

SOME MEASUREMENTS OF THE TRITIUM CONTENT  
IN THE NATURAL WATERS OF SOUTHERN OAHU, HAWAII

by

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Technical Report No. 34

November 1969

Project Completion Report

of

WATER TRACING AND DATING IN HAWAII HYDROLOGIC CYCLE - PHASE II

OWRR Project No. A-016-HI, Grant Agreement No. 14-01-0001-1630

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Project Period: February 1967 to September 1969

The programs and activities described herein were supported in part by funds provided by the United States Department of the Interior as authorized under the Water Resources Act of 1964, Public Law 88-379.



## ABSTRACT

*An analysis was made of the tritium content of samples from various sources of surface and subsurface waters of the island of Oahu. Twenty rainwater samples collected from various rain gages located on the Koolau Range over a period of four months showed an activity between 16.3 and 28.2 tritium units. Samples from four streams in the Pearl Harbor area ranged from 14.5 to 21.3 tritium units, while samples from two springs and five wells in this same area all showed an activity below 5 tritium units.*

*The instrumentation consisted of an electrolysis enrichment apparatus, a vacuum distillation unit and a liquid scintillation counter. A detailed description is given of the various parts of the electrolysis apparatus and the vacuum distillation unit. Both the enrichment and counting procedures are reviewed as well as the calibration of these systems.*



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## INTRODUCTION

The analysis of natural water for tritium is based on the detection of the beta radiation which accompanies the radioactive decay of  $^3\text{H}$  (tritium) to  $^3\text{He}$  (helium). Two important factors play a role:

- 1) The beta radiation is very weak (maximum = 0.018 MeV), and
- 2) The tritium concentration in natural water is usually very small.

It is obvious that these two factors place special demands on a radiation detection system. Although very sensitive systems have been developed, tritium activity is usually so low that direct measurement by counting is only rarely possible.

To solve this problem, water samples are generally subjected to some type of enrichment procedure. At present several methods are used for tritium enrichment, such as, distillation, gas chromatography, thermal diffusion, and electrolysis. Of these, distillation is the simplest method. However, the enrichment factor obtainable is small and difficult to reproduce. Thermal diffusion is a fast and efficient method but complicated. Gas chromatography is slow and complicated as well. Electrolysis is simple and quite reproducible and hence is the most commonly used method.

The counting of tritium can be performed using either a gas proportional counter or a liquid scintillation counter. For gas proportional counting the water samples must be converted to hydrogen gas. Proportional counting is very sensitive and has low background counting rates but is a rather complicated and expensive process. For liquid scintillation counting the water samples must only be mixed with a scintillation solution.

Furthermore, liquid scintillation counting is simple and very useful for routine analysis since most counters are equipped with automatic sample changers (Jacobs, 1968). Water samples can also be converted to benzene and subsequently counted in a liquid scin-

tillation counter. Although the latter method is quite sensitive a pre-enrichment is still necessary for samples of low activity.

The tritium analysis of natural water thus consists of two main operations: enrichment and counting. At the Water Resources Research Center's Tritium Laboratory the enrichment is carried out by electrolysis and the counting of the enriched samples is performed by the liquid scintillation method.

## INSTRUMENTATION

The present equipment is, in several aspects, still the same as that described in the previous report by Lau and Hailu (1968). However, important changes have been made in the electrolytic enrichment apparatus and associated procedures. The electronic devices for the automatic addition of sample water to the electrolysis cells were so troublesome that they have been discarded. The control that automatically turns off the current when the water level in the cells reached the 5-ml mark was also discarded. Adapters were constructed to make it possible to mount the sample-addition funnels on top of the cells. These cell-funnel units can easily be lifted out of the water bath, hence, the water volume in the cells can be visually inspected whenever so desired. The temperature control of the water bath has been improved through the installation of a new refrigerator as well as through some modifications in the heating unit circuitry.

### Electrolysis Apparatus

*GLASSWARE.* The electrolytic enrichment of water samples is performed in glass cells purchased from the "Wicklunds Glasinstrument" factory in Sweden. These periodic-addition electrolysis cells were designed specifically for tritium enrichment work by Ostlund and Werner (1962). The lower half of the cells are flattened and follow the shape of the electrodes (Figure 1A). Hence, even a small volume of water can



be in contact with a relatively large electrode surface, a condition especially important during the final stage of the electrolysis process. Several horizontal lines have been engraved into the glass surface marking the 10- 25- 50- 75- and 100-ml volume levels. The heads of the cells are provided with side arms through which the reaction gases can escape (Figure 1A). Pieces of nickel-tungsten wire are sealed into these sidearms to provide a gas-tight inlet for electrical wires.

Funnels are mounted on top of the cells by means of adapters. Each adapter consists of an upper and a lower part which are held together by a clamp and an o-ring seal (Figure 1A). The upper part, which is attached to the funnel, has a sidearm for argon flushing. The lower part rests in the cell head and has a long stem to facilitate sample water addition into the cell. Argon gas can be introduced into the funnel through the gas inlet which is fitted in the funnel stopper.

*ELECTRONICS.* Each electrolysis cell contains a nickel anode and a mild-steel cathode which are held together by appropriate disks and spacers made of teflon (Figure 1B). New electrodes are polished and, in addition, the cathodes are treated in a phosphoric-acid bath. This surface treatment of the electrodes improves the effectiveness and reproducibility of the enrichment process. A preliminary electrolysis, using 200 ml of tap water per cell, is carried out to further "break in" the electrode surfaces. After these treatments the electrodes are ready for regular use.

Electrolysis continuously produces a mixture of oxygen and hydrogen gas. Any heated or sparking connector inside a cell will cause these gases to react with each other resulting in a violent explosion. All connectors are therefore regularly inspected to make sure that they are not loose or corroded. In addition, new cellheads with plastic side arms are being tested. These side arms eliminate the need for the glass-sealed wires which are too fragile and become warm when subjected to high currents.

