN - CNEN/São Paulo are described. The methods used for hydrological characterization include use of artificial tritium for *unsaturated zone and radioactive tracers like* I - 131, $Cr^{51}Cl_3$, $Cr^{51}EDTA$, Br^{82} for the saturated zone.

The use of stable isotopes for surface and groundwater investigations and use of non-absorbing radioactive tracers together with field value of time transformation/retardation factors for prediction of radionuclide migration around shallow disposal site is recommended.

INTRODUCTION

Shallow ground disposal of solid low and intermediate level wastes may be suitable provided that their radioactivity decays or released to acceptable levels within the time the institutional control of the site is expected to be maintained. The category of wastes which may be disposed include wastes of category (IAEA) IV and III (with engineered barriers).

Suitability of a particular waste type would ultimately be decided by national regulatory authority on the basis of the safety assessment. The components of a national strategy for radioactive waste disposal may vary among countries depending upon the national objectives, but the basic components should include, among others, the following:

1. establishment of national laws and regulations according to the internationally accepted standards/criteria and recommendations, taking into consideration the socio-politic realities and expectations of the country. These regulations should also outline relationship between the various bodies, viz the regulatory body, *the waste transporters, waste generators and* the disposal site operators-*herein called the technical community.* Matters of national policy such as the limiting of exposure levels, institutional control periods, de minimis levels, should be developed within the framework of regulations along with the means of assuring financial responsibility for long term care.

2. development of suitable criteria, methods and assessment techniques for performing safety

analysis.

3. development of plans for surveillance over the period of institutional control. Certain surveillance data are needed before the construction of the repository for comparing with data collected during operational and post operational phases.

Criteria

Experiences of ground disposal of radioactive wastes in many countries, have now resulted in technically sound criterias and safety analysis methodologies for safe disposal of the wastes in shallow/deep ground. These criterias elaborated by Nuclear Regulatory Commission of the U.S. (26, 25) and International Atomic Energy Agency (9 16) based on recommendations of International Commission on Radiological Protection (11) are available as guidelines for application for new sites in developed countries and the first sites in developing countries. The comparison between the criteria developed by various agencies have been discussed in an earlier literature (27, 28, 29).

The site selection process used by any applicant may vary considerably due to a wide variety of factors such as geographical, diverse geology, hydrologic, meteorologic, climatic, ecologic and socioeconomic settings and provisions on site. However, the site selection process involved in each site should share the same basic steps.

The first step will consist of defining the region of interest. The second step will consist of screening the region of interest to identify potential sites. In the third step the potencial sites will be screened against a common set of criteria, including the minimum technical requirements (26) and the environmental standards (25). The third step involves a coarse screening process which will identify candidate sites for more detailed review.

The fourth step will consist of a detailed review of the candidate sites. The difference between the steps will be the depth of detail and inclusion of additional items to be evaluated in each step. For example, the fourth step may include items such as conceptual designs, preliminary cost estimates, release scenarios and pathway studies which were not included in the previous steps. The fourth step will conclude with the selection of a preferred site from among the candidate sites.

It is in the site selection process that the site suitability requirements play their first and most important role. The potencial sites must pass the screening tests before receiving serious consideration. The screening process need not separate the useful sites but discriminate the not-useful sites. The major portion of characterization studies will be performed for the preferred site after selection from among the candidate sites. Only if detailed site characterization studies identify unanticipated adverse conditions at the preferred site, would detailed investigations (16) be performed at more than one site.

Analysis of the past experience with several shallow ground repositories showed that many of the difficulties that resulted in inadequate containment of the wastes could be attributed to the improper siting. A number of problems listed in literature (10) can be avoided if appropriate criteria are applied in the site selection process. It is difficult to define a site with all favourable attributes in a region of interest, and hence attention should be paid to the waste form, radionuclide content and repository design in order to provide the overall system with more engineered barriers to compensate for the less favourable characteristics of the site. Some important aspects of site selection process are briefly discussed in the following description.

Climate

Arid zones are favoured when selecting a site for shallow ground repository. Water management (run-off, safety against flood, erosion) is a critical component in obtaining good performances of the repository system. Sites in arid climates do not generally need an elaborate surface water management system. However, even at arid sites short heavy rainstorms can result in local flooding and erosion (10). Dykes and ditch system should be considered for flood control.

Topography

In order to avoid the risk of flooding, sites should be located outside flood plains in topographically higher areas. Disposal below water table may be acceptable provided engineered barriers prevent any long term contact of ground water with waste or if groundwater movement is exceedingly slow (of the order of 10^{-8} – 10^{-6} cm/s) (10). In order to minimize erosion by surface water, sites should have minimum topographical relief.

Geology and Hydrology

Geohydrological aspects and criteria of ground disposal have been extensively discussed in literature (3, 22, 6). Additionally, the Earth Science Technical Plan (US) (24) or Geoscience Technical Plan (Canada) (1, 21, 23) serve as useful guides for elaborating a national plan for shallow ground disposal (although they are intended for deep repositories).

Favourable performance of site can be expected when water table is reasonably deep, unsaturated zone has low porosity and moderately low permeability, underlying strata has high sorption properties and groundwater flow is of the order of few centimeters per day . If the site does not meet geohydrological suitability requirements, it should be constructed only if appropriate engineering design features are incorporated into the repository and waste form and operational procedures are safety oriented.

A few of the principles of site selection and geohydrological criteria followed in Soviet Union (10) recommended that the minimum depth of the water table below trench bottom should be 4m and the minimum permissible distance from a surface water body be 500m and seismic areas with occurrences at or above force 7 on the 12-point Richter scale should be avoided.

The need to locate the repository within the bounds of a nuclear facility can be understandable mainly from the operational point of view but it could have a long term implications in differentiating impacts from normal releases from the nuclear facility/plant and possible releases from neighbouring waste repository site into the same recipient environment.

Surveillance

Surveillance program should:

- be designed on the basis of safety assessment
- comply with licensing controls
- provide early warning before radionuclides cross the site boundary
- detect any failure of the system for remedial action

- involve measurements of radionuclide content, radiation dose rate, surface contamination in conjunction with disposal options and site practices/operations

- collect data to facilitate prediction of radionuclides via possible pathways

- validate previous safety assessment

- reassure and promote public confidence

Disposal of radioactive wastes is a complex system and safe disposal is controlled by a number of factors of the system. In practice we do not find anyone in charge of all the system. Many factors are controlled by waste generators while others are under the control of the site operators-all of them guided/controlled by regulators. The regulatory authorities impose conditions and limitation on the classification and types, the quantities and the level of activity etc. of the waste that may be disposed off at the repository site. The technical criteria for a safe disposal establish minimum technical requirements (26) for each component of the system. The technical requirements deal with site suitability, design and operation of the site, waste classification/characteristics, site closure and institutional controls. Each of these technical components contribute to the system to meet the overall performance objectives.

Successful performance of a shallow land burial facility is the integration of the various phases of the activity (ie. the site selection, site design and development, operations and closure) to ensure the most cost effective achievement of the performance objectives. A systematic approach should be used in predicting site performance, considering both the characteristics of the wastes to be disposed and the characteristics of the site itself. This performance assessment establishes the basis for design, development and operation. It also provides a useful tool in auditing the performance of the facility.

The requirements for the site suitability should ensure that the overall performance objectives for migration, long term maintenance/stability and intrusion protection are met. Stability of the disposal site is the single and most important aspect and is directly related to the performance objectives.

The site suitability requirements must be:

- based on sound technical footing

- reasonable to implement operationally

- workable in identifying sites which are suitable

- widely discussed among the technical community to achieve consensus (for example between waste generator, disposal site operator, licensor/institutional controller).

A consensus among the technical community greatly facilitates the licensing process. This consensus set of site suitability serves as guide for future applicant and allays public concern and moderates intervention. On the contrary, public concern and intervention appear to be fueled by disagreement and controversy within the technical community.

The final phase in site selection involves an optimization of the radiation protection provided using cost/benefit analysis consistent with ICRP recommendations.

This requires calculations of the expected collective radiation doses from the use of each site. The information that will be needed to perform this safety analysis includes:

- scenario analysis ie. identification and definition of process (retention or leaching) or event that could result in release of radionuclides and their transport from the site to man. Cost of the waste form(s) and conditioning option (s) chosen.

- protection from water, barriers and cost of these

- evaluation of pathway. Arrival of radionuclides and their quantities of release using all pertinent data of geology, hydrology and ecology of the site.

- analytical methods or models for predicting the activities of radionuclides leaving the site and their impacts in terms of collective doses

- comparison of these results with established acceptability criteria.

A sensitivity analysis should be made to define uncertainty and variability in parameters used to perform the safety analysis involving geological and environmental features etc.

Some 300 to 400 sub-routines/system codes have been developed to calculate radionuclide pathways for various cases of repositories-ranging from fairly simple one dimensional to complex 2 or 3 dimensional codes. In many cases, simplifying hypotheses and conservative assumptions have been made to reduce enormous complexities of hydrogeological systems which are further complicated by the many radiochemical and biochemical processes that influence the behaviour of radionuclide in an aquifer.

The methodologies adopted in codes can be a combination of deterministic and probabilistic approaches. A simpler deterministic code may be used in preliminary assessment and as the project develops towards final safety assessment, the codes tend to be more complex and probabilistic in nature in order to calculate radionuclide pathways mathematically taking into account the sensitivity analysis.

Few guidelines to assist in selecting or developing a code for a new repository have been discussed in the recent document prepared by the IAEA. The point to highlight is that in many of the codes migration calculation is based on distribution coefficients which are only valid for equilibrium conditions. In practice this condition is rarely met and therefore attention should be paid to the relative velocities of radionuclides verified by laboratory or field tests rather than to distribution coefficients themselves. Efforts made in the proper selection, design and development of a repository help to avoid costly remedial actions at a later date.

Having reviewed the criteria of shallow land burial of radioactive wastes I like to give a brief description of the studies carried out in IPEN for geohydrological characterization of the site of temporary storage of low level solid wastes produced in the Institute. These studies were carried out as per requisite of National Nuclear Energy Comission-CNEN for licensing the temporary storage of low level solid radioactive wastes inside area of IPEN-São Paulo. The studies carried out and the techniques employed for geohydrological evaluation/characterization, are for our site for temporary storage but are recommended for a site of CNEN where the radioactive wastes will ultimately be disposed off.

For hydrological evaluation it is recommended that, use of tracerslike deuterium and oxygen-18 and radioisotopes like KBr^{82}, $Cr^{51}Cl_3$, $Cr^{51}EDTA$, $Au^{198}Cl^3$, NaI^{131} and reactor produced tritium may be used whenever possible. The stable isotopic tracers furnish almost a fool-proof method of investigation of mixing of ground and surface waters (5) more so when a repository is near a surface water body. The radioactive tracers furnish information about hydrological parameters of the unsaturated and the saturated zone. The parameters of unsaturated zone ie. upward/downward movement of soil moisture, rate of rainfall infiltration at a shallow waste repository can be studied best by artificial tritium tracer (4). The determination of velocity and direction of groundwater should be made in and around the site frequently so as to cover the precipitation cycle of the region. This data should generate representative velocity and direction maps (7) indicating minimum and maximum values observed during the one-year precipitation cycle-evidently excluding off-normal (flood) conditions. The use of radioactive tracers for determination of travel/arrival times of water/waste radionuclides either in laboratory models or in the field give the in-situ values (taking into account the geological heterogeneity) of the parameters used for computing migration rates of the waste radionuclides. Significant work of radionuclide migration in laboratory models in the field is now being reported from GSF/IfR (19, 20, 18) after generating fundamental data for many years (17). The use of non-absorbing type radioactive tracer for describing hydrodynamic dispersion and the use of in-situ time transformation factor/retardation factor (2, 8) could be recommended for predicting migration of waste radionuclides in the field.

