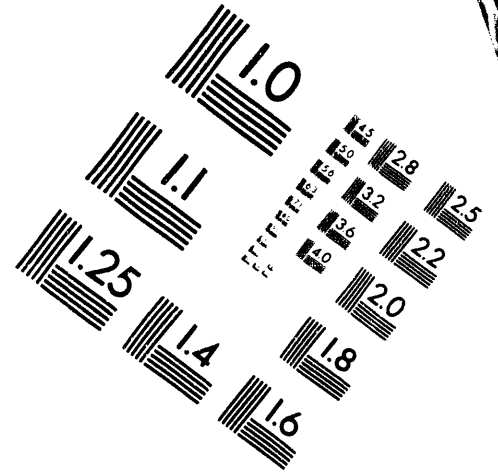
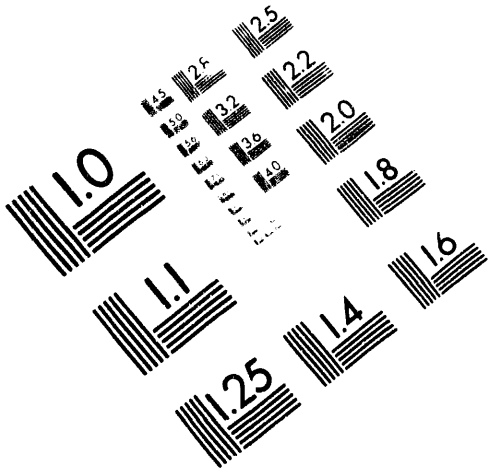




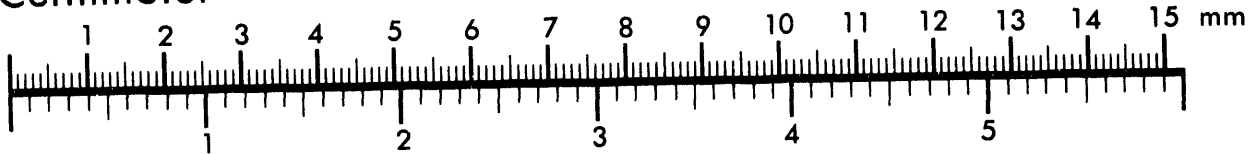
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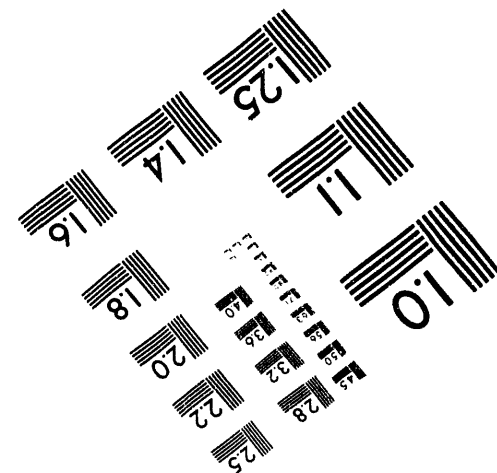
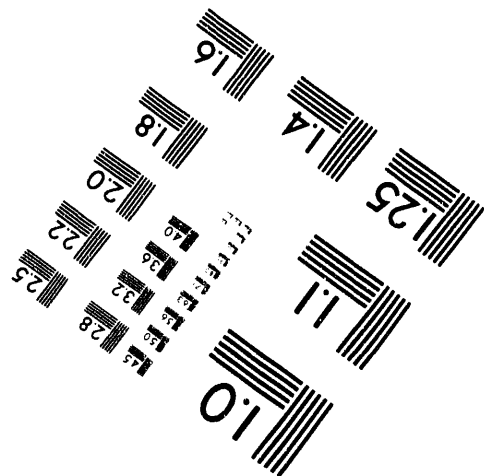
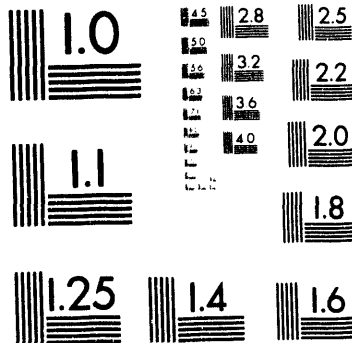
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RECYCLING OF ACETONE BY DISTILLATION

by


Analytical T.Q. Team Members

Deborah L. Brennan
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Mark Harper