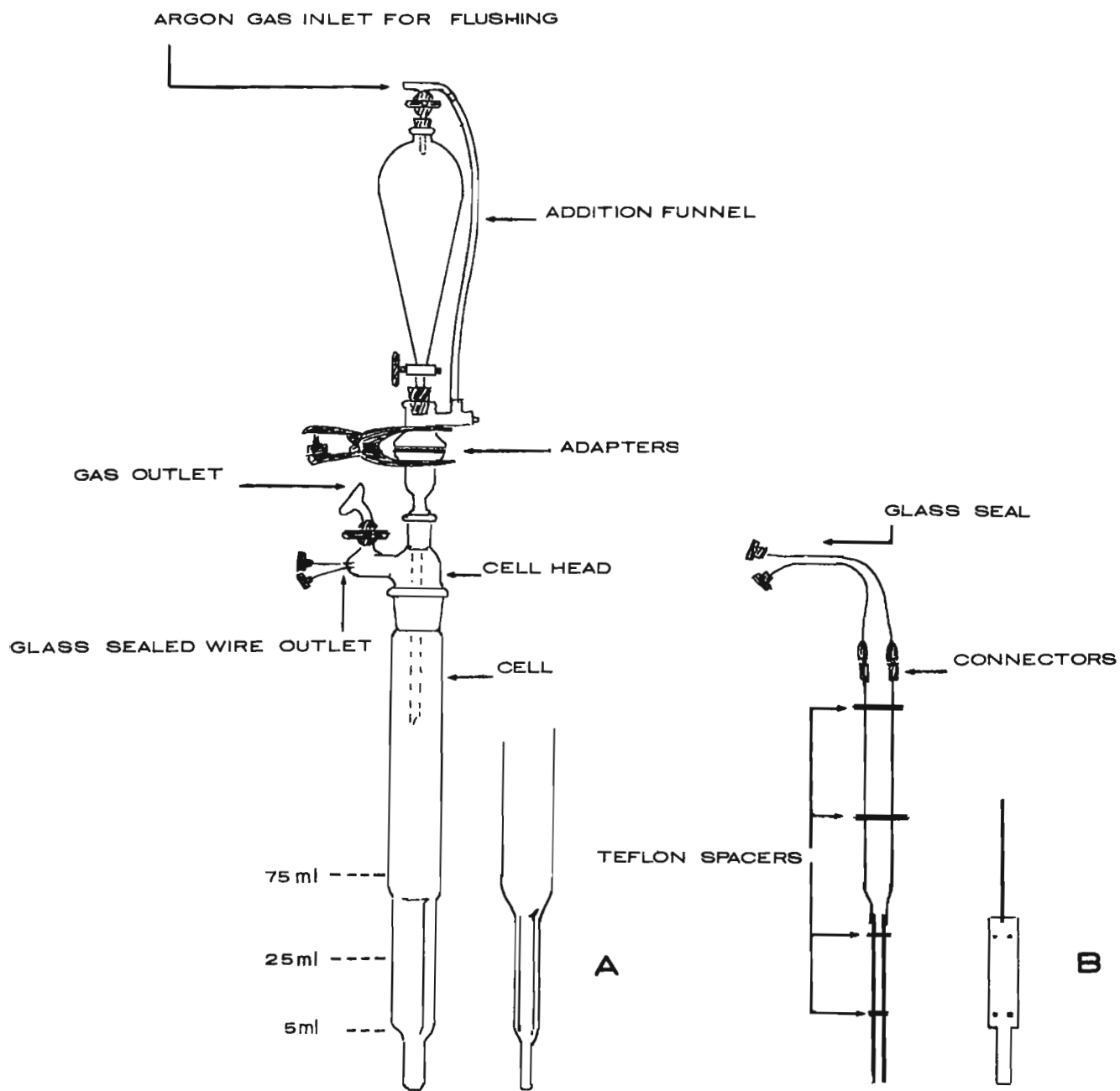


FIGURE 1 a) ASSEMBLY OF ELECTROLYSIS CELL AND ADDITION FUNNEL  
b) ELECTRODES AND WIRING

FIGURE 1. A) ASSEMBLY OF ELECTROLYSIS CELL AND ADDITION FUNNEL.  
B) ELECTRODES AND WIRING.

Two power supplies provide the required electricity. Each unit delivers a constant "direct current," which is monitored by an ammeter. The capacity of the smaller unit is 0-5 amp at a maximum of 35 volts. The large unit delivers 4-7 amp at a maximum of 40 volts. (See Appendix C for the schematic of the circuitry.) During the electrolysis process, the electrodes are connected in series; thus all cells receive the same current. Banana plugs and jackets are used for the electrical wire connections (Figure 1B and 2).

*COOLING BATH.* Several investigators have found that the electrolysis process is most efficient if carried out at reduced temperature ( $\sim 0^{\circ}\text{C}$ ), (Roy, 1962; Hoffman and Steward, 1966). Thus the cells are placed in a cold-water bath, a converted hydrocollator with thick insulated walls, maintained at  $2 \pm 0.5^{\circ}\text{C}$ . A 0.8 h.p. refrigeration pump circulates cooling gas through copper tubes coiled along the inner walls of the tank.

An electric heating element is installed on the bottom of the tank making a more precise temperature control possible. This heater operates in conjunction with a thermistor placed in the water. The supply of current to the heater is proportional to the temperature differential as sensed by the thermistor. (See Appendix C for schematic of the circuitry.)

A heavy duty stirrer does the necessary mixing and aids in cooling the cells.

### Distillation System

*GLASSWARE.* Upon the completion of the electrolysis process a few milliliter of enriched sample water is left in each cell. This water must be quantitatively distilled so it can be weighed and prepared for counting. Figures 3 and 4 shows the arrangement used for this distillation, performed under reduced pressure. The receiving unit can be cooled by using a Dewar flask with liquid nitrogen.

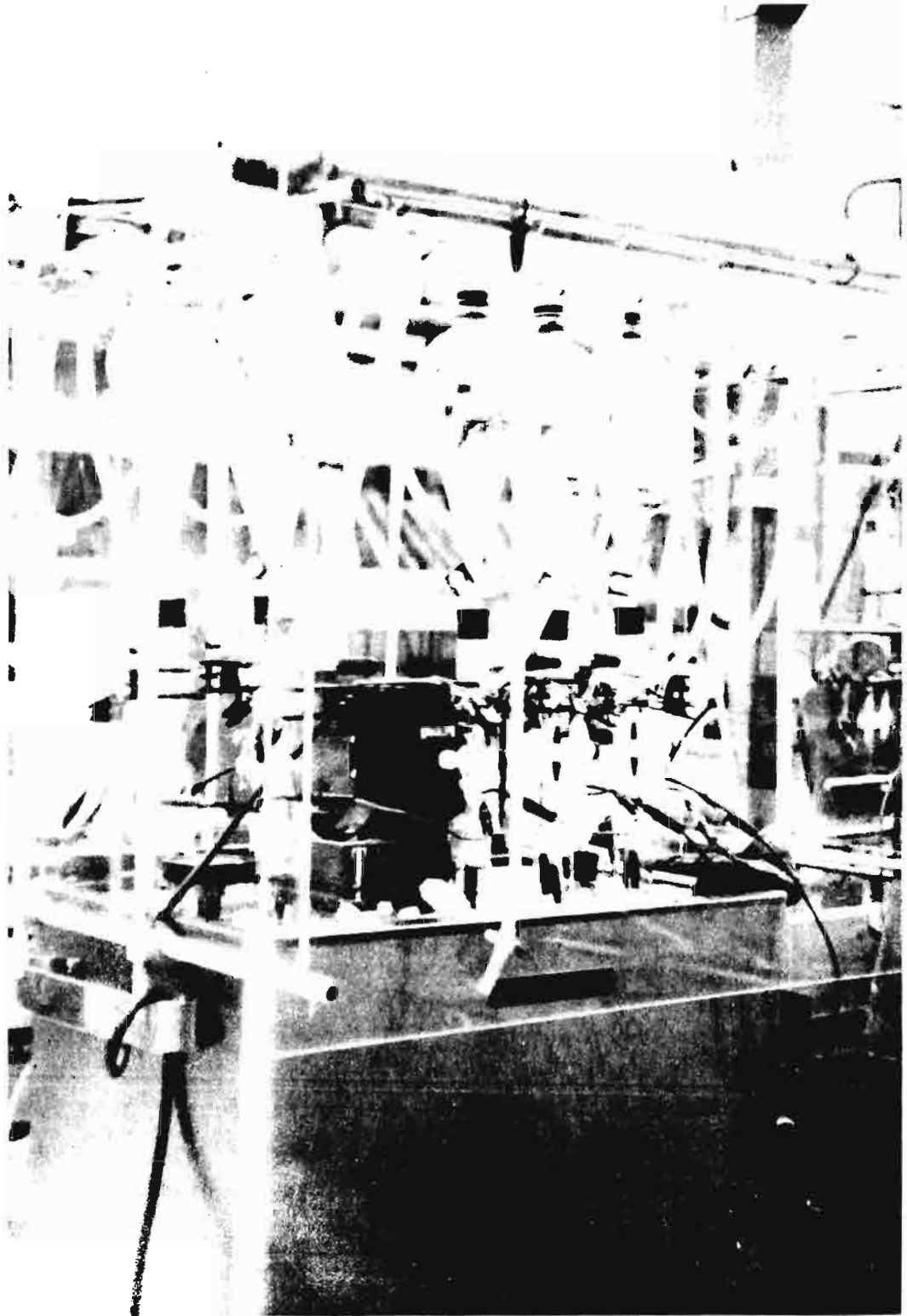


FIGURE 2. ELECTROLYTIC ENRICHMENT SET-UP.