The sub-surface geology of our site consists of the following:

- quartenary alluvial sediments with clay, thickness of this layer is variable within 10m
- tertiary fluvial sediments of São Paulo basin thickness of which varies from 40 to 150m, thickness decreases in the western direction
- precambrian crystalline basement with occurrence of cracks and fissures between 70 to 120 and 250m
- the water table aquifer containing the rainfall infiltration consists of the first 10 to 15 m of the sediments

The local topographical contours vary from 750m to 735m sloping towards the Pinheiros canal. The site is at sufficiently high topographical level to take care of flooding. Average annual rainfall is about 1300 mm and evaporation about 850 mm.

Rainfall infiltration rate (297 cm/y) was determined by tritium labelling technique. Ground water velocity (max. 46.1 cm/d) and direction (to north) was determined by various radioactive (^{82}Br, ^{131}I, ^{51}Cr) tracers using single well techniques. The earliest arrival time of groundwater to Pinheiros canal from the site is about 1209 days (ie. without considering absorption and dispersion). There is no potable use of groundwater in the vicinity. The superficial water also is not potable up to about 20kms.

The safety margins via aquatic pathway are quite high. The site is under routine environmental monitoring through various groundwater monitoring wells constructed around it. Further studies required for evaluation are in progress.

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LEACHING OF NUCLEAR POWER REACTOR WASTES FORMS

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ABSTRACT

The waste management research program foresees a number of tests that can provide data for characterization and quality assurance of the final radioactive wastes forms and its safe disposal from the human environment. In this way, leaching tests are developed and achieved in order to understand the mechanism of radionuclides loss from immobilized wastes and to predict its long-term performance under storage and disposal conditions.

This paper describes the leaching tests for immobilized power reactor wastes carried out at IPEN. These wastes forms consist mainly of spent resins and boric acid concentrates solidified in ordinary Portland cement. All tests were conducted according to the ISO and IAEA recommendations. Three years leaching results are reported. The cesium diffusivity coefficients determined out of these results are about 1×10^{-8} cm^2/s for boric acid waste form and $9 \times 10^{-[illegible]}$ cm^2/s for ion exchange resin waste. Strontium diffusivity coefficients found are about 3×10^{-11} cm^2/s and 9×10^{-11} cm^2/s respectively.

INTRODUCTION

The leachability of radionuclides from solidified wastes forms is a significant data to evaluate the suitability of a given material as a safety mean to immobilize radioactive wastes, to risk analysis assessment when it concerns to radionuclides release into the environment and also to predict long-term performance of the waste form under disposal conditions.

Cement is widely used by the nuclear industry to conditioning intermediate and low level radioactive wastes, because it presents some advantages as low cost, radiation and thermal stability.

In the present work, within the on going program of wastes forms characterization, leaching from radioactive cement-based wastes forms containing specifically simulated reactor wastes were determined. Intermediate and low level reactor wastes consist mainly of evaporators concentrates and spent ion-exchange resins. For a PWR type reactor these concentrates have a high boric acid contents, which may vary between 90 to 260 g/kg of concentrates.

The procedures for leaching tests and data analysis were conducted according to the IAEA and ISO recommendations (1, 2). Since it is desirable to know the mechanisms of radionuclides release, in order to predict or extrapolate from short term data the behavior of the far future, experimental data were statistically analysed according to some equations found in the literature (3).

EXPERIMENTAL

Evaporator Concentrates Immobilization

As it is well known boric acid has a strong retarding action on the cement setting. This problem can be solved by using some accelerating additives such as sodium silicates or by neutralizing such action on the cement by transforming the boric acid into a neutral chemical form.

The direct neutralization of boric acid with sodium hydroxide leads to the tetraborate form, which precipitates at pH 9. It can be avoided by heating the solution to approximately 70° C, but the cement setting doensn't occur at short time. Hence the addition of a calcium salt or calcium hydroxide is convenient to form a calcium metaborate which has no retarding effect over the cement setting.

To prepare the leaching test samples, simulated evaporator concentrates were made, by neutralizing a boric acid solution (120 g/kg concentrate) with sodium hydroxide until pH 8. The solution was then heated to 70° C and added to a cement/additives/vermiculite blend. The additives consisted of setting accelerators as calcium cloride, sodium carbonate and barium hydroxide. Due to its adsorption capacity, a clay (vermiculite) was used to improve the cesium retention. The whole composition of the borate wastes forms is given in table I.

The cement used was the ordinary Portland cement, and the water/cement (W/C) ratio was 0.42. To all samples were added as radioactive tracers cesium-134 and strontium-90.

The experimental conditions for the leaching test were as follows:

– Samples are right cylinders with 5 cm diameter by 10 cm height, resulting in a volume to exposed surface of sample (V/S) equal to 1.

– Curing time was 28 days in a sealed atmosphere, i.e., 90-100% relative humidity.

– Leaching was conducted at room temperature, with renewal of leachant which was 1600 cm^3 of distilled water and synthetic sea water whose composition is given in table II.

Ion - Exchange Resins Immobilization

Cemented samples containing simulated spent ion-exchange resins were prepared according to the compositions shown in the table III.

The experimental conditions were the same for evaporators waste, unless the leachant. In this case leaching was investigated only in distilled water.

Comparison with Actual Reactor Waste Cement Immobilized

In order to compare leaching behavior between actual and simulated wastes, it was made some samples containing actual reactor evaporators concentrates. It distinguishes two compositions of waste-forms: without vermiculite and with vermiculite. See table IV.

The activity measurements of the leachant were made by using NaI(Tl) well type gamma detector for cesium-134 and liquid scintillation detector for strontium-90.

RESULTS AND DISCUSSIONS

Up to now leaching time has reached 3 years for simulated wastes and about one year for the actual waste. As recommended by IAEA, leach results are reported as a cumulative fractional release ($\Sigma\ a_n/A_0$), corrected for the volume to surface ratio as a function of the total elapsed leaching time ($\Sigma\ t_n$) up to each sampling time. Leached fractions of cesium and strontium to distilled water and sea water from evaporators concentrates wastes are plotted in figure 1 . Figure 2 and 3 show the results obtained for spent resins wastes forms and actual reactor wastes. The comparison of both, simulated evaporators concentrates and actual wastes is shown in the figure 4.

Since diffusion seems to be the main process of the radionuclides loss from immobilized cement-based wastes forms, it can be characterizing the leach behavior according to the mass transport equation solution for a semi-infinite medium (3) as follow:

$$\frac{\Sigma a_n}{A_o} \cdot \frac{V}{S} = 2\left(\frac{D.t}{\pi}\right)^{1/2} \qquad (1)$$

Where V is the volume and S surface of the specimen. D is the diffusivity coefficient or leach coefficient, which characterizes leach behaviour of mobile species. Linear least square regression was used to fit experimental data to the equation in order to determine D.

Table I

Composition of the cemented evaporator concentrates

Compounds	% Weight
Cement	61.25
Vermiculite	3.0
Waste	30.15
Additives (*)	5.5

(*) 2% calcium cloride, 1.5% barium hydroxide and 2% sodium carbonate

Table II

Synthetic sea water composition

Compounds	Concentration (g/l)
NaCl	27.213
$MgCl_2$	3.807
$MgSO_4$	1.658
$CaSO_4$	1.260
K_2SO_4	0.863
$CaCO_3$	0.123
$HgBr_2$	0.076

pH = 8.1

Another approach to determine D was attempted using the diffusion equation with concentration dependent dissolution rate, as described by Godbee (3). In this equation was introduced an analysis by error function (erf) expressed as:

$$\frac{\Sigma a_n}{A_o} \cdot \frac{V}{S} = (K.D)^{1/2} \left[\left(t + \frac{1}{2R} \right) \text{erf } (K.t)^{1/2} + \left(\frac{t}{K.\pi} \right)^{1/2} e^{-K.t} \right] \qquad (2)$$

Where K is dissolution rate constant.

The diffusivity coefficients obtained from first model are listed in table V. It can be observed that strontium has release rate smaller than cesium by a magnitude factor of 2, even for the specimens that contain vermiculite as additive, it was demonstrated that vermiculite has a deleterious effect on the

Table III

Spent resins wastes - forms composition

Compounds	% Weight
Cement	63.3
Waste (*)	32.3
Additives (**)	4.4

(*) Wastes with 20% strong cationic resin-IRA 120

(**) 1.1% barium hydroxide, 1.65% calcium cloride and 1.65% sodium carbonate

Table IV

Composition of actual reactor concentrates immobilized into cement.

Compounds	Without clay % Weight	With clay % Weight
Cement	58.8	53.8
Calcium hydroxide	6.0	6.0
Waste (*)	35.2	36.2
Vermiculite	–	5.0
W/C ratio	0.53	0.54

(*) Wastes arising from Germany reactor Stade gmBH.

strontium leaching. The higher the vermiculite load the higher is the strontium release (4), also the mechanical strenght decreases with increasing of the vermiculite load.

The analysis of this waste has resulted:

pH = 7.6
d = 1.130 kg/m^3
Solid content: 165 kg/m^3 (mainly boric acid)

Isotopic analysis:

Isotope	Isotopic %
Co-60	32.26
Sr-90	14.79
Y-90	14.79
Sb-125	2.65
Te-125	0.53
Cs-134	1.90
Cs-137	16.54
Ba-137m	16.54
U-238	4.3 E-04

Table V

Comparison of the effective diffusivity constant calculated by transport equations (model 1) in distilled water and seawater leachant medium. D(cm^2/s)

Isotope	Simulated waste form		
	Borate		Resins
	Distilled water	Seawater	Distilled water
Cs-134	9.61×10^{-9}	2.05×10^{-8}	8.28×10^{-9}
Sr-90	3.75×10^{-11}	7.92×10^{-12}	8.24×10^{-11}

In table VI are presented cesium diffusivity constants calculated by means of equation 1, for simulated and actual borate wastes.

