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MEASUREMENT OF LOW LEVELS OF DISSOLVED RADON AND RADIUM IN WATER

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ABSTRACT

Standard method of measuring dissolved radon in water consists of sealing a known volume of water sample in a jar, measuring airborne radon concentration using an E-PERM radon monitor, and calculating the radon concentration in water. The method is also usable for measuring dissolved radium in water. Anew method described in this paper improves the sensitivity by two orders of magnitude. This method consists of immersing an E-PERM radon monitor encased in a water barrier (thin plastic or tyvek) which permits exchange of radon between water and the detector air volume, and measuring radon gas concentration over the desired time interval. Radon in water is then calculated using appropriate partition constants and other parameters. Results of such measurements agreed well with other standard methods. The method is usable for measuring extremely low levels such as 0.25 to 1.0 pCi/L of dissolved radon. For measuring radium in water, water is filled in a 4 liter glass jar and sparged to remove any radon already present in water. An E-PERM radon monitor encased in a suitable water barrier is immersed in water and the jar sealed. Using appropriate theoretical equation relating the average radon concentration as measured by E-PERM radon monitor and the exposure time, the radium concentration in water can be calculated. Levels as low as 1 to 5 pCi/L is measurable by this method in about 10 days. Ability to measure such low level extends this method to several basic research areas in oceanography and geophysical research.

INTRODUCTION

A small water sample is placed in the bottom of a glass jar. An E-PERM is suspended in the air phase above the water. The lid of the flask is closed and sealed to make it radontight. Radon reaches an equilibrium between the water and air phase. At the end of the desired exposure period, the flask is opened and the E-PERM removed. The average radon concentration in the air phase is calculated using the standard E-PERM procedure. A calculation using this air concentration in conjunction with the other parameters gives the radon concentration of the water. The basis for the analysis is described in detail by P. Kotrappa et al (1) Detailed method of sampling and analysis is described fully in E-PERM System Manuals(2). Schematic of the method is shown inFigure 1. This method gives results with acceptable accuracy for radon concentrations down to 100 pCi/liter of dissolved radon in water. Following equation gives the parameters for the calculations.

$$Rn\,Water\ (\frac{pCi}{\ell}) = \frac{RCA \times (e^{\lambda \times TD}) \times (\frac{V_a}{V_w} \times OC) \times \lambda \times TA}{(1 - e^{-\lambda \times TA})}$$

where λ is the decay constant for radon

Rn Water is the dissolved radon in water
RCA is the average radon concentration as measured by E-PERM
TD is the delay time in days
TA is the analysis time in days
TD is the delay time in days
V_a is the volume of air in liters
V_w is the volume of water in liters
OC is the Ostwald Coefficient, which as 0.2593 at 68°F
Decay constant for radon is taken as 0.1812 day^{-1}
e stands for exponential function

MEASUREMENT OF DISSOLVED RADON IN WATER BY IMMERSION METHOD

It is possible to extend this method for measuring very low concentrations of dissolved radon in water by simply increasing the volume of water and decreasing the volume of air. As a limiting case, the air volume is the air volume of the detector and the water volume could be very large as in the case of an immersion in a large tank. In such case radon in air phase is 4.4 times the radon in water. One can readilly see that 1 pCi/L of radon in water gives leads to 4.4 pCi/L of radon in the air phase of the E-PERM, which is easily measurable in two day measurement using SST E-PERM. In order for this method to work, E-PERMs must be enclosed or sealed in water barrier that lets radon into the air phase of E-PERM. The use of a thin (commercially available half mil thick bags) made of low density polyethylene bags are readilly usable. These could be sealed by a heat sealer. The method is now extended to a measurement of extremely low levels of radon in water. Such measurements are may be too sensitive for measuring radon in well water or in municipal waters. Following table gives the sensitivity achievable by using different E-PERMs in the immersion mode.

Table 1. Lower limits of detection (LLD) using different E-PERM configurations by immersion method. Measurement time: Two days.