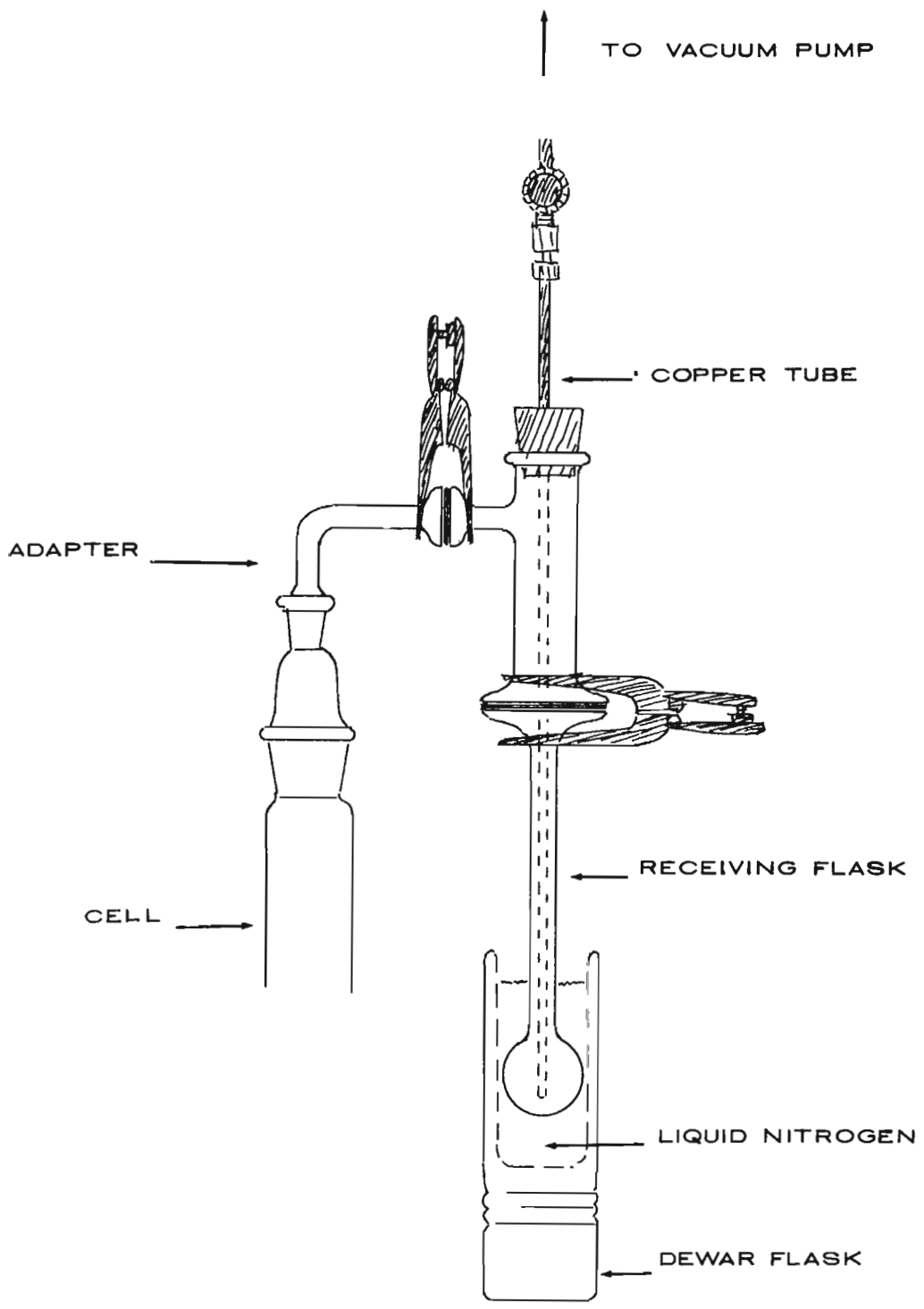


FIGURE 3. ASSEMBLY OF ADAPTER AND RECEIVING UNIT FOR THE DISTILLATION OF ENRICHED SAMPLE.

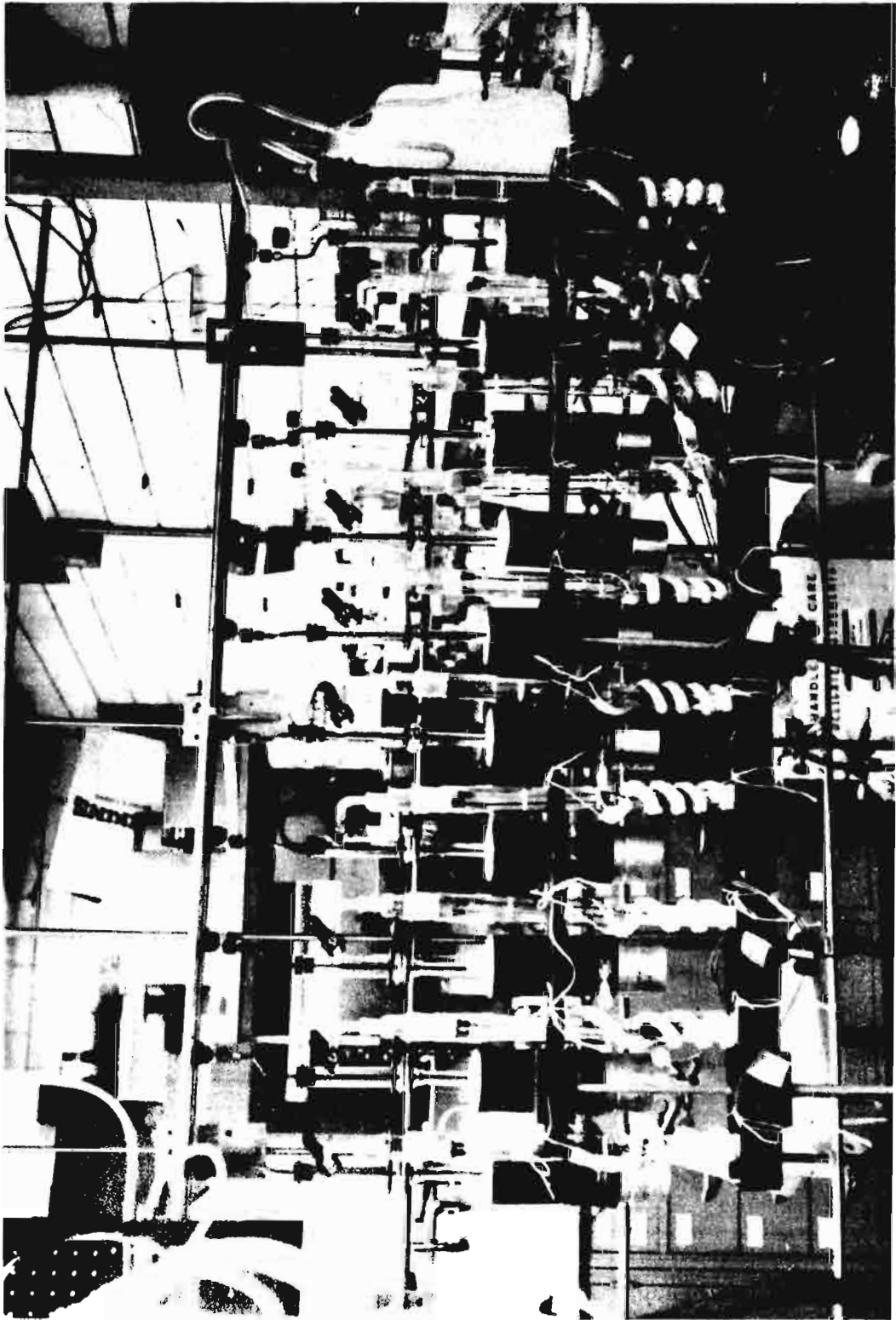


FIGURE 4. DISTILLATION ASSEMBLY.

*VACUUM LINE.* The low pressure required during the distillation is obtained through the use of a 25 l/min vacuum pump. This pump is connected to a stainless steel vacuum line which is provided with several outlets. Each outlet consists of a bellows valve and a long copper tube that extends well into the receiving flask. Each distillation unit can thus be evacuated separately and isolated by means of the valves. The vacuum line is provided with an extra outlet to break the vacuum at the completion of the distillation process. A glass tube with moisture-absorbing material is connected to this outlet to avoid contamination of the samples by atmospheric tritium.

### Radioactive Assay Equipment

The radioactive assay of the enriched samples is performed with a Beckman CPM-100 liquid scintillation counter. A detailed description of this sophisticated instrument is outside the scope of this report and is thus omitted here. A brief description of the system has been given by Lau and Hailu (1968).

Before the enriched samples can be counted in a liquid scintillation counter they have to be mixed with a scintillation solution and placed in special vials. The main component of a scintillation solution is an organic solvent such as dioxane or toluene. To this solvent are added small amounts of organic compounds which are designated as primary and secondary solutes. Often a solubilizer must be incorporated as well to make the solution miscible with water.