Statistical analysis by the second model demonstrated a good fitness for the experimental data of strontium, as its can be seen from table VI, but it cannot be concluded the same for cesium, whose experimental data don't fit adequately to the model. The simple diffusion process cannot explain satisfactorily the leach mechanisms from these forms. This seems to occur after some elapsed time when the water has already penetrated the cements pores. At the first few days the behavior may be due to an initial surface wash-off, on the other hand, at long leaching times, the dissolution rate of the matrix takes

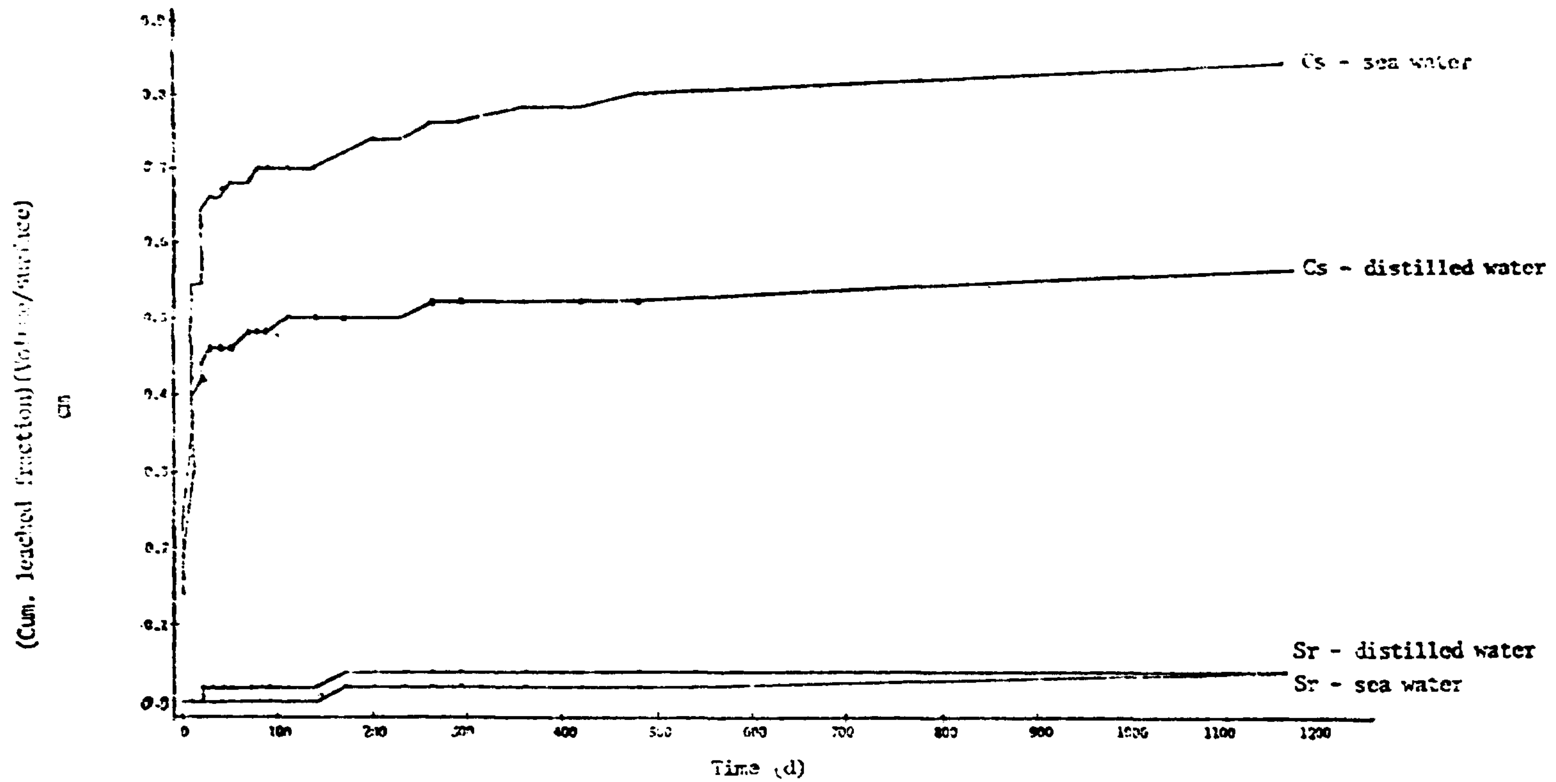

Figure 1 – Cesium and strontium leaching from cemented borate waste-forms, in distilled water and synthetic sea water, at room temperature

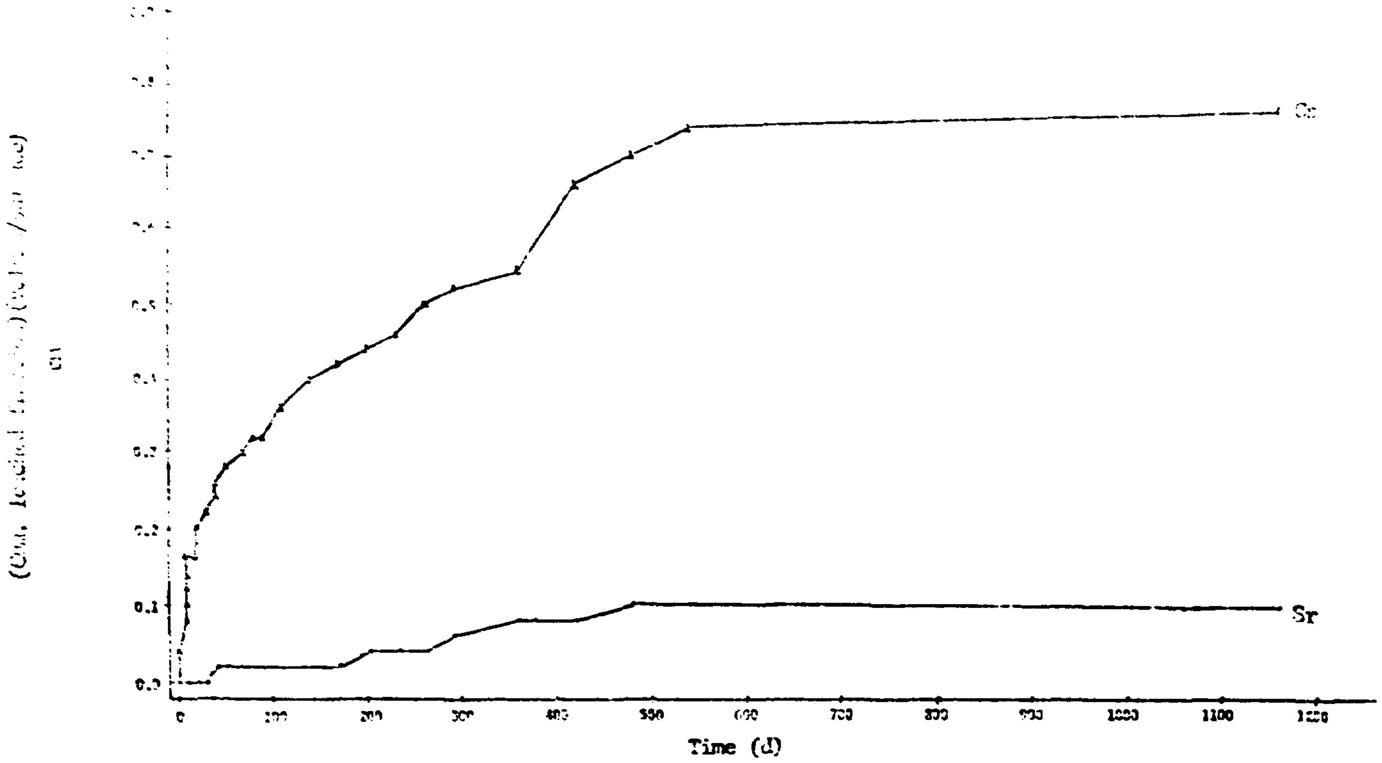

Figure 2 — Cesium and strontium leaching from cemented spent-resins waste-form, in distilled water at room temperature