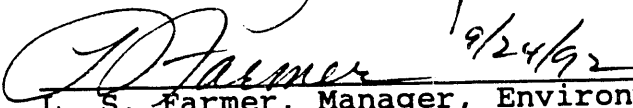
September 1992

**FERNALD ENVIRONMENTAL MANAGEMENT PROJECT
WESTINGHOUSE ENVIRONMENTAL MANAGEMENT COMPANY OF OHIO
P.O. BOX 398704
CINCINNATI, OHIO 45239-8704**

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EXECUTIVE SUMMARY

The mission of this company and the world today is the preservation of our environment. This can be accomplished through re-use, recycling and reduction in waste generation. A two-phase distillation unit was designed and tested for the purpose of recycling spent acetone routinely used in performing analytical laboratory determinations.

We obtained data through experimental work conclusively proving that background levels of radiation can be achieved from highly radioactively contaminated acetone through a controlled distillation process.

The addition of water to a flammable solvent enhances safety by diluting any oxidizing agent concentration below its lower explosion limit. This addition also enhances safety by causing the solvent to distill off at a lower temperature than water, so a boiler high temperature malfunction will not ignite the solvent.

The contaminants, nitric acid and technetium-99 (⁹⁹Tc), have a greater affinity for the water, therefore they are not distilled over.

By using a carefully monitored distillation process, optimum yields of greater than 90% can be achieved providing a recycled product and reducing disposal and/or storage by the same amount.

A two-phase distillation process was found to be effective and the most efficient method tested.

1. INTRODUCTION

The Resource Conservation Recovery Act (RCRA) identifies spent acetone solvent as a listed hazardous waste. At Fernald we have spent acetone that has also been contaminated with radionuclides and therefore is identified as a mixed hazardous waste. At the time of this publication there is no available approved method of recycling or disposal of radioactively contaminated spent acetone solvent. The Consent Decree with the Ohio EPA and the Consent Agreement with the United States EPA was agreed upon for the long-term compliant storage of hazardous waste materials.

The purpose of this project was to demonstrate the feasibility for safely decontaminating spent acetone to background levels of radioactivity for reuse. It was postulated that through heat distillation, radionuclides could be isolated from the spent acetone.

2. THEORY

All radioactive contaminants of interest found in the spent acetone are dissolved solids. The theory of distillation dictates that solids will remain as a distillation residue. Through condensation the distillate can be captured in a pure, decontaminated form that is acceptable for re-use.

One radionuclide, ^{99}Tc , presented a unique concern. In the presence of nitric acid and heat, this component has the potential to volatilize along with acetone, causing it to reappear in the pure distillate. Spiked experiments needed to be conducted to test technetium volatility. The presence of nitric acid, a strong oxidizer, in the spent acetone provided a potential explosion hazard. Water must be added to provide safety by diluting any oxidizing agent concentration below its explosion point. These concerns will be addressed in the Experimental section.

3. CONCLUSIONS

A. The experimental work proves that background levels of radiation can be achieved from highly radioactively contaminated acetone because the contaminants (HNO_3 ; ^{99}Tc) have a greater affinity for the H_2O therefore they are not distilled over.

B. The addition of water to a flammable solvent provides safety by diluting any oxidizing agent concentration below its explosion point; it also reduces the potential for igniting the solvent due to a boiler high temperature malfunction.

The contaminants (HNO_3 ; ^{99}Tc) have a greater affinity for the H_2O therefore they are not distilled over.