The function of the scintillation solution is to absorb the energy of the beta particles emitted by the decaying tritium atoms, and in turn to re-emit this energy as photons of a wavelength which can be detected and processed by the phototubes and photomultipliers in the counter. The mechanism by which a scintillation solution transforms the beta-ray energy into a burst of photons is rather complicated. Basically the beta-particle energy is absorbed by the solvent molecules and subsequently transferred to the primary and secondary solute

molecules which in turn release this energy in the form of photons which are picked up by the phototubes. The purpose of the solutes is to shift the energy released by the solvent to a wavelength that corresponds to the maximum sensitivity of the phototubes (Hayes, 1956).

A substance that interferes with the energy transfer processes in a scintillation solution is called a quencher. Quenchers lower the counting efficiency of the system and thus its detection capability. In counting scintillation mixtures containing water, the most important quenchers are dissolved oxygen and the water itself. Water quenching can be reduced considerably through the addition of naphthalene when dioxane-based mixtures are used. (Hayes, 1956; Baxter, *et al.*, 1964).

Background noise in liquid scintillation counting is rather high. Most of this noise is caused by random radioactivity, electronics, counting vials, and phosphorescence.

The main source of random activity is cosmic rays, which upon interaction with air and other matter, produce electrons, gamma rays, x-rays, etc. Lead shields and special cosmic ray guard detectors can be placed around the counting chamber to reduce random activity to lower levels (Nuclear Chicago, 1966).

Electronic noise is caused by the photomultiplier tubes. Reduction of phototube noise is difficult due to the required sensitivity of the photocathodes and the high currents under which they operate.

Counting vials made of glass contain small amounts of naturally occurring  $^{40}\text{K}$  which emits beta and gamma rays. Borosilicate vials with low potassium content are therefore most commonly used. Plastic vials are also in use but their permeability to solvents like toluene requires prompt counting (Nuclear Chicago Corp., 1966).

Phosphorescence is the delayed emission of light after excitation. It can cause erroneously high counting rates if the samples are exposed to light just before they are counted. This effect is most pronounced when glass vials are used. A few hours dark adaptation will eliminate contributions from this source (Beckman, no date).



## Additional Special Equipment

Sample water of low tritium content can very easily be contaminated by atmospheric water vapor of higher tritium content. Distillation of samples prior to electrolysis is therefore done in an air-tight distillation apparatus with provisions for argon flushing.

A glove box filled with nitrogen gas is used for the preparation of NaOH solution from  $\text{Na}_2\text{O}_2$  and tritium-free water. Commercial NaOH is not used in order to avoid possible contamination from chemically bound tritium (Ostlund, 1966).

## ANALYTICAL PROCEDURE

### Electrolytic Enrichment

The method employed by the Water Resources Research Center laboratory for the enrichment of tritium in water samples is called "Periodic Addition Electrolysis." This method was first tested and described by Ostlund and Werner (1962) and has since been adapted by several tritium laboratories.

At the start of the electrolysis, each cell contains 75.0 ml of sample water and 2.5 ml of 30-percent NaOH as the electrolyte. The rest of the sample (300 ml) is contained in the funnel mounted on top of the cell. After 24 hours, at a current of 4.5 amps, the water is reduced to 37.5 ml. From the funnel a fresh 37.5-ml portion is withdrawn thus bringing the volume in the cell back to the 75.0-ml mark. This procedure is repeated until the funnel is empty. When the residual volume in the cell is 37.5-ml the current is lowered to 2.25 amps and when 18.75 ml remains the current is lowered to 0.45 amps. The electrolysis is stopped as soon as the water level in any one of the cells reaches the 5-ml level. The original sample is thus reduced by a factor of about 75, the entire process taking 13 to 14 days.

In order to avoid atmospheric contamination, the cells are flushed with argon before use as well as during sample-water addition. All gases are vented out of the laboratory through plastic pipes.

## Distillation

Before the enriched samples can be distilled they must be neutralized to release any tritium chemically bound in the NaOH. To this end, CO<sub>2</sub> gas is bubbled through the samples by means of an appropriate arrangement of copper and rubber tubing.

After neutralization each cell is provided with a distillation head, the electrodes remaining inside the cell. A glass connector is then fitted between each cell and its receiving unit by means of an o-ring seal and a clamp. Another o-ring seal and clamp holds the pre-weighed receiving flask in position. The distillation is carried out at a reduced pressure of  $10^{-4}$  mm Hg.

During the distillation each receiving flask is surrounded by a dewar flask containing liquid nitrogen. Heat is applied to the lower part of each cell by a heating tape. Heating aids the distillation and converts NaHCO<sub>3</sub> into Na<sub>2</sub>CO<sub>3</sub>.

## Counting

Upon completion of the distillation, the heating tapes are disconnected and the Dewar flasks removed. When the distilled samples are at room temperature the vacuum in each unit is broken by slowly opening the bellows valves. The receiving flasks are then weighed to determine the final water volumes.

From each flask an aliquot of 4 ml is withdrawn and placed in a counting vial. To each vial 15 ml of scintillation solution are added and the vials are capped and shaken until a clear mixture results. (For the composition of the scintillation solution see "Counting System.") Then the vials are placed in the liquid scintillation counter and dark-adapted for 24 hours. This dark adaptation eliminates counting errors caused by phosphorescence of the vials mentioned before.

## CALIBRATION PROCEDURES

### Electrolysis System

The electrolytic enrichment process is based on the different rates of electrolytic dissociation of H<sub>2</sub>O and HTO. In mathematical form this can be expressed as,

$$\frac{dH}{H} = \beta \frac{dT}{T},$$

where H = H<sub>2</sub>O, T = HTO, and  $\beta$  = the enrichment factor.

Obviously the enrichment factor,  $\beta$ , depends upon the efficiency of the electrolysis process and the liquid volume reduction involved. Thus calibration experiments must be carried out first. To this end water samples of known tritium content were processed and the enrichment factors calculated. The results of the calibration experiments are summarized below.

CELL	A	B	C	D	E	F	G	H	I	J
$\beta$	16.5	14.0	18.6	17.1	19.6	17.2	17.7	17.0	16.2	19.6

$$\text{Average } \beta = 17.4 \pm 1.7 \quad (1\sigma)$$

Differences between the values of the various cells are due to variations in the electrode surfaces, as well as their spacings. The average  $\beta$  value given above compares favorably with that reported by Ostlund (1966). For 100 times enrichment he found:

$$\beta = 16.0 \pm 1.1 \quad (1\sigma)$$

It should be noted that this value was obtained from 23 runs of known activity. The volumes and currents used by Ostlund were 250/2.5 ml and 3.0/0.3 amps.

One sample of known tritium content is always included when analyzing regular water samples. The  $\beta$  value obtained from this "electrolysis standard" is then used for the calculation of the sample water activities.

As a check on possible contamination from handling, etc. a sample of "tritium-free" water is processed in each run as well.

### Counting System

As was mentioned in a previous section, the enriched water samples are mixed with a scintillation solution before being counted.

Several experiments have been carried out to determine the most favorable composition of the scintillation solution. The solution used consists of:

600 ml	Dioxane	-	Solvent
60 g	Naphtalene	-	
3.6 g	PPO	-	Primary Solute*
0.18 g	MePOPOP	-	Secondary Solute*

The counting efficiency for a 4-ml water sample mixed with 15 ml of the above solution is 20 percent. (Glass-counting vials). Other mixtures are still being prepared and tested.

Counting efficiency determination is performed using a mixture of scintillation solution and water of known tritium content. The efficiency obtained from this "counting standard" is used for the calculation of the sample-water activities. The preparation and activities of the counting standard and the electrolysis standard is discussed in the next section, "Preparation of Standards."

The measurement of the background noise is very important when counting samples of low activity. With each set of samples a counting blank is included, prepared from tritium-free water and scintillation solution. The chemical composition of the samples and the blank is thus the same. In the computation of the sample activities the counting rate of the blank is subtracted from that of the sample to

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\* See Appendix A for further information

give the net-counting rate. Typical background counting rates obtained in the WRRRC laboratory are 10-10.5 CPM/ml (Counts Per Minute / ml of H<sub>2</sub>O) using glass vials and the mixtures described previously.