E-PERM CONFIGURATION	LLD, pCi/L of dissolved radon in water Two day measurements
SST	1
SLT	12
LST	7
LLT	96
HST	0.25

APPLICATION FOR RENEWING WATER SUPPLY OR CONSTANT RADON LEVELS

Two situations may arise. Water may be flowing through a tank in which the E-PERM is immersed. Then the correction for the decay of radon during measurement is not applicable. Similarly if radon in water is maintained constant by continuous addition of radon into water, the equation(1) is simplified to equation (2)

$$Rn\,Water\ (\frac{pCi}{\ell}) = RCA \times (\frac{V_a}{V_w} \times OQ$$

where λ is the decay constant for radon

EXPERIMENTAL RESULTS

The Department of Oceanography of the State University of Florida maintains large (140 gallon) aquarium tank of water. Please see figure 4. The tank contains a "radon generator", radium absorbed onto exchange resin, which continuously produces a constant amount of radon. This has been in use a reference standard for several applications in their research. The radon in water level is very well characterized using liquid scintillation methods and is held constant at about 139 pCi/L. A set of L chambers sealed in thin polyethylene bags were suspended in water tank and were removed after 2, 3 and 4 days. Results are given in the following table.

Table 2. Measured and reference radon in water using L chamber E-PERMS in immersion mode

Electret Number	Initial volts	Final volts	DV	Exposure days	Radon in air pCi/L	Radon in water pCi/L
LG4343	757	708	49	3	509.69	132.16
LG4382	728	659	69	4	545.77	141.52
SM5701	699	340	359	2	537.43	139.36
SM5669	700	115	585	3	612.51	158.82
Average						142.97
Reference						139

MEASUREMENT OF DISSOLVED RADIUM IN WATER

The method used for measurement of dissolved radon can be adopted for measurement of dissolved radium in water(3). Figure 2 gives a schematic of the measuring arrangement. The principle is simple. If a water sample contains radium (^{226}Ra), then radon is generated in the sample. This radon partitions between air phase and water phase. By measuring average radon concentration in the water phase, it is possible to calculate radium concentration in water. If the dissolved radon is not leaked out of the sample, it is possible to calculate radon concentration in water using the physical laws governing the growth and decay of radon from a known concentration of radium in water.

PROTOCOL FOR MEASURING RADIUM CONCENTRATION IN WATER

1. Heat seal an SST E-PERM in a thin polyethylene bag

2. Lower into the 4 liter jar

3. Sparge water to remove the dissolved radon. Blender can also be used.

4. Fill the jar with water to be tested

5. Close the jar and tighten the collar

6. Allow the jar to stay undisturbed for 10 days

7. Calculate the average radon concentration seen
   by E-PERM

8. Divide the net radon concentration by the calibration factor
(1.6477) to calculate radium concentration in water

The calibration factor CF is defined as the average radon concentration as measured by E-PERMS per unit concentration of radium in water.

$$CF = \frac{ARa_e}{RaC} = \frac{1 - \frac{1 - e^{-\lambda \times T}}{\lambda \times T}}{\frac{V_a}{V_w} + L}$$

λ = *deacy constant for radon*

CF is in units of radon concentration in air per unit concentration of radium in water

ARA_e is the average equilibrium radon concentration in air as measured by E-PERM radon monitor
RaC is the radium concentration in water
V_a is the volume of air cavity
V_w is the volume of water
L is the Ostwald Coefficient

Table 3. Calibration constants for different accumulation times.
Volume of water V_w=3.75 liter Volume of air V_a=0.25 liter

Days	5	10	15	20	25	30
CF	1.0479	1.6477	2.0093	2.239	2.3927	2.5005

Example :

Va = 0.25 L, Vw=3.75 L, T=10 days, L=0.26
CF= 1.6477

EXPERIMENTAL RESULTS

Figure 3 gives the experimental results of the experiments conducted at the State University of Florida. There is a good agreement between the expected and measured radon concentrations.

DISCUSSIONS

Sensitivity can be improved by extending the measurement time. For example, by extending the measurement from 10 days to 30 days, the sensitivity is improved by a factor of 1.5. Further if H chamber is used, in the place of S chamber a further improvement by a factor of 5 is possible. If expected radium concentration is high other suitable E-PERM configuration can be used. This possibility of measuring very low levels of radon and radium in water extends the use of E-PERMs to many areas of basic researches such as the studies of natural waters, sea waters, movement of soils, weatherization and such other studies.

