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SUMMARY

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ABSTRACT

A process to pretreat scintillation solutions composed basically of PPO, POPOP, TOLUENE and ANTAROX (emulsifier), utilized by radioimmunoassay laboratories, is described.

The technique employed is distillation which permits a waste reduction to about 40% of the initial volume with the recovery of the solvent (toluene). The recovered toluene can be reused for the same purpose, since it is free of radioactive material as assured by quality control procedures.

DESTILAÇÃO COMO UM PROCESSO DE PRÉ-TRATAMENTO DE SOLUÇÕES CINTILADORAS

RESUMO

É descrito um processo de pré-tratamento, aplicado à soluções cintiladoras compostas basicamente de PPO, POPOP, TOLUENO e ANTAROX (emulsificante), utilizadas por laboratórios de radioimunoensaio, pela técnica de destilação.

Com essa técnica reduz-se o volume a 40% do volume inicial e recupera-se o solvente (tolueno). O tolueno recuperado pode ser reutilizado uma vez que é livre de material radioativo.

1. INTRODUCTION

In recent years the powerful techniques of radioimmunoassay (RIA) has come into increasing use for measuring small concentrations of hormones and other natural substances in tissues extracts, blood plasma and other materials of interest.

In order to conduct those RIA, kits containing ^{3}H, ^{14}C, ^{125}I as tracers and usually scintillation counters are used. The refused cocktails are considered radioactive wastes and according to their activity they can be released to the environment or treated by incineration[1].

In Brazil the ^{3}H discharge limit, indicated by the federal radiological regulations is 185 $Gbq.a^{-1}$[2], and in general the activity of the scintillation solutions arising from the RIA laboratories are lower than this limit.

Therefore it is allowed to release them to the environment from the radiological point of view. On the other hand the solvent content of the scintillation cocktails is rather high, which would give rise chemical pollution of the sewage stream.

Nevertheless, in recent years pretreatment techniques are being considered very important in the waste management scheme. The basic goals of them are to reduce the waste volume and sometimes to "recycle" products, and thereby to lower the costs in subsequent waste management operations[3].

Having this in mind, it was developed a pretreatment technique aiming to reduce the waste volume and to recover the solvent from scintillation solutions. Few papers[4,5,6] exists in the litterature on the same subject in laboratory scale.

2. COMPOSITION AND CHARACTERISTICS OF THE WASTE

The scintillation solutions utilized in RIA laboratories have various compositions[7], but in order to start the pretreatment research, only one type of scintillation solution (the more frequently utilized) was characterized. Its tipical composition is:

		(%V approx.)
a	scintillators	
	PPO (2,5 -- Diphenyloxazole)	4.4
	POPOP (1,4 – Di(2-(5-phenyloxazolyl)) benzene)	0.4
b	solvent - toluene	56.8
c	emulsifier - antarox	28.4
d	biological material – labelled usually with ^{3}H and ^{14}C	traces
e	water of dissolution	10.0

The volume of the scintillation solution described corresponds aproximately to 70% of the total volume received by our laboratories (more than 3,000 $L.a^{-1}$) and has an average activity of 25.9 $KBq\ L^{-1}$.

The waste is received fortnightly in polyethylene bottles with capacity of 20 L each, and behaves usually like a stable emulsion, but sometimes happens to occur phase separation. In this case the solution must be agitated to promote its homogeinity and in this way to allow that any portion of it has the same composition of the one characterized previously.

3. DESCRIPTION OF THE PROCESS

3.1. Distillation

Many separation processes can be used when the goal is to separate a mixture in relatively pure fractions. The principles and theory are essentially the same for all different kinds of separation processes, however in general, distillation is the best choice because of the simplicity of the procedure and apparatus[8].

After WEISSBERG[9] "In broadest terms, distillation is a process of separation based on the difference in composition between a liquid and the vapor formed from it".

There are four types of distillation processes commonly used by industry and laboratories[9,10,11] that are:

3.1.1. Continuous distillation: -- In this process, which is used mainly for large scale operations, the material to be distilled is fed continuously and the product is continuously withdrawn.