The minimum detection capability of electronic equipment depends upon the relative magnitude of the signal compared to the noise. Due to the low counting efficiency and rather high background noise, the signal to noise ratio for liquid-scintillation counting is not as favorable as one would like. The lower limit for tritium detection is therefore rather high. With a counting efficiency of 20 percent and a background of 10.5 CPM/ml the minimum detectable activity is 0.6 DPM/ml (Disintegrations Per Minute / ml of H<sub>2</sub>O). If electrolytic enrichment is carried prior to counting, the minimum detectable activity of the original sample is 0.02 DPM/ml.

#### Preparation of Standards

Both the electrolysis standard and the counting standard have been prepared using tritium-free water and tritium standard No. 4927 from the National Bureau of Standards (NBS). As of June 3, 1969 the activity of the electrolysis standard was 26.32 DPM/ml and that of the counting standard 2535 DPM/ml. These standards were cross-checked with the standards used in the tritium laboratory of the United States Geological Survey in Washington, D. C.

Tritium-free water was obtained from two sources:

- a) U.H. Well T-133, Ewa Beach, Oahu (1100 feet) and
- b) Deepsea water, offshore Oahu at 22°N 158°W (6560 feet).

Both the well water and the seawater were analyzed along with "zero-tritium" water from the USGS tritium lab. The final counting rates of the duplicate samples showed that they were all within 3 percent of the background counting rate. Both waters are thus considered tritium-free for our purposes.

## SAMPLE CALCULATIONS

For the determination of the activities of a set of samples the following data must be obtained:

From the counting standard (cs):

activity on counting date - DPM/ml (cs)  
counting rate - CPM/ml (cs)

From the electrolysis standard (es):

initial activity on counting date -  $DPM_o$ /ml (es)  
final counting rate - CPM /ml (es)  
initial volume -  $V_o$  (es)  
final volume -  $V$  (es)

From the samples:

final counting rate - CPM/ml  
initial volume -  $V_o$   
final volume -  $V$

(All counting rates refer to "net counting rates")

On the basis of the above data, the calculations can be carried out as outlined below.

*COUNTING EFFICIENCY.* From the activity of the counting standard and its counting rate the counting efficiency is calculated using:

$$CE = \frac{CPM/ml (cs)}{DPM/ml (cs)} \times 100\% . \quad (1)$$

The counting efficiency, CE , thus calculated is taken to be the same for all counting samples, hence no subscript is used.

*FINAL ACTIVITY - ELECTROLYSIS STANDARD AND WATER SAMPLES.* Once the counting efficiency is known the final activities of the electrolysis standard and the water samples can be calculated from their respective final counting rates. Thus for the electrolysis standard:

$$DPM/ml (es) = \frac{CPM/ml (es)}{CE} \times 100\% , \quad (2)$$

and for the samples:

$$\text{DPM/ml} = \frac{\text{CPM/ml}}{\text{CE}} \times 100\%. \quad (3)$$

*ELECTROLYSIS EFFICIENCY - ELECTROLYSIS STANDARD.* The electrolysis efficiency is defined as:

$$\text{EE} = \frac{\text{final activity}}{\text{initial activity}} \times \frac{\text{final volume}}{\text{initial volume}} \times 100\%, \quad (4)$$

hence for the electrolysis standard:

$$\text{EE} = \frac{\text{DPM /ml (es)}}{\text{DPM}_0/\text{ml (es)}} \times \frac{V (es)}{V_0 (es)} \times 100\%. \quad (5)$$

*ENRICHMENT FACTOR,  $\beta$ .* The enrichment factor,  $\beta$ , can now be calculated using the following relation:

$$\log \text{EE} = - \frac{1}{\beta} \log \frac{\text{initial volume}}{\text{final volume}}. \quad (6)$$

(A derivation of this equation is given in the appendix.)

For the electrolysis standard this becomes upon rearrangement:

$$\beta = \frac{- \log \frac{V_0 (es)}{V (es)}}{\log \text{EE}}. \quad (7)$$

*ELECTROLYSIS EFFICIENCY - SAMPLES.* Once the enrichment factor,  $\beta$ , is known, the electrolysis efficiency for each sample can be calculated using equation (6) and the individual sample volumes involved, thus:

$$\log \text{EE} = - \frac{1}{\beta} \log \frac{V_0}{V}. \quad (8)$$

*ACTIVITY-ORIGINAL SAMPLES.* The initial activity of the samples can now be calculated from their final activities, respective electrolysis efficiency values and sample volumes. Hence:

$$\text{DPM}_0/\text{ML} = \frac{\text{DPM/ml}}{\text{EE}} \times \frac{V}{V_0} \times 100\%. \quad (9)$$

*CONVERSION TO TRITIUM UNITS.* The tritium activity of natural water samples is usually expressed in terms of tritium units where: 1 TU = 0.0072 DPM/ml.

*RADIOACTIVE DECAY CORRECTION.* Due to the radioactive decay of tritium, it is necessary to frequently update the activity of the standards. For this correction the following equation is used:

$$A = A_0 e^{-\lambda t}, \quad (10)$$

in which: A = activity on counting date,  $A_0$  = last assay date, t = time elapsed since last assay,  $\lambda = 0.693/12.26$  years.

This same equation can be used to correct sample activities if more than a month has elapsed since their collection. If equation (10) is used for sample activity correction, the symbols will read: A = activity on counting date,  $A_0$  = activity on sampling date, and t = time elapsed since sampling.

The counting efficiency as computed from the counting standard is taken to be the same for all counting samples. This procedure is considered justified because all counting samples are chemically identical and the quench factors recorded by the counter are the same within 1 percent.

For the electrolysis process it is the enrichment factor,  $\beta$ , that is taken to be the same for the samples and the standard. In calculating the error level on sample activities, the variation in the values is taken into account.

## COLLECTION OF SAMPLES

Rainwater samples from various rain-gages on the Koolau range were provided by the Board of Water Supply, City and County of Honolulu (Figure 5). The Board of Water Supply used standard non-recording copper rain-gages to collect samples which were stored in 500-ml brown glass bottles.