C. The optimum yields of greater than 90% can be achieved providing a recycled product and reducing disposal and/or storage problems.

problems.

Recycling of spent solvents reduces waste and decreases any negative environmental impacts.

The initial success with acetone on the experimental scale indicates that additional investigation of solvents with similar physical and chemical properties is warranted.

4. EXPERIMENTAL WORK

A single-phase distillation unit (see Drawing A) was assembled with standard laboratory glassware and a column (3.7cm x 41.25cm) packed with 3 mm glass beads. The beaded column was implemented to expedite separation of acetone and water. The still (see Drawing A) consisted of packed column connected via a Claisen head to a condenser and connector into a three-necked 1000 mL round bottom collection flask. The collection vessel was vented via a water scrubber to maintain an open system. Heat was provided by a standard laboratory heating mantle, regulated by a 110-volt AC (VAC) variac monitored by an in-line voltmeter. Thermometers inserted into the distilling flask and the Claisen head recorded operating temperature parameters. After assembly, a leak test was successfully conducted on the unit prior to conducting any experimental work.

To establish preliminary optimum parameters, 500 mL of deionized water was charged to the distilling flask. The system was started at 35 VAC and raised in 10 VAC increments at 30 minute intervals until a VAC of 75 was attained. It was then raised an additional 5 VAC to 80 VAC with a visible vapor level measurement in the beaded column at 6 inches. This level was decidedly too high, and the voltage was reduced 5 VAC incrementally every 30 minutes until the visible vapor level in the column was between 1 to 1-1/2 inches. This was achieved at 60 VAC, maintained for 1 hour, and then shut down. (See Table 1) The next day, the system was turned on at 60 VAC to verify that the system could maintain levels duplicating that of a cold start-up. The unit stabilized after two hours and maintained a visible level of 2 inches for 4-1/2 hours after which it was turned off.

4.1 RUN #1 [5/29/91]

A mixture of 100 mL of acetone and 500 mL of deionized water was introduced into the still at 0800 hrs and heated to boiling at 60 VAC. At 0840 hrs the acetone vapor level had reached the top of the column and the temperature had reached 56°C. Acetone distillation had begun. By 1000 hrs, the acetone appeared to stop distilling and the temperature in the Claisen head was cycling between 46°C and 54°C. The coldest pot temperature we could observe was 95°C. At this time the voltage was raised to 65 VAC. After 40 minutes, the distillate level in the reception flask remained constant with no acetone distillation occurring. The cyclic nature

of the temperature change in the Claisen head indicated that the packed column had flooded. One hundred mL of acetone was added in an attempt to overcome the flooding in the column. After twenty minutes acetone was distilling again as evidenced by an increased level of acetone in the reception flask. At 1125 hrs, the pot temperature reached 104°C. At 1215 hrs the vapor temperature was cycling between 35°C and 45°C, and distillation had stopped, indicating that the packed column had once again flooded.

4.2 RUN #2 [5-31-91]

Prior to the second run, the entire still was dismantled and cleaned. A charge consisting of 500 mL of water, 100 mL of acetone and 5 mL ⁹⁹Tc solution at a concentration of 110.74 disintegrations per minute per milliliter (d/m/mL) was introduced at 0930 hrs. The variac was set to 65 VAC and the temperature throughout the unit was constant at 27°C. A vapor temperature of 55°C was achieved at 1010 hrs and remained constant for 30 minutes, when it began cycling between 45th and 55th. It remained at the cycling levels stated for an additional 50 minutes. The pot temperature reached 100th, indicating that once again the packed column had flooded.