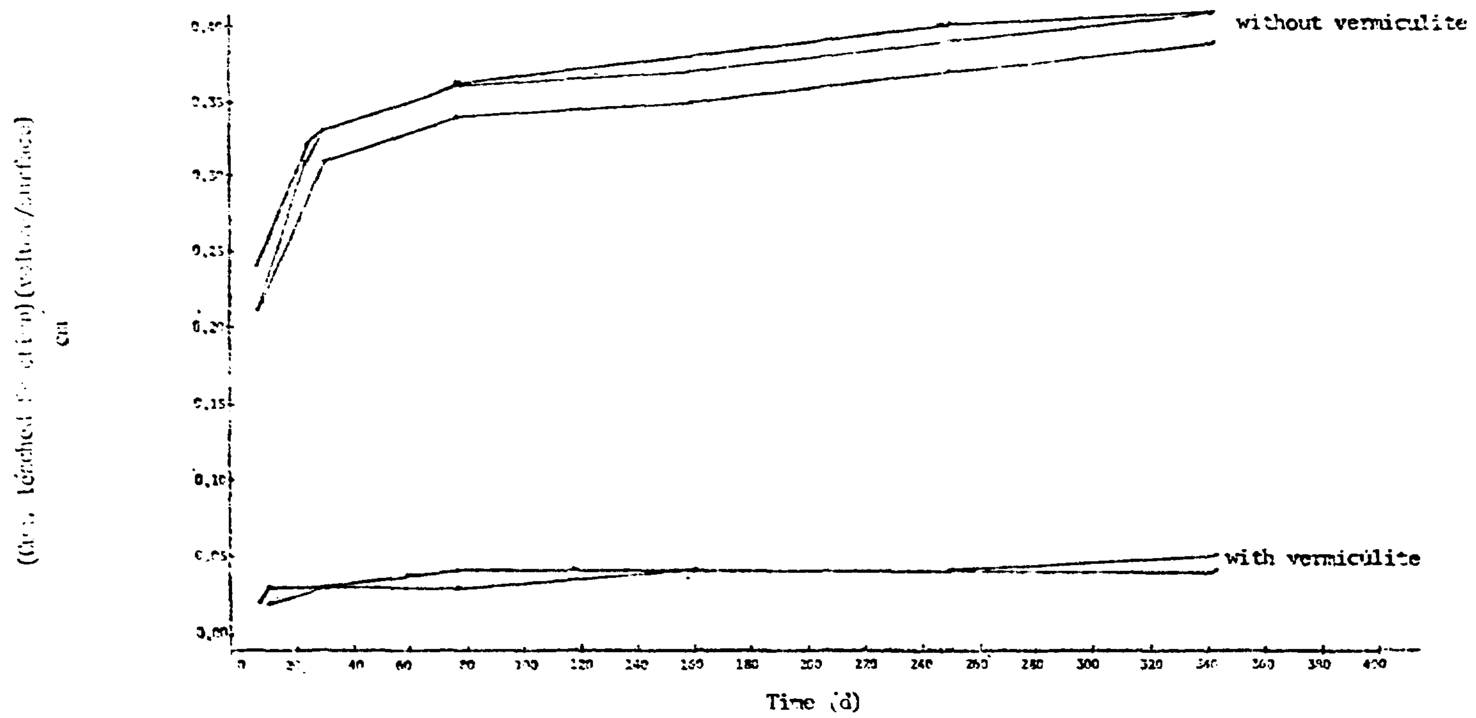

Figure 3 – Cumulated leached fraction of cesium from actual cemented waste

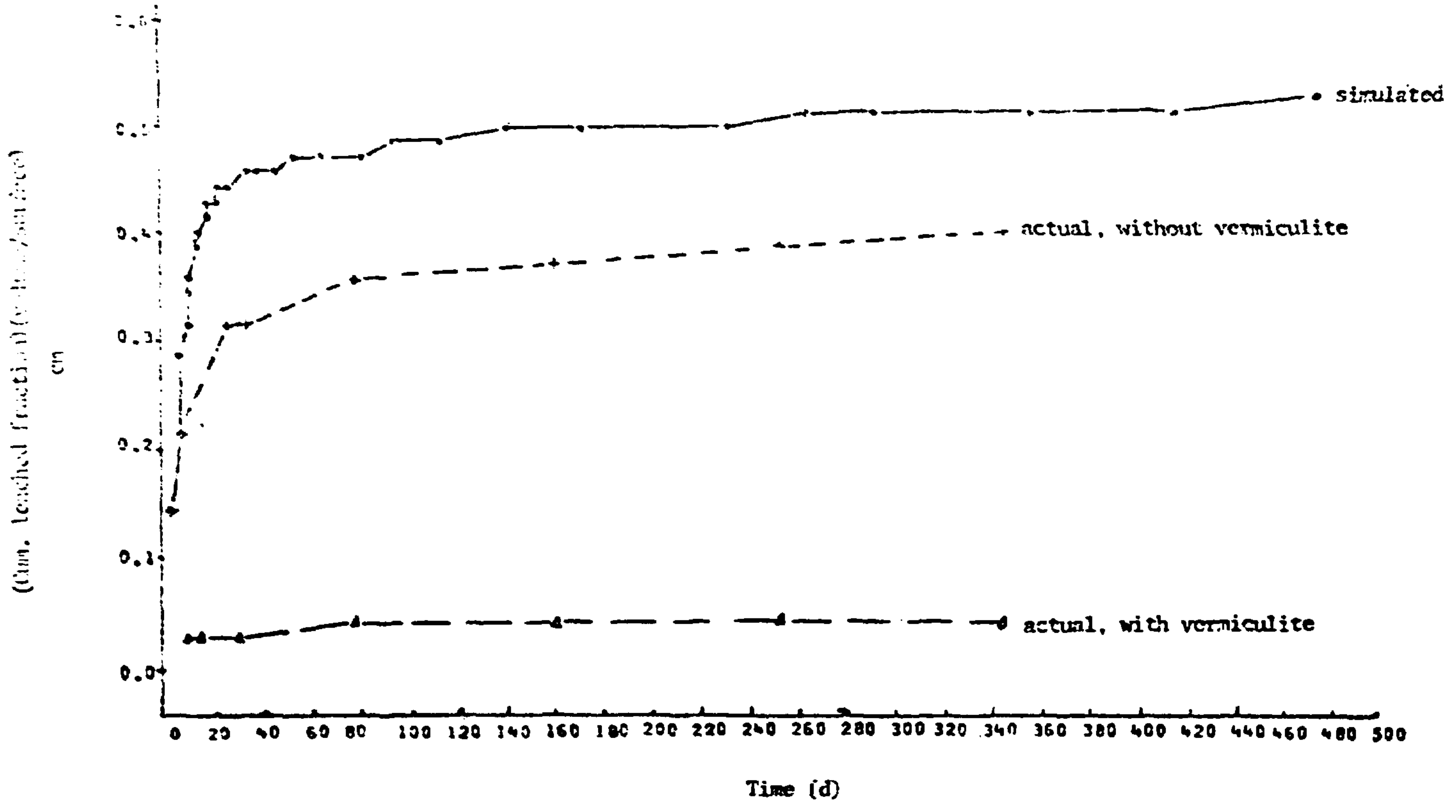

Figure 4 – Comparison between cesium leached fraction from actual and simulated cemented wastes

place as a predominant factor. So, to model the kinetics of the leach mechanisms for wastes forms containing additives, it must be taken into account another factors that contribute to the leach rate. Reactions as ion-exchange or retention by clay or ion-exchange resins between cement-additives, cement-wastes, waste-additives are some of those reactions that may occur, other than the interactions between whole waste form of the surrouding aqueous medium. Thus it becomes necessary to determine quantitatively each of these contributions and the degree of the modifying factors that should be introduced to compose a kinetic model for a quantitative approach for the leaching behavior.

More accurate laboratory investigations on the impact of some of these interactions on the leach rate is the purpose for a future work, in order to obtain adequated data that could be helpful to the comprehension of leaching mechanisms.

Table VI

Effective cesium diffusivity constant calculated by transport equations (model 1) for actual and simulated borate waste.

Simulated	Actual	
(cm^2/s)	With vermiculite (cm^2/s)	Without vermiculite (cm^2/s)
9.61×10^{-9}	1.56×10^{-10}	2.54×10^{-10}

Table VII

Effective diffusivity constant, D(cm^2/s) and dissolution rate constant, K(s^{-1}) calculated by transport equations (model 2) for strontium-90

Leachant	Simulated waste form			
	Borate		Resins	
	D (cm^2/s)	K (s^{-1})	D (cm^2/s)	K (s^{-1})
Distilled water	3.91×10^{-11}	7.02×10^{-9}	6.53×10^{-11}	1.24×10^{-8}
Seawater	6.74×10^{-12}	5.93×10^{-9}		

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EVALUATION OF SOLIDIFIED CEMENT WASTE FORMS

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ABSTRACT

This work is part of a program to get expertise in the treatment of low-and intermediate-level liquid wastes. Concrete has been suggested as a promising encapsulation or overpack material for long-term storage of radioactive waste from the nuclear fuel cycle, particularly in conjunction with the disposal of low-and intermediate-level waste.

This paper was conducted to investigate the influence of sodium nitrates, as a possible integrating component of waste, on some properties of national Portland cement.

The waste form properties, discussed in terms of their dependency on waste type and amount, include water-cement ratio - W/C, setting times, compressive strength and homogeneity.

Criteria to be applied in the assessment of the final solidified waste are proposed.

INTRODUCTION

The objective of solidification is to convert radioactive waste into a stable monolithic form, which minimizes the probability of radionuclide release to the environment during interim storage, transportation and final disposal. The solidified waste should be of such chemical, mechanical, thermal and radiolytic stability that its integrity can be assured over the time required for the decay of contained radionuclides to an acceptable level.

The setting of suitable criteria for the analysis of the waste forms depends on local circunstances and to the specific problem considered.

The present work analyses the influence of nitric waste streams on the mechanical properties of national Portland cement used as an immobilization matrix.

As a start point to this study it was not considered any pre-treatment of the waste prior to the immobilization except the neutralization with sodium hydroxide. For this reason the interval of salt content by weight was zero to 8% while W/C was kept between 0.30 and 0.40, by weight, in order to keep a reasonable workability.

One of the most fundamental physical property required for any kind of immobilized waste form is the homogeneity which is quite important in the solidification process and during long-term storage. It is starting point to specify and define the other physical properties and also some chemical one as density, porosity, leaching rate, degradation, permeability, compression strength, radiation damage, thermal conductivity etc which can not be studied if the matrix is not homogeneous.

So, in order to evaluate the homogeneity of the final product it was used the high sensibility of the delayed neutrons detection technique to measure the distribution of very small quantities of a soluble uranium salt or thorium oxide in powder form. The uranium salt was used to simulate the soluble radioactive wastes and the thorium oxide simulates all the insolubles which could be present as precipitates or suspension solids. Any kind of immobilization process aim to result in an homogeneous product. To evaluate its performance when compared to a standard one use can be done of this technique together with appropriate statistical tests. The advantages of this method (*) are the fast response and high sensibility associate with the needlessness of using radioactive tracers which could increase the waste generation.

The next property which is directly related with the immobilization process is the setting time.

According to the immobilization process devised the setting time either initial or final can be quite important.

In practice, the terms initial and final set are used to describe arbitrarily chosen stages of setting.

Experiments were made to observe the influence of sodium nitrate as well W/C on the setting time. At the studied extend there was none anomalous behaviour in setting time beyond the retardation effect.

The compressive strength of concrete waste forms is an important parameter in safety and accident evaluations. The final product must exhibit sufficient strength to survive accidents or long term stresses during handling, storage, transport and final disposal. The solidified product has to have sufficient resistance to avoid damages that can increase the superficial area and enhance the leach of radionuclides.

Normally, cement pastes have compressive strengths on the order of 60 to 70 MPa which are higher than the minimum required by the final repositories acceptance criteria. Portland cements generally develop ≈ 90% of ultimate strength after 28 days but the addition of sodium nitrate as simulated wastes to cement reduces greatly the compressive strength.

EXPERIMENTAL PROCEDURES

The cement pastes were prepared using Portland cement, CP320, whose average measured composition is : 58.4% tricalcium silicate (C_3S), 12.5% dicalcium silicate (C_2S), 9.85% tetracalcium aluminoferrite (C_4AF), 8.99% tricalcium aluminate (C_3A), 4% gypsum and 0.8% of insolubles. To it was added different solutions of sodium nitrates to simulate neutralized nitric wastes.

The mixture of all components was done by following the standard procedure given by ABNT-MB-1/79 (1). For that purpose it was employed a planetary paddle mixer with two speeds.

Homogeneity Tests

For the homogeneity evaluation of the final product two types of tracers were used: uranil nitrate-$UO_2(NO_3)_2$ used to simulate the soluble radioactive wastes and thorium oxide-ThO_2 used to simulate all the insolubles which could be present as precipitates or suspension solids.

The metallic can used as mould of the blocks for the cement matrix had cylindrical shape with diameter of 165 mm and 180 mm height.

After the cure time those blocks were disassembled and cutted into five parts, as shown in figure 1 . From them were extracted, with the help of a carbide drill, around ten samples in which uranium and thorium concentrations were determined.

(*) Patent Pending

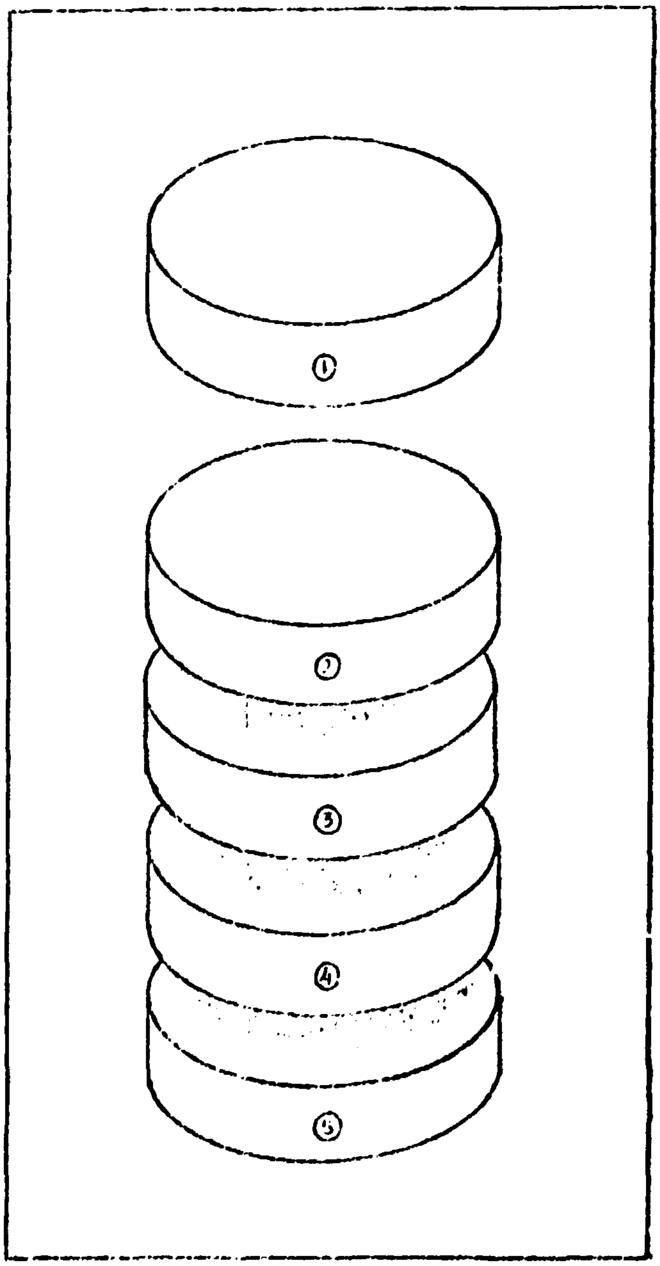


Figure 1 – Cutted cement blocks for homogeneity tests.