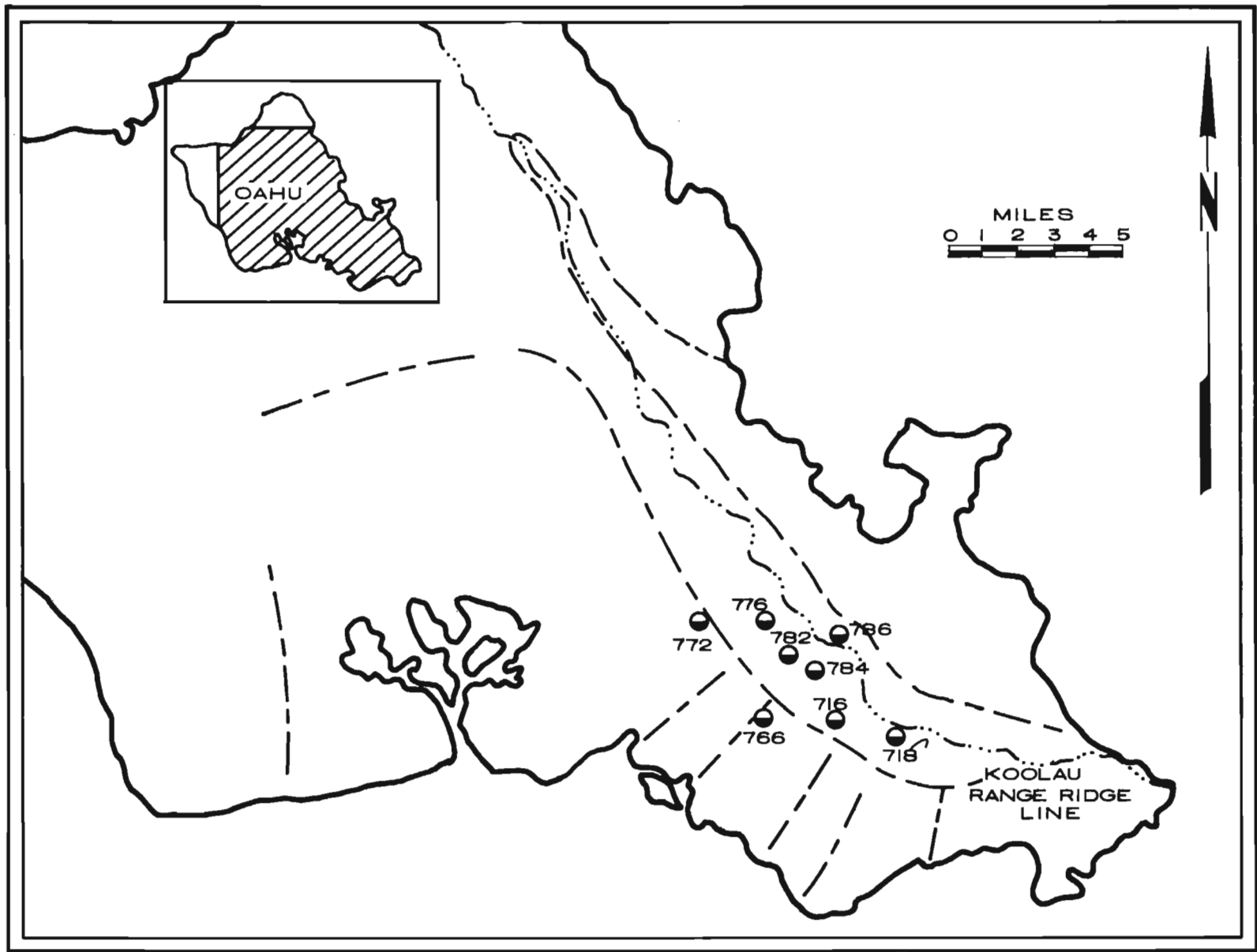


FIGURE 5. RAINWATER SAMPLING POINTS ON THE KOOLAU RANGE, OAHU.

The stream, spring, and well water samples were collected by WRRC personnel. Figure 6 shows the Pearl Harbor area with the various locations and the type of water obtained at each point. The collection of the well water was performed at two depths using a 500-ml sampler. Extra care was taken to keep the exposure of the samples to atmospheric air at a minimum. Composite samples were taken from the streams and the springs. All samples were placed in 1000-ml nalgene bottles and capped tightly immediately after filling and stored in a dark and cool closet until they were analyzed. (A detailed account of the sampling method is given by Tenorio, Young, and Whitehead, 1969.)

#### ANALYTICAL DATA

Analytical data are listed in Tables 1, 2, and 3. In the calculation of the accuracy of these data, the maximum error associated with each measurement was taken into account along with a 1 $\sigma$  error on the counting rates.

The activity of the 20 rainwater samples ranges from  $16.3 \pm 2.5$  TU to  $28.2 \pm 3.1$  TU. There are no marked differences between the samples from one month to another or between the various stations. For comparison some data were taken from the report by Lau and Hailu (1968) and listed in Table 4. The present tritium activity of Oahu rainwater, as reflected by our new data, indicates a lower level than that prevailing in 1964-65. The tritium content of the four stream water samples ranges from  $14.5 \pm 2.4$  to  $21.3 \pm 2.9$  which is of the same order of magnitude as the rainwater samples. The two spring water and ten well water samples however are all below 5 TU. Hydrologic interpretation of the data herein presented is being made as additional data are continually obtained.

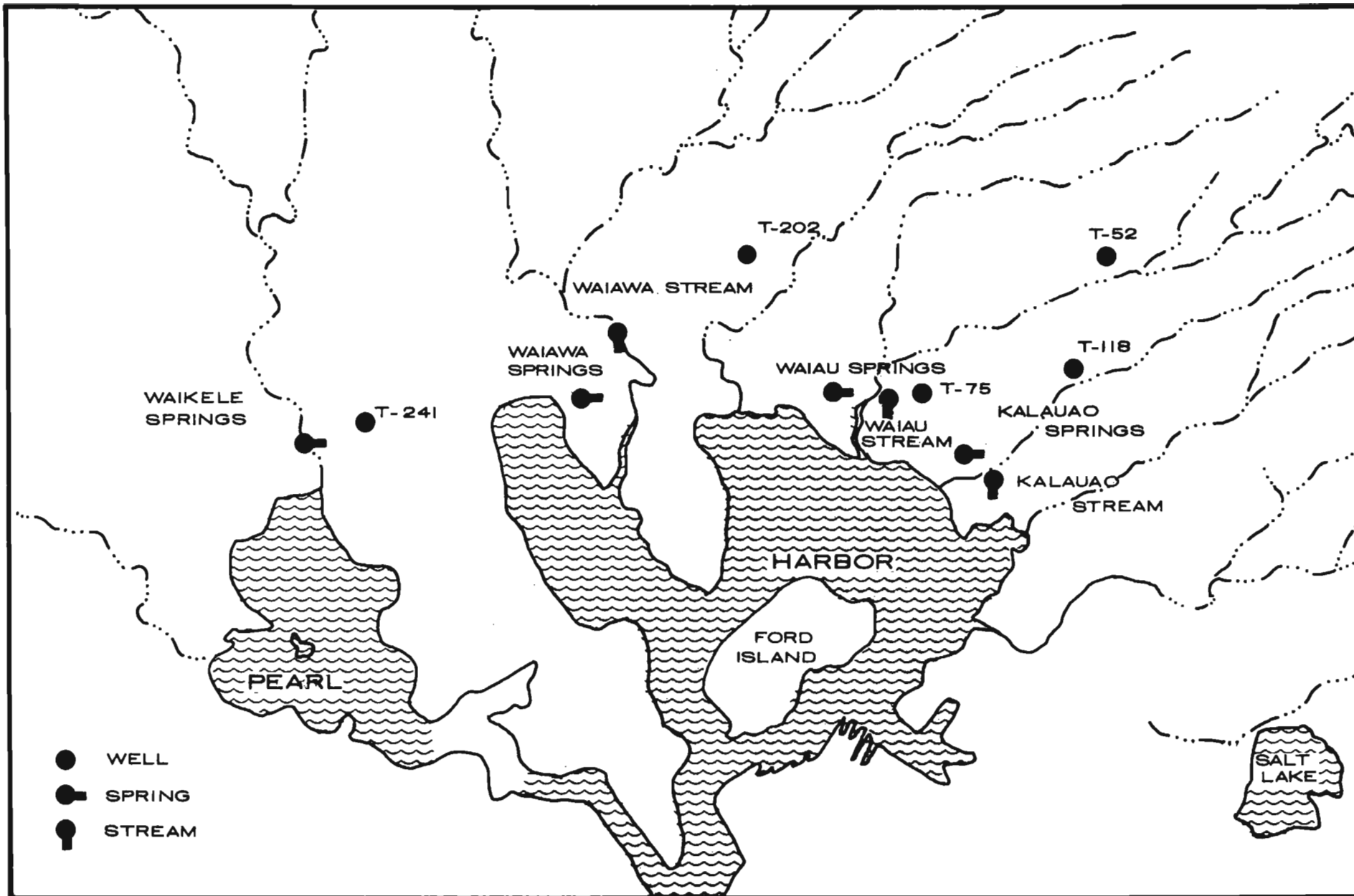


FIGURE 6. SAMPLING POINTS IN THE PEARL HARBOR AREA, OAHU.

TABLE 1. ANALYTICAL DATA OF RAINWATER ON OAHU.

TABLE 1. ANALYTICAL DATA OF RAIN WATER ON OAHU  
(COMPOSITES OF 2 - 4 WEEKS)

STATION	SITE	DATE OF COLLECTION	ACTIVITY IN T.U.
706	PACIFIC HEIGHTS	3- 3-69	23.6 ± 2.7
706	PACIFIC HEIGHTS	4- 1-69	28.2 ± 3.1
782	LOWER LUAKAHA	"	19.2 ± 2.5
786	NUUANU PALI	"	22.9 ± 3.1
706	PACIFIC HEIGHTS	5- 5-69	18.8 ± 2.6
782	LOWER LUAKAHA	"	23.6 ± 2.9
786	NUUANU PALI	"	21.5 ± 3.1
716	MANOA VALLEY	5-20-69	23.6 ± 2.5
718	PALOLO VALLEY	"	22.0 ± 2.9
776	KALIHI TUNNEL	"	19.7 ± 2.7
706	PACIFIC HEIGHTS	6-16-69	24.6 ± 3.4
786	NUUANU PALI	"	19.3 ± 2.8
706	PACIFIC HEIGHTS	7- 7-69	23.0 ± 2.4
782	LOWER LUAKAHA	"	22.9 ± 2.4
786	NUUANU PALI	"	23.8 ± 2.4
776	KALIHI TUNNEL	8- 1-69	17.9 ± 2.5
782	LOWER LUAKAHA	8- 4-69	16.3 ± 2.5
706	PACIFIC HEIGHTS	"	19.5 ± 2.6
718	PALOLO VALLEY	8-26-69	25.1 ± 2.7
772	MOANALUA VALLEY	8-18-69	20.6 ± 2.9

TABLE 2. ANALYTICAL DATA OF STREAM  
AND SPRING WATER ON OAHU.