Recovery rates on Runs 1 and 2 were 50% (See Table 2). Samples of distillate submitted from Run 2 for alpha/beta analysis were determined inconclusive due to the relatively low activity of the spike. (See Table 3)

4.3 Run #3

The still was charged with spent acetone collected from the Isotopic and Organic Group (IOG) Satellite Accumulation Area (SAA). Distillation was completed under the same parameters as used in Run #2. Four distillate samples were prepared for alpha/beta and uranium analyses. Alpha/beta analyses were performed on a Tennelec LB5100 low background alpha/beta proportional counter by IOG. Uranium analyses were done by the Bioassay Group using the Fusion Method. Results of these analyses indicated a return to background levels of uranium content and radioactivity. (See Table 4 & 5)

4.4 Run #4 & 5

In an attempt to improve yield, clamps were added and the beaded column was insulated. All other parameters were the same. The yield increased from 50% to 70%. (See Table 2)

4.5 Run #6

The still was charged with 500 mL deionized water, 200 mL of clean acetone and spiked with 10 ml of ^{99}Tc at 6427 d/m/ml. Activity levels in the spiked sample were high enough to conclude that the ^{99}Tc did not volatilize and reappear in the distillate. (See Table 6)

4.6 Run #7

The unit was operated using previously established parameters with the exception of the water/acetone ratio. This was done in an attempt to improve yield. The volume of water was reduced from 500

mL to 200 mL and the acetone was increased from 200 mL to 500 mL. Although yield remained the same, efficiency was improved. For the first time since beginning this experiment, more reclaimed distillate was produced than still bottoms as waste.

4.7 Run #8

At this time the still was expanded to a two phase distillation unit (See Drawing B). Phase I was used to separate radionuclides from the water and spent solvent. Phase 2 separated the acetone from the water. The phase 1 radioactively contaminated still bottoms can be discarded as radioactive mixed waste. The phase 2 still bottoms can be reused as a diluent in phase 1 of subsequent distillations, thus optimizing yields to greater than 90%.

4.8 Run #9

Parameters identical to Run #8 yielded results identical to Run #8. At this time, all distilled acetone was sent to the WEMCO Organic Lab for a GC/MS Purity Assay. No impurities were found in any of the samples. (see table 7)

APPENDIX A

ACRONYMS

WEMCO	-	Westinghouse Environmental Management Company of Ohio
FEMP	-	Fernald Environmental Management Project
RCRA	-	Resource Conservation & Recovery Act
EPM	-	Environmental & Process Monitoring Laboratory
TQ	-	Total Quality
SAA	-	Satellite Accumulation Area
IOG	-	Isotopic & Organic Group
VAC	-	Volt Alternating Current
DPM	-	Disintegrations Per Minute
EPA	-	Environmental Protection Agency
DI H ₂ O	-	Deionized Water

DEFINITIONS

MIXED WASTE - Radioactive RCRA waste as identified by WEMCO.

RCRA WASTE - Waste classified under the Resource Conservation Recovery Act.

RADIOACTIVE WASTE - Waste containing radioactive nuclides.

SPECIFIC GRAVITY - The ratio of the density of a substance to the density of some substance (as pure H₂O on H₂) taken as a standard when both densities are obtained by weighing in air.

ALPHA PARTICLES - A positively charge nuclear particle identical with the nucleus of the helium atom that consists of 2 protons and 2 neutrons and is ejected at high speed in certain radioactive

transformations.

BETA PARTICLES - An electron or positron ejected from the nucleus of an atom during radioactive decay.

GAMMA RAYS - A photon or radiation quantum emitted spontaneously by a radioactive substance.

SPENT ACETONE - Used acetone containing radioactive and other contaminants which is no longer useful for its intended purpose in the analytical lab's glass cleaning procedure.

STILL BOTTOMS - Residual contaminants left by distillation of a spent solvent.