3.1.2. Descontinuous distillation: – discontinuous or batch distillation the material to be distilled is fed before the distillation begins and the product is withdrawn during the distillation.

3.1.3. Extractive distillation: – The extractive distillation is made in the presence of a substance which has a boiling point much higher than the operation temperature to be utilized and besides it promotes the increase of the relative volatility of the component to be separated.

3.1.4. Azeotropic distillation: – There are different definitions for this type of distillation, but in the present discussion, it is defined as that which requires the additions of a volatile component to promote the attainment of an azeotropic mixture.

3.2. The choise of distillation as a pretreatment process

Approximately 75% in volume of the waste (with composition characterized previously) received annualy, that is 1,500 $L.a^{-1}$, corresponds to the solvent. This solvent, toluene, has a boilling point equal to 110.6°C and it forms with water an azeotrope (20.2% water and 79.8% toluene) which boilling point is 85°C. These temperatures are lower than the boilling point of the other components. As for example PPO has a boilling point equal to 360°C.

Based on these data distillation was selected as the process for pretreatment of liquid acintillation waste. The basic goal of this pretreatment is to reduce the waste volume to be treated either by incineration or by discharge in the environment[2] besides to recovery the toluene.

The type of distillation selected was the batch distillation process based on the volume received annualy

Three differents distillation procedures were assayed in laboratory: simple distillation, reflow distillation and bi-distillation all followed by phase separation and withdrawal of toluene. Samples of recovered toluene were utilized to prepare scintillation cocktails to allow a comparison with scintillation cocktails prepared with A.R. (Analytic Reagent) toluene, in order to obtain data about decontamination and reproductiveness of the process. The results are presented in Table I.

Table I

Data obtained with scintillation solutions prepared with A.R. toluene and recovered toluene

Toluene utilized in scintillation solution	1st batch count (cpm)	2nd batch count (cpm)	3rd batch count (cpm)	average count rate (cpm)
A. R.	32,9 ± 4,2	33,7 ± 3,8	32,7 ± 3,9	33,1 ± 2,3
recovered by simple dist.	32,5 ± 2,2	35,0 ± 1,6	34,2 ± 2,9	35,4 ± 1,2
recovered by reflow dist.	32,5 ± 5,2	32,7 ± 1,4	33,7 ± 1,9	33,0 ± 1,2
recovered by bi-dist.(*)	36,3 ± 7,8	36,4 ± 3,4	34,2 ± 2,2	35,2 ± 1,9

(*) Obs.: The operation time in this procedure is about the double of the others procedures.

Based on statistical analysis of the data[12], the three procedures can be considered reproducible within a 95% confidence interval. However when the recovered toluene is compared with A.R. toluene, only the reflow distillaltion procedure presents acceptable results at the same confidence interval Therefore, the toluene revovered by simple and bi-distillation can be considered as undecontaminated products.

These results can be interpreted as a consequence of the likely entrainment of contaminated mate..al droplets during the distillation.

3.3. The experimental unit

Aiming to pretreat the scintillation solutions utilized for the majority of the radioimmunoassay laboratories it was done a desing for mounting and operation of an experimental unit.

The choice of glass material for the unit, as we l the capacity of it as being 20 L d^{-1} (two batches of 10 L each) was based on the waste volume received annualy. A lay-out of the unit and a flow diagram are in the Figures 1 and 2 respectively, and the apparatus especifications are in Table II.