TABLE 2. ANALYTICAL DATA OF STREAM AND SPRING WATER ON OAHU

SITE	DATE OF COLLECTION	ACTIVITY IN T.U.
KALAUAO STREAM	5- 6-69	21.3 ± 2.9
WAIAWA STREAM	"	14.5 ± 2.4
WAIMALU STREAM	"	20.8 ± 2.6
WAIKILE STREAM	"	18.5 ± 2.3
WAIANA SPRING	"	0.3 ± 2.1
WAIKIAU SPRING	"	0.3 ± 2.8

TABLE 3. ANALYTICAL DATA OF WELL WATERS LOCATED IN THE PEARL HARBOR AREA, OAHU.

TABLE 3. ANALYTICAL DATA OF WELL WATERS LOCATED IN THE PEARL HARBOR AREA, OAHU

WELL	DEPTH	COLLECTED	ACTIVITY IN T.U.
T- 75	237'	5- 6-69	1.0 ± 1.9
"	80'	"	1.3 ± 1.8
T-118	300'	5- 6-69	1.5 ± 1.8
"	220'	"	2.4 ± 1.9
T- 52	260'	5- 8-69	2.9 ± 1.9
"	185'	"	3.1 ± 1.9
T-202	360'	5- 8-69	0.5 ± 2.1
"	28'	"	3.5 ± 3.3
T-241	180'	5- 6-69	1.6 ± 2.1
"	52'	"	1.1 ± 2.0

TABLE 4. LITERATURE DATA OF RAINWATER ON OAHU.

TABLE 4. LITERATURE DATA OF RAINWATER ON  
21°45'N-157°45'W

(MONTHLY COMPOSITES IN TRITIUM UNITS)

YEAR	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JUL.	AUG.	SEPT.	OCT.	NOV.	DEC.
1964	115	147	165	162	120	103	159	96	61	94	51	36
1965	109	92	106	81	56	88	43					

## ACKNOWLEDGEMENTS

The authors would like to thank Mr. Clarence Williams for his high quality glass blowing work, Mr. William Barnes for the building and revision of the electrical circuits and Dr. Donald Gordon for providing us with the deep-sea water. Many thanks also go to Mr. Theodore Jordan of the Hawaii Institute of Geophysics Machine Shop for his help in solving many problems associated with instrumentation and hardware, as well as to Mr. Dale Hammond for his many helpful suggestions.

Deep appreciation also goes to Mr. Frank Brookman of the U. S. Geological Survey Tritium Laboratory for his many services and suggestions.

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## APPENDICES



## APPENDIX A

Abbreviations used for components of scintillation solution.

PPO = 2, 5- Diphenyloxazole

MePOPOP = 1, 4-Bis [2-(4-methyl-5-phenyloxazolyl)] benzene.

## APPENDIX B

Derivation of enrichment factor equation.

The expression relating the electrolysis efficiency and the enrichment factor,  $\beta$ , is derived from the linear differential equation

$$\frac{dH}{H} = \beta \frac{dT}{T}$$

This equation can be integrated over the limits  $H_0$  to  $H$  and  $T_0$  to  $T$ .

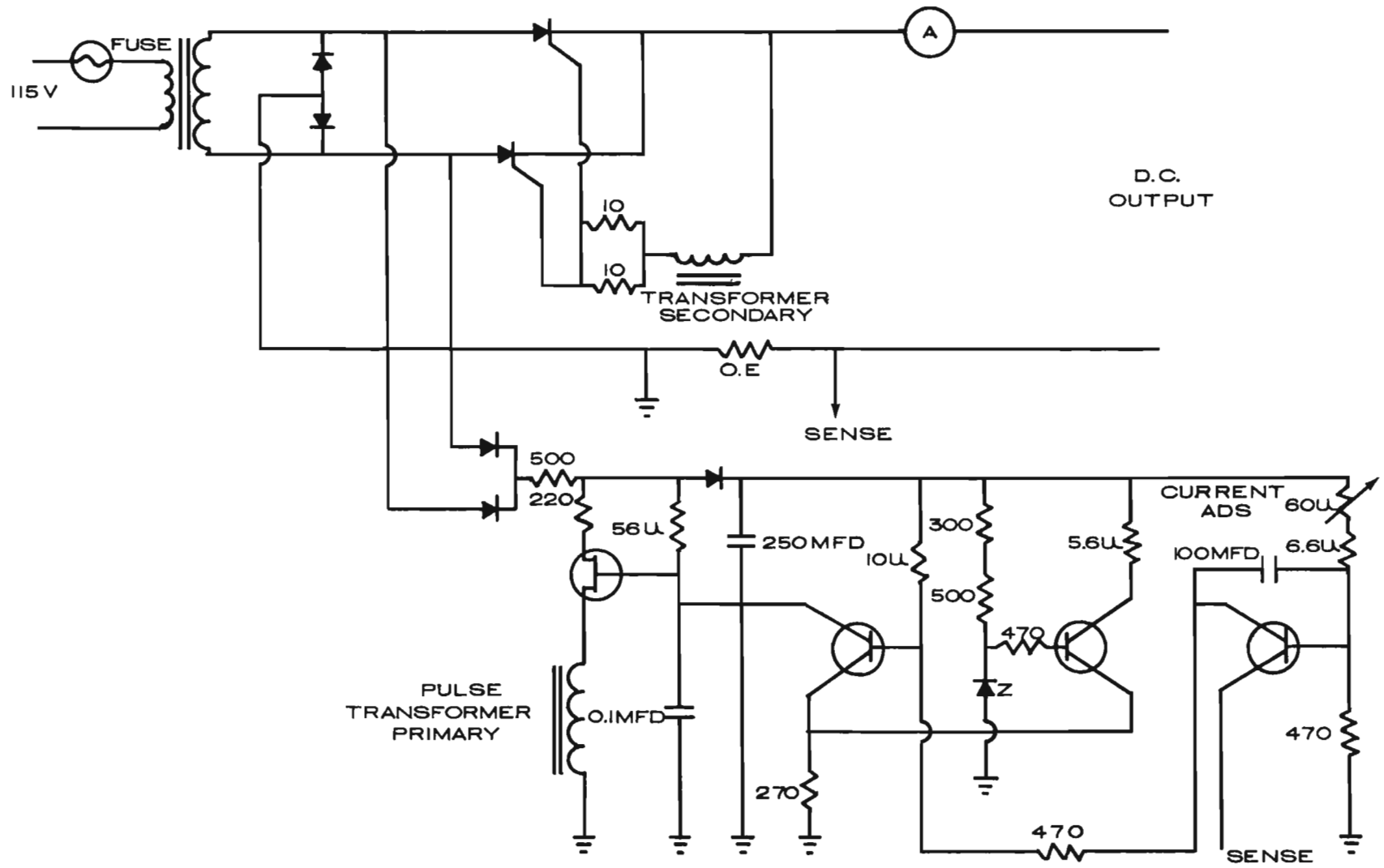
$$\log H \Big|_{H_0} = \beta \log T \Big|_{T_0} \quad \text{Hence} \quad \log \frac{H}{H_0} = \beta \log \frac{T}{T_0}$$

Since at any time during the electrolysis process the bulk of the water consists of  $H_2O$  it is quite valid to substitute the volume ratio  $V/V_0$  for that of the hydrogen atom ratio  $H/H_0$