Those concentrations were evaluated by use of the delayed neutron counting facility existing at the Radiochemistry Division of IPEN-CNEN/SP (2).

Setting Time Measurements

These measurements were done by using the Vicat apparatus shown in figure 2. After pouring the cement paste mixture into the conical mould the initial time of setting was considered as the interval of time between the moment of contact of cement with the solution and the time when the needle released with $\approx$ 1.3 mm/s stops 1 mm from the mould bottom. Final set is obtained when the needle do not penetrate more than 0.5 mm from the top of the mould.

Mechanical Strength

The samples production for the mechanical strength tests of pastes followed also the standard ABNT-MB-1/79 and they were Ø 50 x 100 mm sized. All samples were prepared by using sodium nitrate solution with temperature of (23 ± 2) °C, pH $\approx$ 8 and sealed cure. The mechanical strength was observed for different cure times ranging from 3 to 360 days, for different W/C, and different percentages, by weight, of sodium nitrate.

For the mechanical strength tests a loading speed of (2.5 ± 0.5) $kgf/cm^2/s$ was used.

RESULTS AND DISCUSSIONS

Homogeneity Tests

The uniformity statistic examination of the uranium and thorium distribution in cement matrix was done by using the same criteria used to test the uniformity of pseudo-random number generators used in computers, that is , χ^2, Kolmogorov and Smirnov-Cramér-Von Mises tests (3).

The hypotesis of uniform distribution of experimental values of a given quantity must be rejected if the χ^2 value calculated out of the experimental values exceeds the upper limit $\chi^2_{\ell-1}(p)$ of the confidence interval, where p is the assigned confidence probability and $(\ell - 1)$ is the number of degrees of freedom.

Confidence probability p means a number close to 1 such that, if the hypothesis of uniform distribution is correct, the probability is p that the value obtained for χ^2 will not exceed $\chi^2(p)$. If however the confidence limit is exceeded, this means that the measure of discrepancy, χ^2, indicates a significant departure, and the hypotesis of uniform distribution must be rejected.

Extremely small values of χ^2 must be considered an indication of failure of randomness, since in the case of correctness of the hypotesis the probability for a random quantity to assume too small values is extremely small.

The statistic test is some measure of the "distance" between the experimental and hypothetical distribution function. For the Smirnov-Cramér-Von Mises test this is the average square difference between two functions and the Kolmogorov test takes the maximum or minimum difference. The hypothesis of normality is accepted at 100 p% level of significance if the computed value of the *statistic don't exceeds the 100 p% point of the null distribution of the statistic chosen.*

In this paper the homogeneity tests were realized in two ways: in one experiment the blocks were contaminated with uranium salt only while in the other the blocks were contaminated with both uranium and thorium compounds.

As the method is more sensitive to the uranium determination due to its higher cross-section, for those samples where both tracers were used it was necessary to increase the thorium content.

In table I are shown the results obtained in the homogeneity tests for blocks contaminated with uranium alone and both uranium and thorium. As can be seen in this table, both tested blocks satisfied all applied statistical tests.

Hence cannot be rejected the hypothesis that the observations come from a normal population with average value, $\bar{X}$, and standard deviation, $\hat{S}$, within a confidence level of 95%.

Setting Time Measurements

At table II are shown the results for the initial and final setting time obtained for different W/C and salt contents in cement paste. These data represents the average of at least four measures.

As can be seen from the data the effect of sodium nitrate as well the water content is to retard the setting time. From figure 3 is possible to observe that there is a linear dependence between the setting time and the W/C and salt content.

It is known that when the stiffening of the cement paste is developed external actions as movements of the cement container can introduce undesirable damage to the final cement block.

So, the knowledge of that dependence is important when taking decisions related to the cementation mixture process.

Before any cementation process care has to be taken in order to evaluate the caracteristic setting time of the cement used since it varies according to the lot utilized. Those variations can be so higher as 50%, as was observed.

Mechanical Strength

In figure 4 is shown the dependence of axial compressive strength with the salt content for different W/C obtained for 28 days cure time. Each plotted value is an average of 12 measurements.

The main purpose of these measurements was to establish the interval of values for W/C and salt content that satisfy the final disposal site acceptance criteria. Since the cement matrix provides nearly all the strength of the cement paste, increasing salt content decreases load-bearing area of the hydrated cement particles and so the compressive strength.

From figure 5, where it is shown how the compressive strength varies with time, is possible to see that the estimated final compressive strength varies according to the W/C and also with the salt content of the matrix.

The obtained dispersion of values for the compressive strength was around 12% so, in order that the 17MPa for the compressive strength in the final repository be observed with a 95% confidence level, the W/C has to be lower then 0.40 and the salt content not higher then 8% by weight.

Table I

Homogeneity statistic tests for samples * with U and Th tracers.

Sample	Average ± Standard deviation ($\overline{C}$ ± σ) (ppm)	χ^2 Test		Kolmogorov test		Von - Mises test	
		χ^2 Experimental	χ^2 (95%) Theoretical limit	$\sqrt{N}\,D_N$ Experimental	$\sqrt{N}\,D_N$ (95%) Theoretical limit	Nw^2 Experimental	Nw^2 (95%) Theoretical limit
With U	106.8 ± 5.7	1.225	5.991	0.47	1.33	0.08	0.46
With U and Th	$(68.5 \pm 5.8)_U$ $(1237 \pm 101)_{Th}$	1.068 0.404	3.841 3.841	0.63 0.58	1.33 1.33	0.04 0.05	0.46 0.46

* W/C = 0.35 $NaNO_3$ (% by weight) = 4%

Table II

Initial*(t_i) and final*(t_f) setting times for cement pastes with different W/C ratios and $NaNO_3$ percent by weight.

W/C	Salt content (wt %)							
	0		2		5		8	
	t_i (min)	t_f (min)	t_i (min)	t_f (min)	t_i (min)	t_f (min)	t_i (min)	t_f (min)
0,30	171 ± 14	238 ± 15	232 ± 17	327 ± 12	247 ± 27	371 ± 33	351 ± 23	444 ± 13
0,35	203 ± 9	311 ± 16	240 ± 16	361 ± 29	329 ± 21	452 ± 5	398 ± 8	522 ± 9
0,40	229 ± 22	379 ± 26	288 ± 25	433 ± 30	375 ± 13	500 ± 28	457 ± 15	621 ± 21

* average ± standard deviation

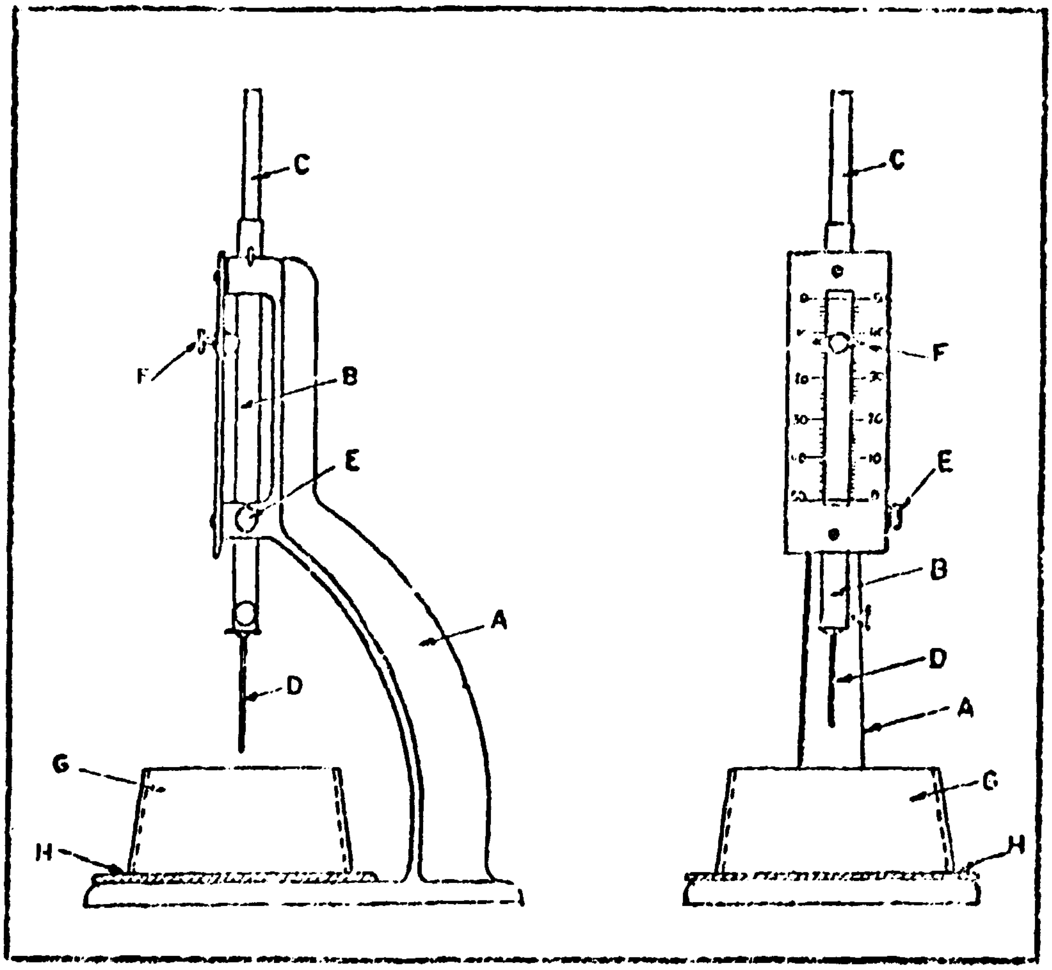


Figure 2 – VICAT Apparatus used for setting time measurements and consisting of : frame, A; movable rod, B, weighing 300g; one end, C, for consistency determination; other end, D, a removable steel needle for setting determination; set screw, E, for held the rod B in any desired position; adjustable indicator, F, which moves over a scale graduated in milimeters; conical ring, G; lucite plate, H.