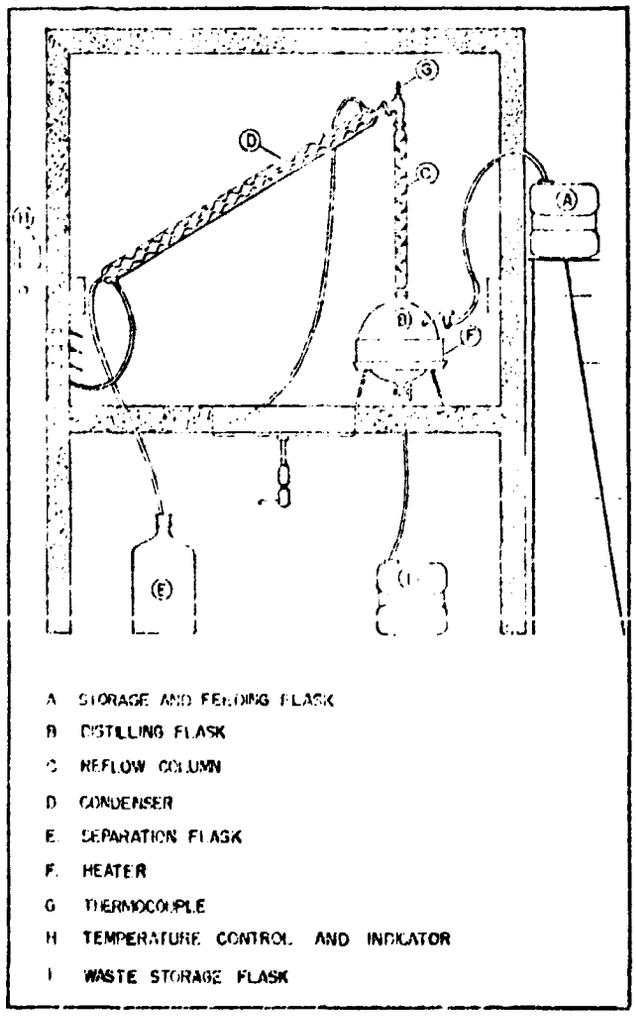

Figure 1 – Lay-out of the unit for pretreatment of scintillation solutions

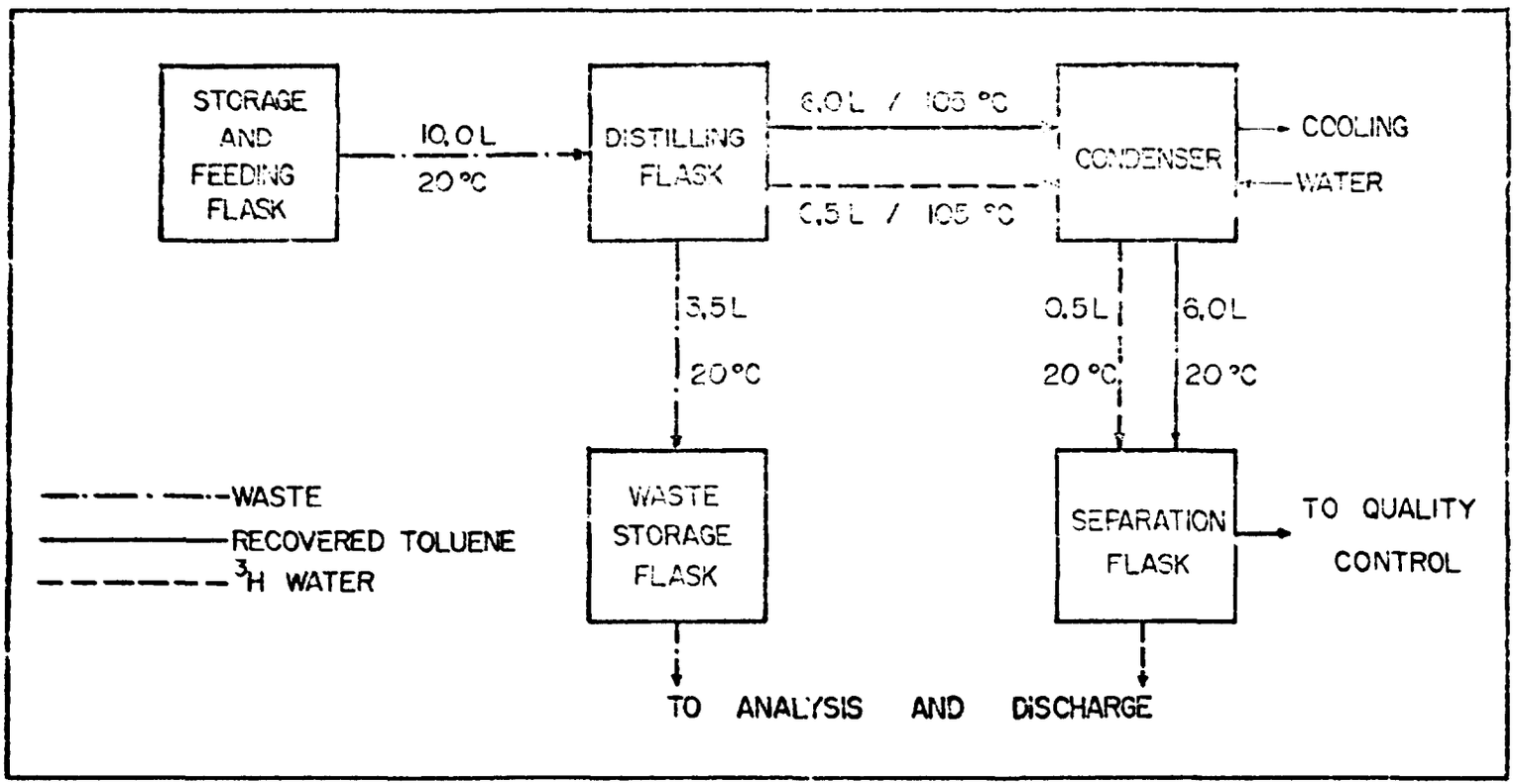

Figure 2 – Flow diagram of the process for pretreatment of scintillation solutions

In short, the operation procedure is the following: The waste is fed from storage flask to the distilling flask by gravity, then the heating system is turned on. When the temperature achieve 85°C the azeotrope mixture (water and toluene) is distilled. When the water contents ends the temperature rises to 110°C and the rest of toluene is distilled. When the temperature rises again the heating system is turned off.

The azeotrope mixture and toluene are received in the separation flask after condensation. After the phase separation the toluene is withdrawal to analysis and reutilization.

The data obtained in the operations that were realized whit this process until now are the following:

a. waste volume reduction 40%
b. process yield 80%
c. cooling water consumption ..... 150 L h^{-1}
d. power ..... 3.5 kW
e. operation time per batch of 10 L ..... 50 min.
f. phase separation time ..... 24 h

3.4. The quality assurance

After each toluene recuperation procedure, it was performed a quality control in order to assure the absence of radioactive material in it, as well that its chemical grade is acceptable for scintillation cocktail preparation with reproducible count efficiency.