APPENDIX B

EQUIPMENT

DISTILLATION UNIT (STILL) -

Three necked flask - 1000ml round bottom - (3)

Distillation column - 24" - (2)

Fractionating column 3.7cm x 41.25cm - (1)

Glass beads - 3 mm - packing

Claisen heads thermometer fitting (2)

Thermometers - 20 - 150°C - (3)

Heating mantle - (2)

Variac - (1)

Voltmeter - (1)

Scrubber - (1)

Miscellaneous glassware

TENNELEC LP 1000 BETA COUNTER - Method # OSH-P-34-006

APC ALPHA COUNTER - Method # OSH-P-34-008

FLUOROPHOTOMETER - Fluorometric Uranium Method # ESHP 34004

TENNELEC LB 5100 A/B PROPORTIONAL COUNTER - Method # FMPC-5100

HP 5970B - GC/MS - Method # FMPC-9002

METTLER/PAAR DMA 46 DENSITY METER - Method # FMPC-3043

ESTABLISHMENT OF OPTIMUM SETTING

VARIAC SETTING	VAPOR LEVEL	VAPOR TEMP.	POT TEMP.
35 VAC	---	22 ^{oh}	22 ^{oh}
45 VAC	---	22 ^{oh}	25 ^{oh}
55 VAC	---	25 ^{oh}	obscured by thermometer holder
65 VAC	---	25 ^{oh}	obscured by thermometer holder
75 VAC	---	25 ^{oh}	103 ^{oh}
80 VAC	some vapor on beads at bottom of column up to 6 in.	25 ^{oh}	103 ^{oh}
75 VAC	6 in.	25 ^{oh}	103 ^{oh}
65 VAC	8 in.	25 ^{oh}	103 ^{oh}
60 VAC	5 in.	26 ^{oh}	103 ^{oh}
55 VAC	2.5 in.	26 ^{oh}	103 ^{oh}
60 VAC	---	26 ^{oh}	103 ^{oh}
60 VAC	1.5 in.	26 ^{oh}	103 ^{oh}
60 VAC	1.5 in.	26 ^{oh}	103 ^{oh}
60 VAC	1.5 in.	26 ^{oh}	103 ^{oh}

TABLE 1

YIELD EFFICIENCY

RUN #	AMT. CHARGED	H ² O ADDED	AMT. RECOVERY	STILL BOTTOMS	% RECOVERY
1	200	500	110	590	55%
2	100	500	50	550	50%
3	400	500	230	670	58%
4*	200	500	100	600	50%
5*	200	500	150	600	75%
6*	200	500	190	510	95%
7*	500	100	370	330	74%
8 *,**	500	100	470	130	94%
9 *,**	500	100	470	130	94%

TABLE 2

* Joint clamps & insulation added
 ** Two-phase distillation used

⁹⁹Tc α & β COUNTS

	α cpm	β cpm
pure distilled	0.14	0
spiked charge	0.11	0.48
spike - still bottom P distillate	0.15	0.047
P distillate	0.12	0.15
purchased	0.12	0.21

TABLE 3

Samples were spiked with 5 mL ⁹⁹Tc @ 110.74 d/m/ml.
Spike was not large enough, therefore results considered inconclusive.

URANIUM ON SPENT ACETONE FOR RUN #3

SOURCE OF ANALYTICAL DATA	mg/L
as purchased	0.005
distilled	0.023
charge	25
still bottoms	*15

TABLE 4

* Difference between results attained on charge & still bottoms is a direct result of the water added.

ALPHA / BETA COUNTS ON SPENT ACETONE FOR RUN # 3

ACETONE DATA	α COUNTS	β COUNTS
not distilled, as purchased	14	116
distilled from contaminated acetone	12	129
not distilled, from Isotopic and Organic Group Satalite Accumilation Area	2641	3588
acetone & water mixed	1437	2151