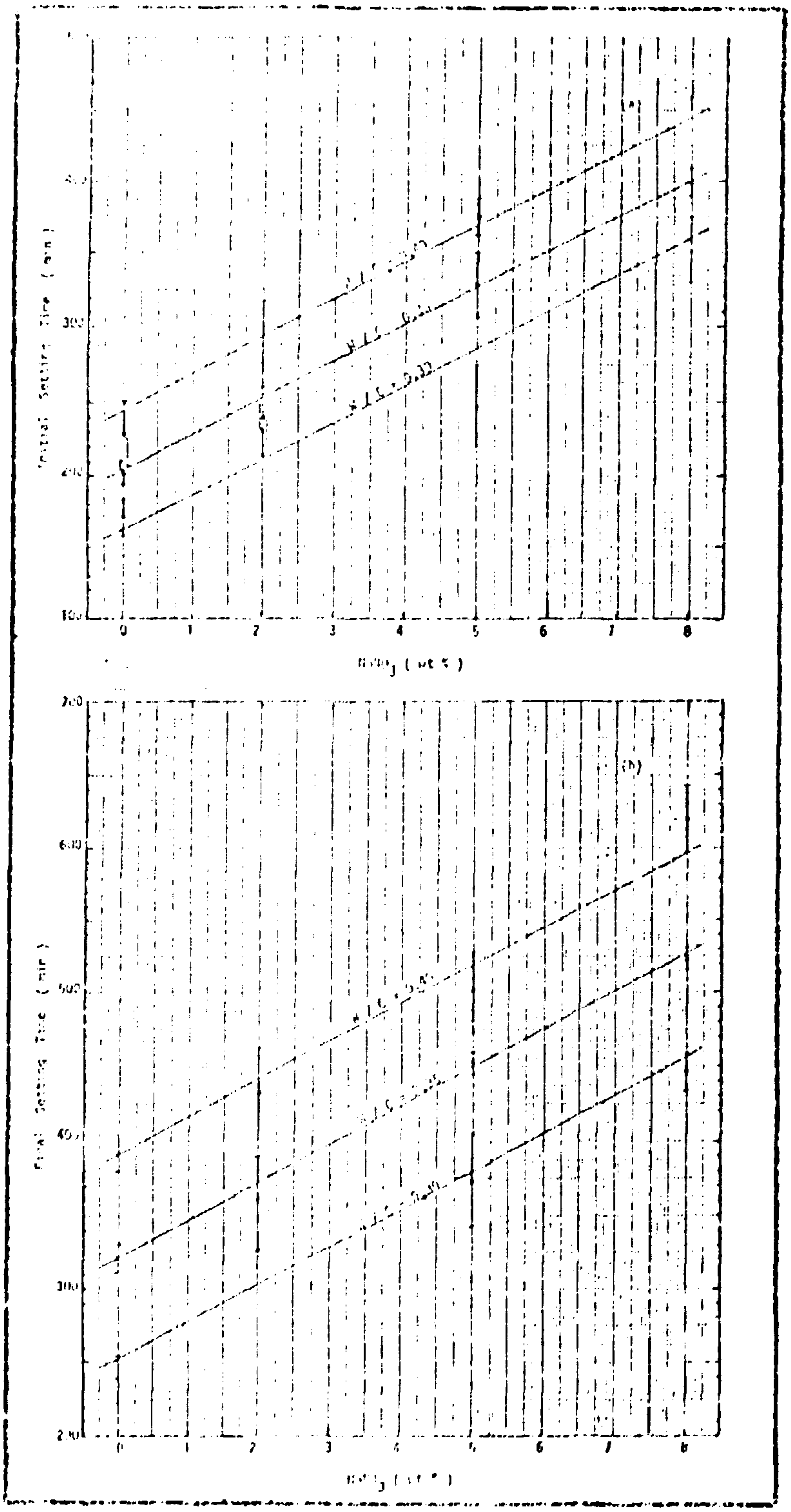


Figure 3 – Relation between initial (a) and final (b) setting times and salt contents for different W/C ratios

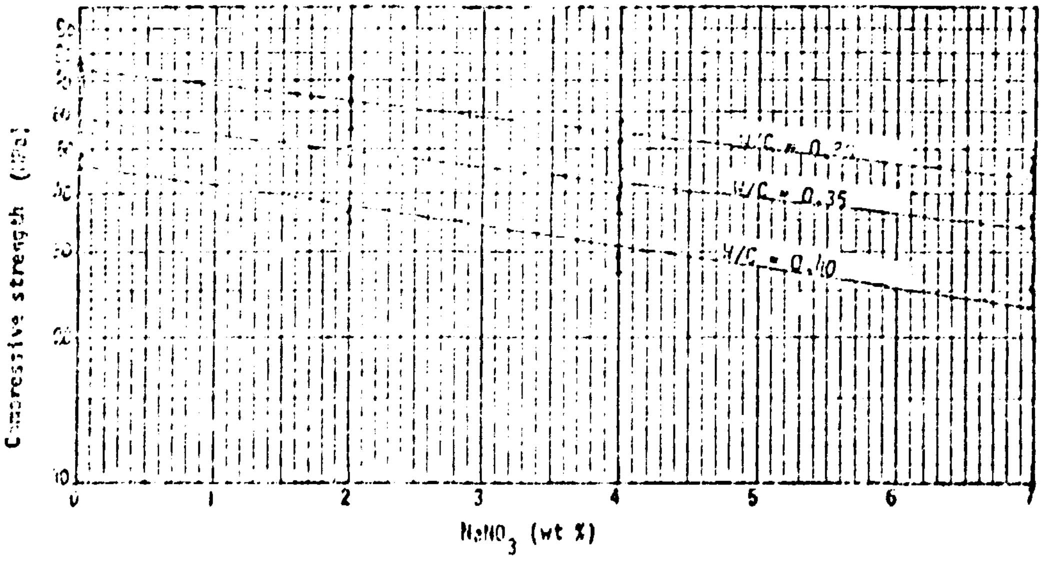
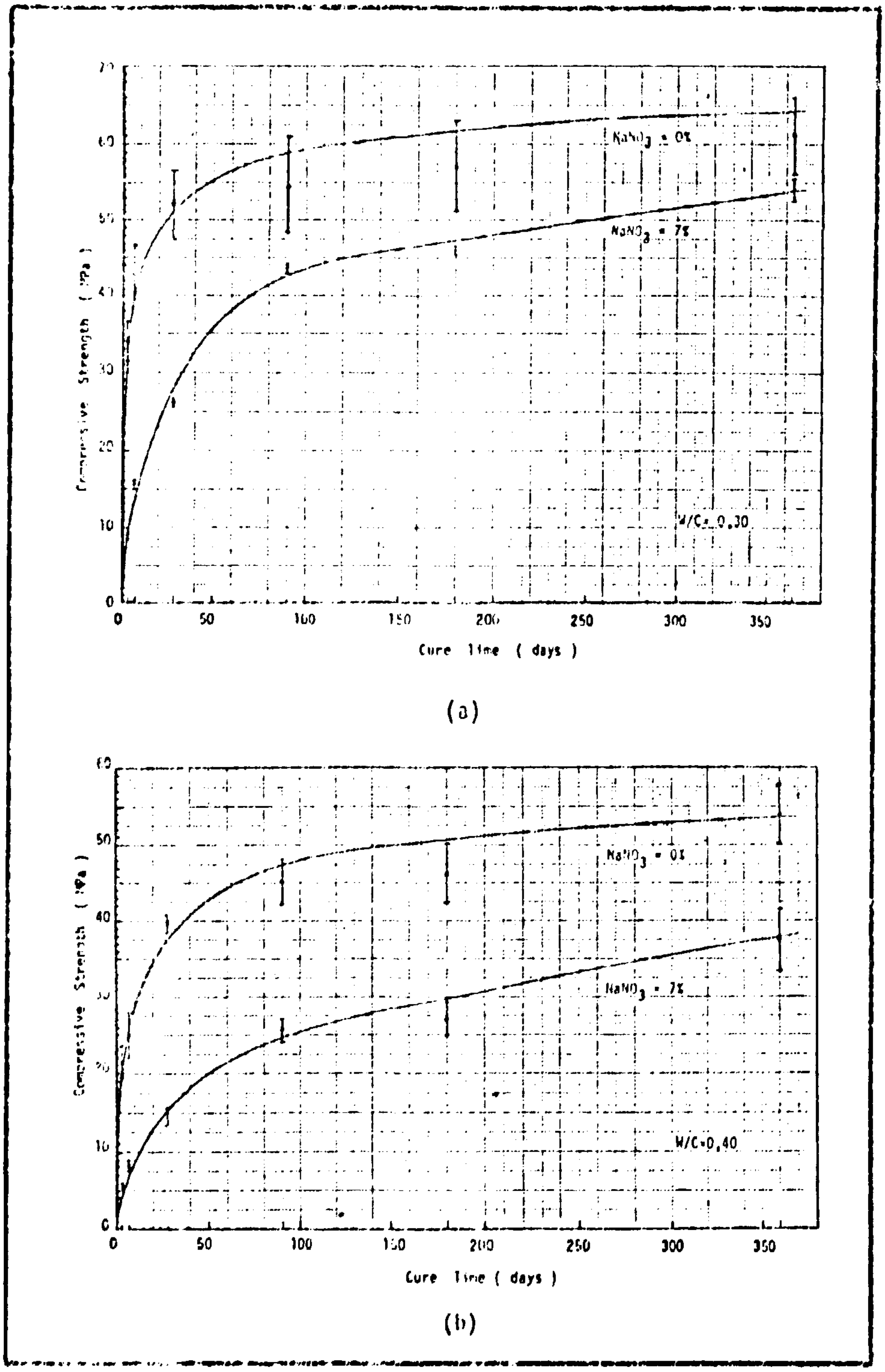


Figure 4 – Relation between compressive strength and salt content for different W/C



(a)

(b)

Figure 5 – Relation between compressive strength and cure time for: (a) W/C = 0.30, NaNO$_3$ = 0% and 7%; (b) W/C = 0.40, NaNO$_3$ = 0% and 7%.

CONCLUSIONS

It was studied some properties of national Portland cement aiming to qualify it as a potencial matrix material for incorporation of nuclear wastes. At the same time it was developed a process to evaluate the homogeneity of the solidified product. According to that process and the statistical tests by using the planetary paddle mixer, one can consider that the process results in an homogeneous product.

Within the range of parameters used it was possible to establish criteria to be followed in order to satisfy the final repository premise. Any way is recommended that for each new lot of cement some tests be realized in advance in order to keep confidence with the proposed compressive strength limits. On the contrary, special control for some other parameters are necessary to be done, i.e. cement composition, fineness, curing time etc.

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EXPERIENCE ON THE OPERATION OF A LOW-LEVEL SOLID WASTE TREATMENT FACILITY

H. Miyamoto, A. A. Suarez, R. Vicente

ABSTRACT

In the middle of 1982 a low-level solid waste compaction facility was commissioned to run cold tests aiming at licensing the facility to handle radioactive wastes. In August 1983 begun normal operation and wastes arising from IPEN and many other institutions have been treated and packed successfully since then.

The following radionuclides are present in the wastes reaching the facility: ^{3}H, ^{14}C, ^{24}Na, ^{32}P, ^{35}S, ^{42}K, ^{51}Cr, ^{60}Co, ^{82}Br, ^{89}Sr, ^{90}Sr, ^{99}Mo, ^{99m}Tc, ^{125}I, ^{125}Te, ^{127}Te, ^{129}Te, ^{131}I, ^{134}Cs, ^{140}Ba, ^{192}Ir, ^{198}Au, ^{226}Ra, natural U and Th, and other radionuclides in smaller quantities.