Chemical analysis was also made in order to evaluate the chemical purity degree of toluene. The average purity degree obtained was 99.5% and the impurities found were benzene and aliphatic hydrocarbons.

Table II

Some specifications of the apparatus of the unit for pretreatment of scintillation solutions.

Code in lay-out	name	specification
A	Storage and feeding flask	Polyethylene bottles, 20 litter capacity.
B	Distilling flask	Round bottom flask with 12 litter capacity and 24/40 ground-joint. Adaptation of stopcocks for feeding and withdrawal at 2/3 height and on the bottom respectively.
C	Reflov/ column	Vigreaux column with 450mm lenght and 24/40 ground joints.
D	Condenser	Allihn condenser with 1,000mm lenght and 24/40 ground joint.
E	Separation flask	9 litter bottle with one-hole rubber stopper.
F	Heater	Heater wrap-around with 300mm of diameter. This heater operates on 220V and offers heat output of 3,500W.
G	Thermocouple	Iron-constantan sensor 20 gauge with 40mm lenght and AISI 316 stainless steel protection.
H	Temperature control and indicator	Temperature control and indicator system special for iron-constantan sensor with temperature renge from 20°C to 250°C.
I	Waste storage flask	The same bottles that were utilized as feeding flask (now empty).

4. CONCLUSION

The distillation technique used is shown as an alternative pretreatment for liquid scintillation wastes, because it reduces the waste volume to be treated, through the separation of a reagent that can be reutilized, and in this way decreases the chemical pollution of the environment.

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