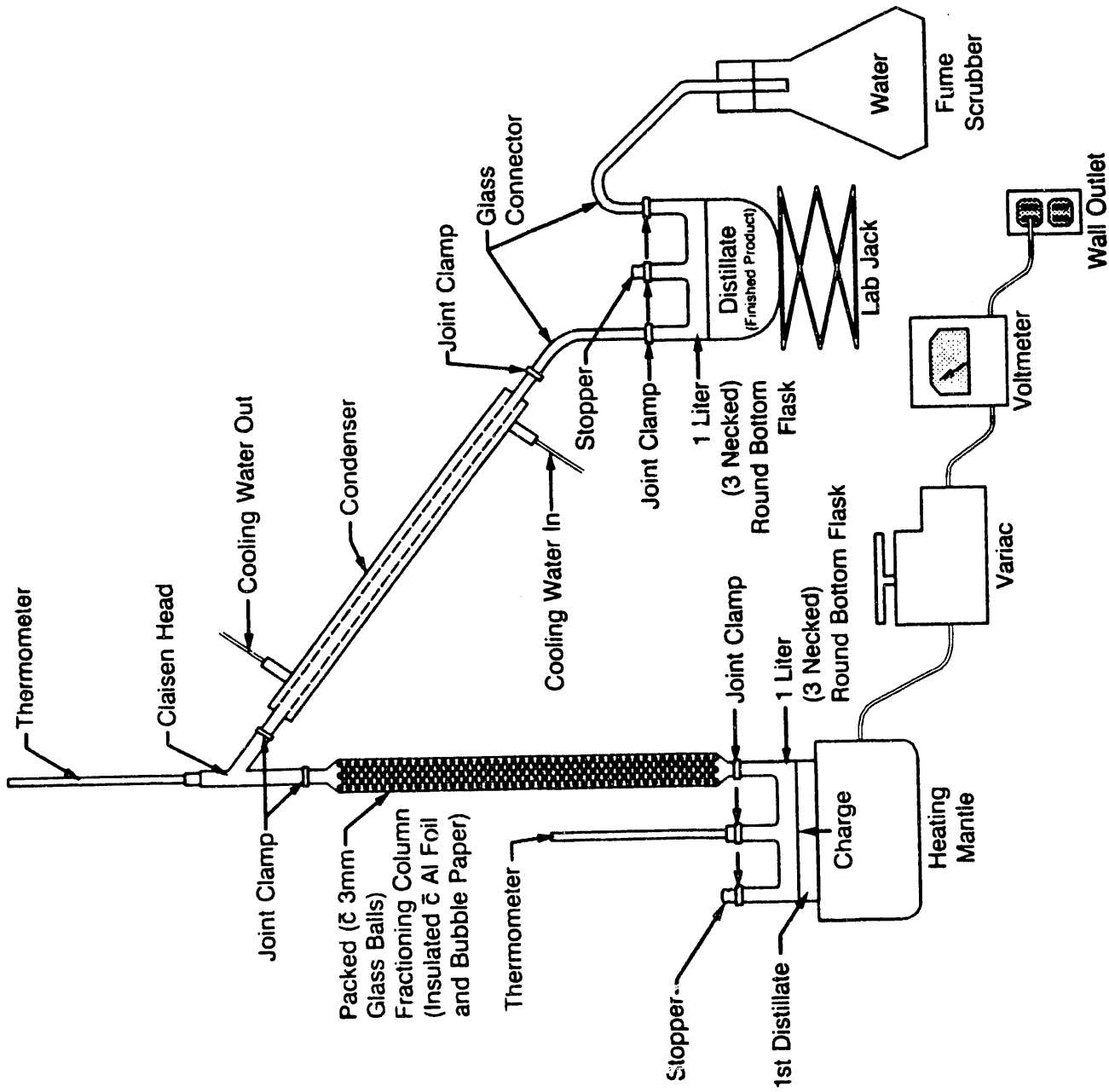
TABLE 5

ALPHA/BETA COUNTS FOR ^{99}Tc IN RUN #8

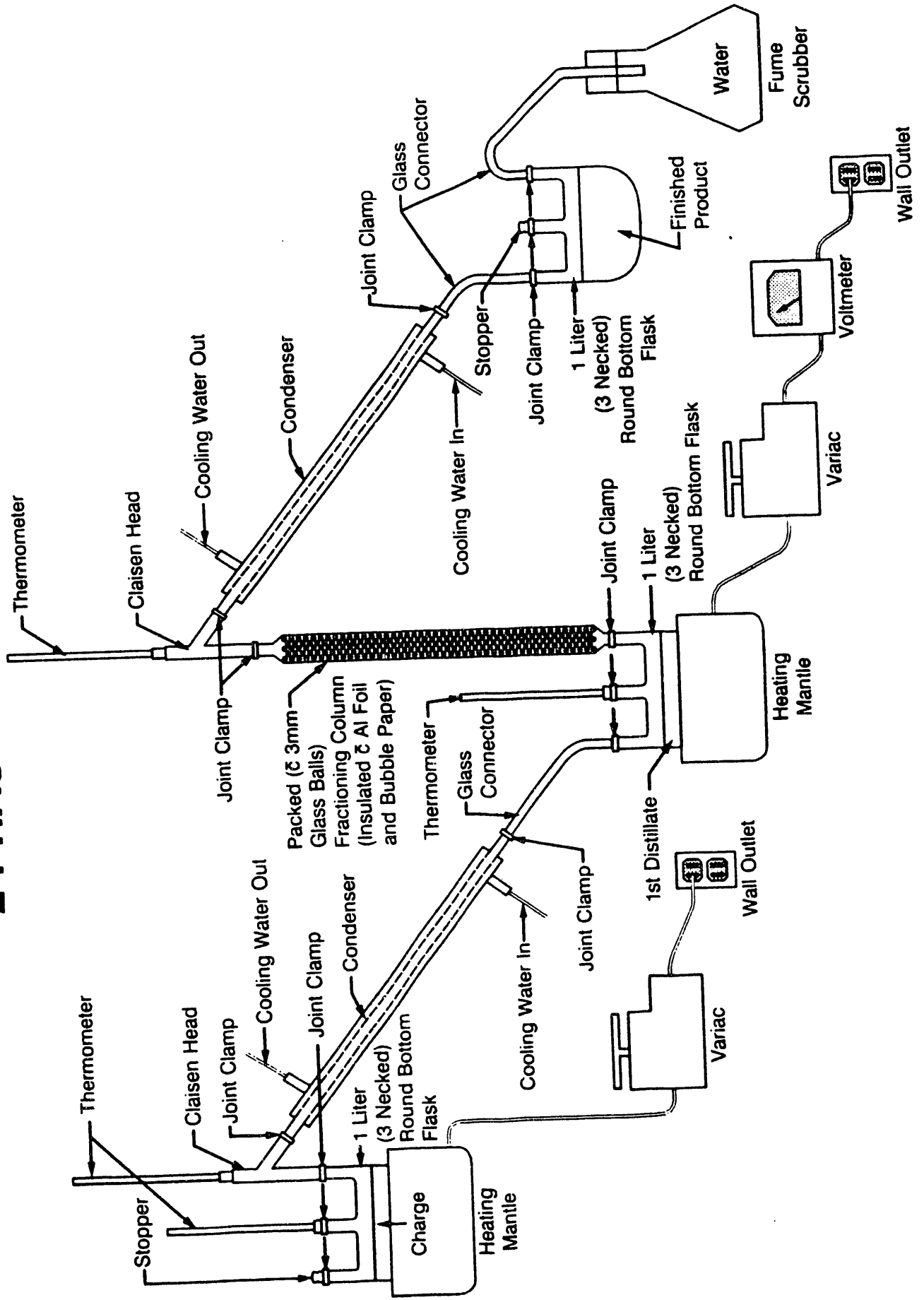
SOURCE OF ACETONE DATA	α COUNTS	β COUNTS
as purchased	31	141
distilled from dirty acetone	12	87
D,T water	17	85
still bottoms #1 spike with 10 mL ^{99}Tc @ 6427 d/m/ml	397	38064
still bottoms #2 distilled from still #1	10	104

TABLE 6

SINGLE PHASE DISTILLATION



2 PHASE DISTILLATION



DATE

FILMED

9/20/93

END