The compactible wastes are mainly paper, glass bottles, refused gloves, clothing, overshoes cans, plastic pieces, rubber, polyethylene bags, and others. An average volume reduction factor of 4 has been achieved with the 10 tons hidraulic press used. Up to now, about 144 cubic metres of solid compactible wastes were received, yielding about 180 drums of compacted waste.

Non-compactible wastes are also received and are mainly wood pieces, metal scrap, contaminated soil, ion-exchange resins, debris from dismantling and decontamintion operations. These wastes were encapsulated in 200 liters drums, the voids being filled with cement grout. Resins and soils were mixed with cement in a concrete mixer and poured in the drums. About 8 drums have been filled with non-compactible waste since the operation started.

The modifications on the original installation aiming at improving the performance of the facility are described. These include modifications on the baling press shaft, exchange of the original epoxi-based paint in the floor of the building, modification on the original concept for the storage site, and others.

The procedures for the waste management and the associated radiological safety procedures are detailed.

Up to now, despite some incidents, no contamination of personnel have been occurred.

INTRODUCTION

The existence of the low-level solid waste treatment facility whose operation is described in this paper became necessary with the natural increase of the institute activities in the nuclear field and to its attendance to community. A detailed description of the facility is presented in other paper of this seminar (IAEA-SR-110/22).

Before the installation of this facility the amount of waste material was small and had concentrations below the limits indicated by the federal radiological regulations. The applied treatment technique was to delay and decay prior dispose to the environment. Since then it was desired to get experience in waste treatment techniques so, in 1983 the compaction facility begun operation and in the beginning of the next year a small incineration plant fo radioactive wastes will start to burn cavy carcasses and others combustibles solid wastes.

A lay-out of the treatment facility is shown in figure 1.

Experience accumulated, so far, indicates that 95% of solid waste to be treated have dose rates below 2 mSv/h (200 mR/h), and only 5% have higher ones.

The solid waste produced at IPEN and arrising from other institutions, whether combustible or non-combustible, was mostly reduced in volume till now by simple compacting directly in the waste drums. Although compaction results in volume reduction factors of only 4:1, it is characterized by low operating costs.

Solid radioactive wastes are produced mainly during cleaning and decontamination activities in the form of rags, paper, cellulose, plastics, gloves, clothing, overshoes etc. Laboratory materials such as cans, polyethylene bags, glass bottles as well as bulky exhaust air filters which are contaminated by the activity adhering to dust particles and aerosols also contribute to the solid waste inventory. Most of this solid waste is low-level.

A small parcel of non compactible long lived wastes are also produced and received for treatment at the facility. They are wood pieces, metal scraps, exhausted sealed radioactive sources, defective components and tools, debris from dismantling and decontamination operations, ion-exchange resins etc. These wastes usually are put in 200 liters drums and immobilized by pouring cement into the voids.

Because of the widely divergent nature and quality of solid waste, it must be graded right at the place of origin to allow optimum methods of treatment to be applied. This requirements extends not only to the different nature of the waste in terms of burnable and non-burnable, but also to a clear separation into and classification as $\beta - \gamma$ contaminated and α contaminated solid wastes.

According to the experience accumulated so far, 80% of the low-level solid waste produced at IPEN can be incinerated. Practice has shown however that for most of the waste produced the delay and decay treatment is still valid. Thus, in the next future when the incinerator plant be in operation its main use will be for animal carcasses, wastes contaminated with long lived radionuclides ($T_{1/2} > 60d$), combustible liquids, resins or even short lived wastes if the volumetric production justify.

Contaminant Radionuclides

There is no quantitative limit on the contamination level of radioactivity in low-level wastes. While small quantities can be quite highly contaminated, the majority have only relatively low-levels of contamination. In fact, a large percentage of the total wastes handled are only suspect because of their source of generation.

The radionuclides present in the wastes received at the compaction facility of IPEN depends from which laboratory they come. The main radionuclides present in the wastes are: ^{3}H, ^{14}C, ^{24}Na, ^{32}P, ^{35}S, ^{42}K, ^{51}Cr, ^{60}Co, ^{82}Br, ^{89}S, ^{90}Sr, ^{95}Mo, ^{99m}Tc, ^{103}Ru, ^{106}Ru, ^{125}I, ^{125}Te, ^{127}Te, ^{129}Te, ^{131}I, ^{134}Cs, ^{140}Ba, ^{198}Au, ^{226}Ra, U_{nat}, Th_{nat} and minor quantities of other radionuclides.

In figure 2 is shown the relative contribution of the main solid waste contaminants and in figure 3 is shown the relative contribution according to the producers laboratories. The total volume produced during last years and according to the waste origin is shown in table I.

Collection of Solid Wastes

During the first years of operation of the compaction facility none special separation of radioactive waste was used except the subdivision in non-compactible and compactible categories and