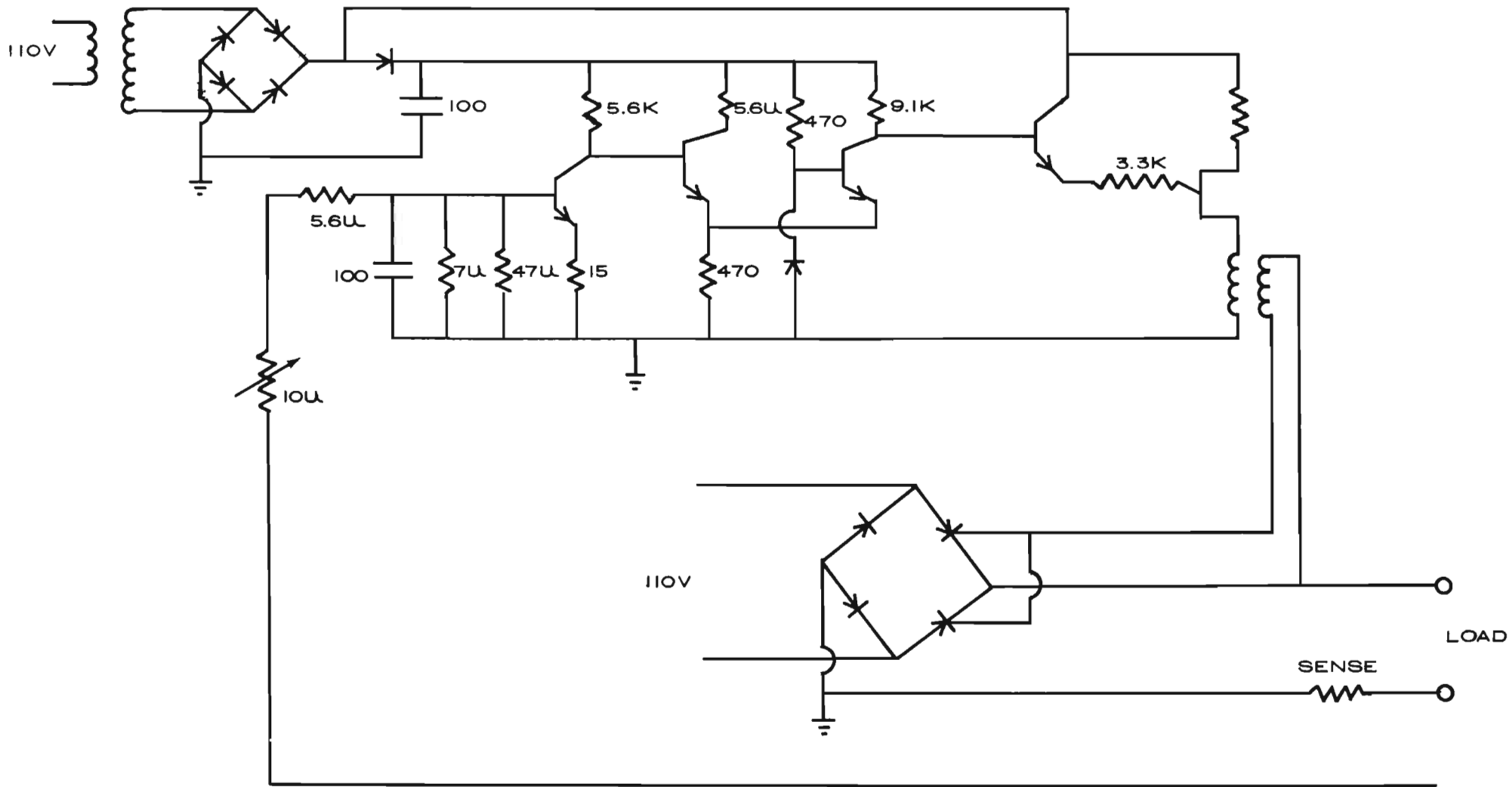
$$\text{So: } \log \frac{V}{V_0} = \beta \log \frac{T}{T_0}$$

The ratio  $T/T_0$  has been defined as the electrolysis efficiency and therefore can be substituted by it. Thus upon substitution and rearrangement:

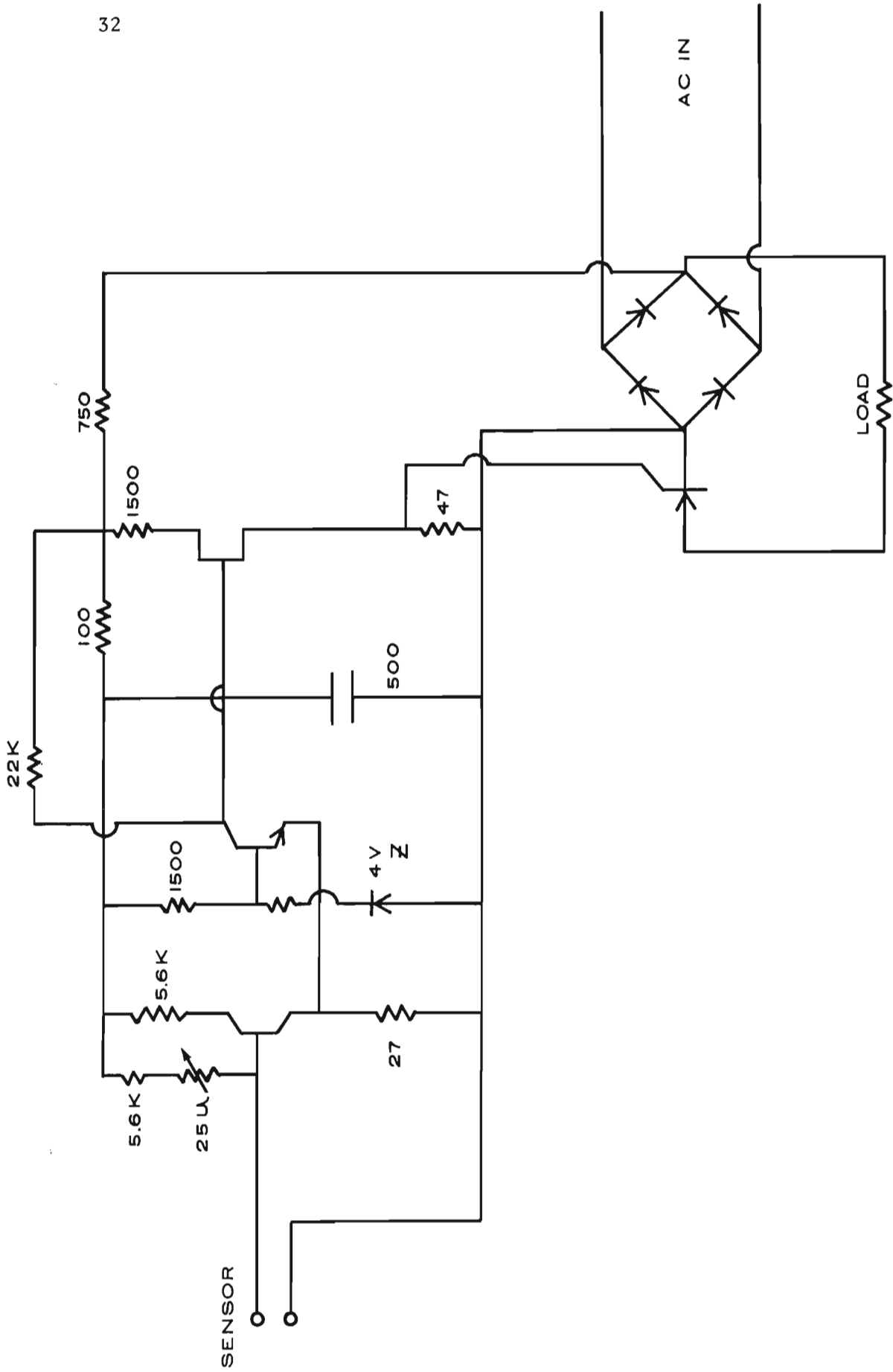
$$\log EE = - \frac{1}{\beta} \log \frac{V_0}{V}$$



APPENDIX C-1. CONSTANT CURRENT POWER SUPPLY CIRCUIT: 0-5 AMP.



APPENDIX C-2. CONSTANT CURRENT POWER SUPPLY CIRCUIT: 4-7 AMP.



APPENDIX C-3. BATH TEMPERATURE CONTROL CIRCUIT.