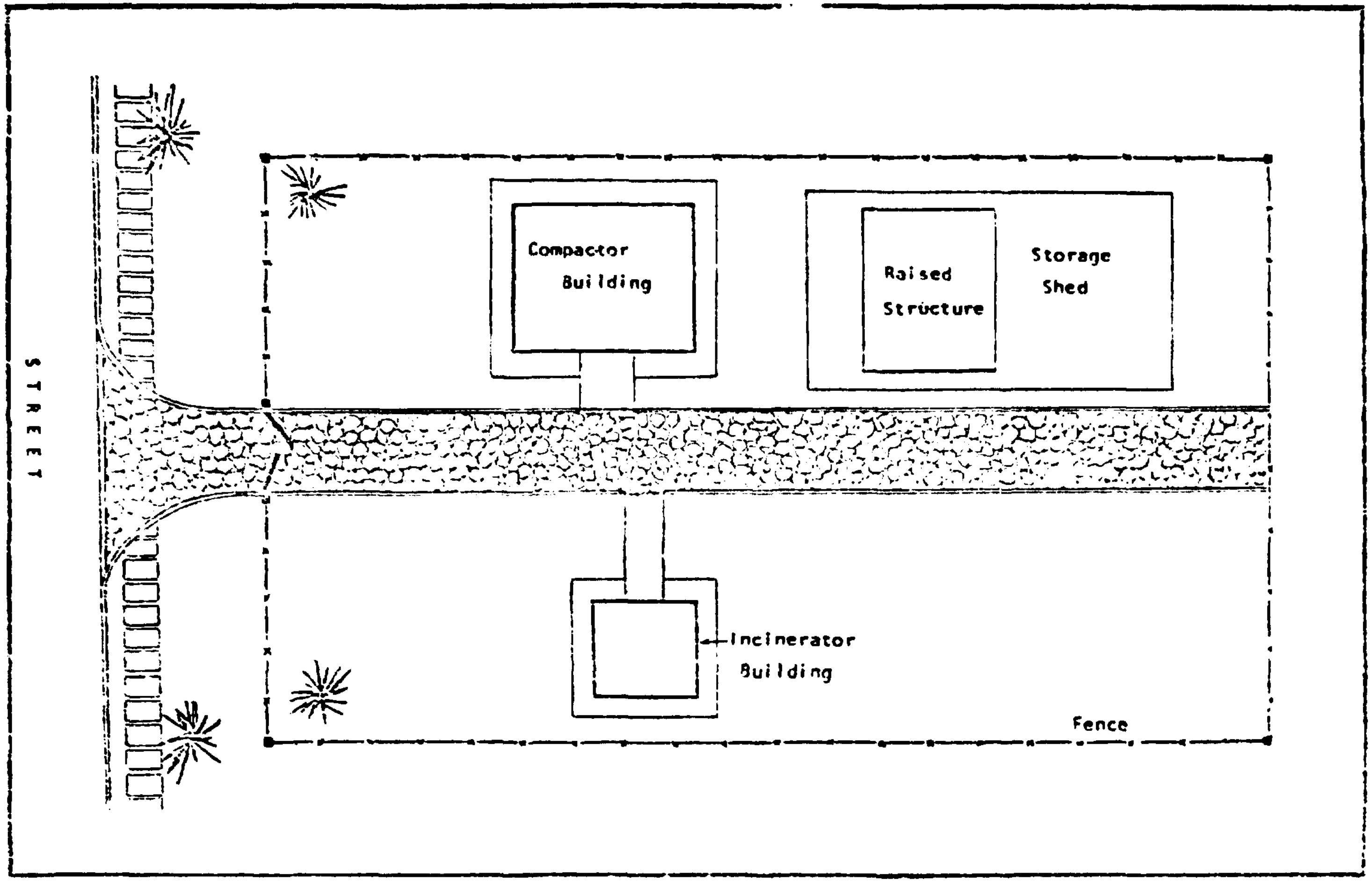

Figure 1 – Lay-out of the treatment facility

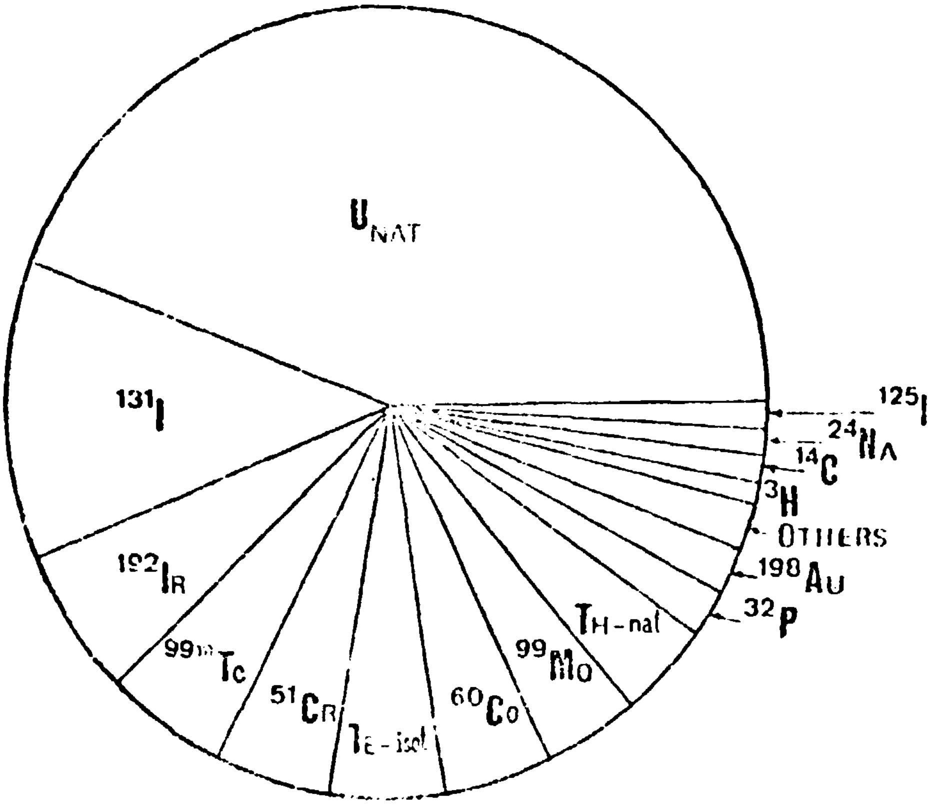

Figure 2 – Solid waste contaminants, volume %

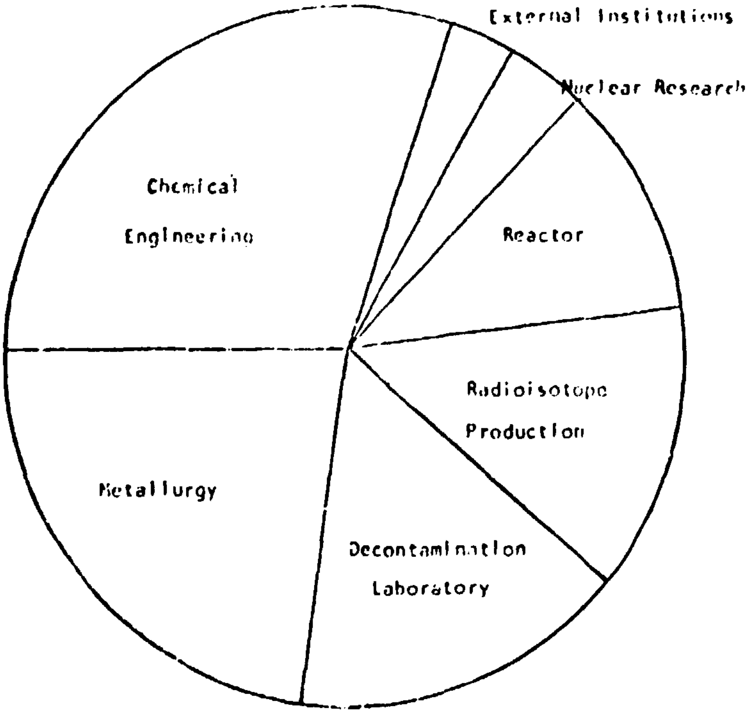

Figure 3 – Waste producers, volume %

Table I

Radioactive solid waste treatment at IPEN

Waste producer	1983 (m^3)	1984 (m^3)	1985 (m^3)
Chemical engineering	5.20	8.90	8.01
Metallurgy	0.80	4.00	5.30
Decontamination lab	0.60	2.60	1.80
Reactor	1.50	0.80	0.70
Nuclear research	0.30	0.50	1.10
Radioisotope production	5.47	4.90	5.65
External institute	–	–	0.4

according to the activity level (greater or lower than 2 mSv/h (200 mR/h) at the external surface of the container). Recently however, in order to comply with the incinerator installation and begin the education of the waste producers, the compactible radioactive wastes are segregated in combustible and non-combustible wastes.

The radioactive wastes are collected in paper bags (40 ℓ) placed inside to polyethylene bags of 0,2 mm thickness. The closed bags are transported weekly to the treatment facility for compaction, temporary storage or eventually in the next future to incineration.

The non-compactible wastes are collected also in polyethylene bags and addressed for special treatment.

All radioactive wastes bags are identified at the points of production. This is done by the health superviser which attaches a properly filled tag to the transfer bag with the following information: a) level of activity; b) radioactive isotopes present; c) estimate of quantity (Bq or Ci); d) general description of waste and e) location of generation operation.

Transportation

For transport of wastes items inside the institute a couple of vehicles are available such as trucks, trailers and fork-lift trucks.

Bulky contaminated wastes are packed in plastic foils and require sometimes special arrangements for transport.

Compactor Operating Procedures

Before the compaction procedure the wastes bags are visually checked by the operator to determine if the waste is properly packaged. Radiation reading are taken as well the bags are weighed prior the compaction operation.

All data relative to the waste and compaction procedure are registered in a logbook.

The compactor exhaust is put on prior the compactor is placed in the operation mode. The operator places plastic bags of wastes inside a drum, move it to the enclosure, closes and locks the door and depresses the control valve lever down. The platen moves down and compresses the waste inside the drum with a maximum compaction force of 10 tons. The operator then depresses the control valve lever to the up position for the return of the platen to the start position, open the door, moves the drum for the filling area and continues this cycle operation until the drum be roughly 90% full. Then, is put a lid covering the drum.

The outside surfaces of the drum are them surveyed and cleaned as necessary prior to its identification, weighing and transference to the interim storage area.

The compactor and compactor room is monitored periodically throughout the day during compaction operations. During all the compaction operation gloves are worn by the operators as well additional clothing requirements.

The facility is operated once each two weeks depending of the waste arrival.

Interim Storage Area

For temporary storage of conditioned or untreated low-level wastes it is provided a storage shed with 200 m^2 area. It has steel structure and is under construction a light external sheathing. The floors have been designed to support a load of at least three piled drums. For those solidified wastes a reserved area for higher loads is provided with 63 m^2.

Up to now, about 144 cubic meters of solid compactible wastes were received, yielding 180 drums of compacted wastes stored in the facility. About others 8 drums have been prepared with non-compactible wastes.

Facility Modifications

During the initial operation of the treatment unit some modifications have been made in order to correct malfunctions of the compactor as well in the storage area.

For instance during normal operations of the compactor and depending of the compression waste resistance the drums were deformed causing difficulties in the closing of them. Once occurred that the driving shaft bended slightly due to a resistent piece of wood placed in the drum. To solve this problem it was installed a knee-cap between the driving shaft and the platen which is there since then.

The floor of the compactor building originally prepared with an epoxi-based paint was substituted by a quartz powder and araldite mixture which has higher mechanical resistance and is more convenient for eventual decontamination procedure.

Previously the drums were stored in raised structures covered by canvas. This procedure was given up because that arrangement was propitious to rust. Besides, the canvas size difficults the handling and sometimes rips out.

Final Discussion

It is recognized that if it was to rebuild all the facility it would be designed in a different way. For instance the lay-out of the compactor building should be replanned in order to arrive at a more favorable flow of the waste stream. Even some minor modifications in the compactor unit is desirable, however despite some objections the treatment facility has been operated successfully.

CONDITIONING LIQUID WASTE FROM I 131 PRODUCTION BY NATURAL TeO_2 NEUTRON IRRADIATION

L. Dalaqua Junior, J. C. Dellamano

ABSTRACT

This paper describes the treatment techniques assayed to decontamination and volume reduction of the waste from I 131 production.

The assayed techniques were: $(NH_4)_2$ TeO_4 precipitation in alcoolic medium and chemical reduction using Sn^{2+} in acid medium and Fe^{2+} in alcaline medium.

There is some considerations about products features and final results.

INTRODUCTION

At IPEN-CNEN/SP the I 131 is produced by neutronic irradiation of natural TeO_2. The products of this irradiation are listed-bellow:

$$\text{Te 121m} \xrightarrow[154d]{IT} \text{Te 121} \xrightarrow[17d]{EC} \text{Sb 121}$$

$$\text{Te 123m} \xrightarrow[117d]{IT} \text{Te 123 } (1.2 \times 10^{23} y)$$

$$\text{Te 125m} \xrightarrow[58d]{IT} \text{Te 126}$$

$$\text{Te 127m} \xrightarrow[109d]{IT} \text{Te 127} \xrightarrow[9.4h]{\beta^-} \text{I 127}$$

$$\text{Te 129m} \xrightarrow[34d]{IT} \text{Te 129} \xrightarrow[69m]{\beta^-} \text{I 129} \xrightarrow[1.7 \times 10^7 y]{\beta^-} \text{Xe 129}$$

$$\text{Te 131m} \xrightarrow[30h]{IT} \text{Te 131} \xrightarrow[25m]{\beta^-} \text{I 131} \xrightarrow[8.05d]{\beta^-} \text{Xe 131m} \xrightarrow[11.8d]{IT} \text{Xe 131}$$

After irradiation the TeO_2 is dissolved with a NaOH solution and oxidized, with MoO_4^{2-} and H_2O_2, to TeO_4^{2-} ions. The solution is treated with concentrated H_2SO_4 and the I 131 is then distilled.

After washing the cell equipment about 1 ℓ of waste is produced with an annual production of 300 ℓ.

This waste basically contains Te isotopes and a residual I 131 activity in 5M H_2SO_4 solution.

The waste is then collected in a polyethylene bottle and stored to allow decay of short-lived isotopes.

However, for reasonable decay, the storage time must be about five years due to the presence of Te 121m (154d half life).

Based on the activity of Te 121m produced, only after this time is possible to deal with this waste by dilution and liberation to environment.

This time is long to store sulfuric solutions securely and the shielded area must be considered.

A more attractive way to solve this problem is to convert the waste to a solid form. By this way a little area is necessary and a more secure storage is achieved.

Bearing this in mind a decontamination and volume reduction treatment study was started aiming to produce a solid waste.

Some initial ideas as the use of ion exchange resins to TeO_4^{2-} ions retention or waste neutralization and cement incorporation were rejected due to the high SO_4^{2-} concentration.

DESCRIPTION

The assayed treatment methods described here were conduced under a non diluted waste. All waste samples were analyzed by γ ray spectrometry before and after the assay to find the decontamination factor.

I. $(NH_4)_2$ TeO_4 precipitation in alcoolic medium:

This treatment was carried out by NH_4OH waste neutralization, using phenolphtalein as indicator, and ethanol addition until total salt crystallization.

This procedure produces a gel precipitate which is adherent to the glass-ware.

The disadvantages of this technique is the low decontamination factor and the use of flammable (ethanol) and volatile compounds.

In a previous study the best results obtained were conduced under a 1: 20 dilution. This procedure gives rise to high volume waste to be treated which is undesirable mainly by the work conditions in a real scale plant.

II. Reduction of TeO_4^{2-} ions:

The reduction technique was carried out using Sn^{2+} as a reducing agent, without neutralization, directly on the waste.

A fine Te precipitate was obtained and there was a high Sn^{2+} consumption probably due to the MoO_4^{2-} ions presence.

After a filtration the filtrate initially transparent was becoming cloudy with slow Te precipitation.

Another reduction essay was made in alcaline medium to prevent I 131 volatilization.

In this case Fe^{2+} ions were used as a reducing agent producing, after reaction, a $Fe(OH)_3$ precipitate.

The $Fe(OH)_3$ acts as a scavenger carring out the ions from the solution.

A great volume of colloidal precipitate was formed with a high water content.

In this case a dewatering process like evacuated filtration or solid-waste compaction is necessary to obtain volume reduction.

In the table I the decontamination factor D. F. and volume reduction V. R. is presented.

Table I

Decontamination factor and volume reduction as $V_{final}/V_{initial}$ ratio

	$(NH_4)_2\ TeO_4$ Precipitation	Reduction	
		Sn^{2+}/H^+	Fe^{2+}/OH^-
D. F.	33	574	219
V. R.	0.4	0.7	2

In the three cases the precipitate obtained were fine, giving a slow filtration and a poor volume reduction due to the high salt content.

Another way to do waste treatment is the evaporation of the liquid followed by crystallization of the product.

Simultaneously with this study an evaporator/crystallizer was designed to be applied to this waste.

The considerations about the evaporator design features are described in: "Design of an evaporator/crystallizer for radioactive liquid waste treatment" presented in this publication.

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