REFERENCES

1 P. Kotrappa and W. A. Jester, "Electret Ion Chamber Radon Monitors Measure Dissolved ^{222}Rn in Water" Health Physics 64:397-405 (1993)

2 E-PERM System Manual (1994) Rad Elec Inc., 5714-C, Industry Lane, Frederick, MD 21703

3 United States Patent 5,055,674 (October 1991) "Electret ion chamber for monitoring radium and dissolved radon in water" Inventor P.Kotrappa; Assignee: Rad Elec Inc., Frederick, MD USA

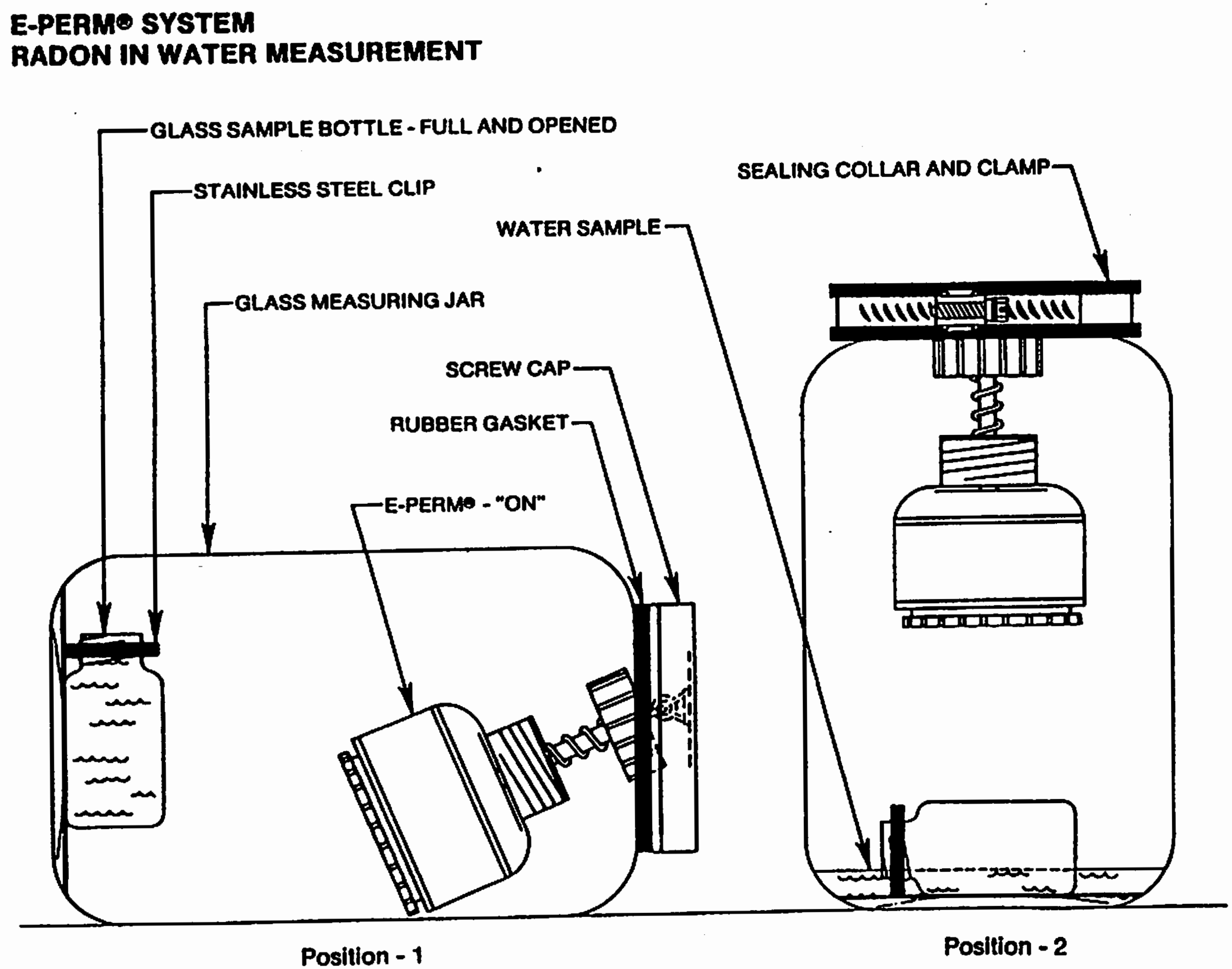
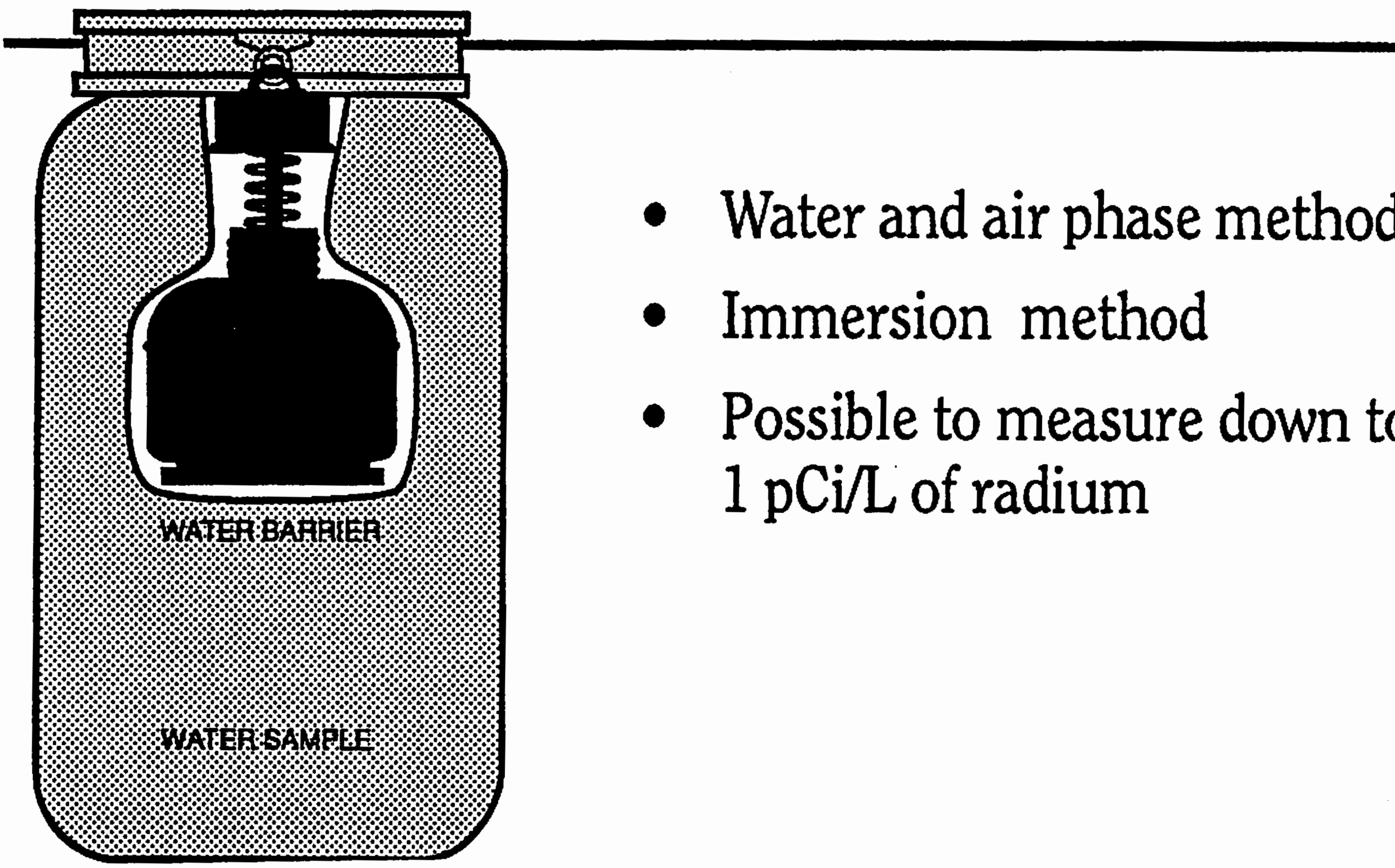


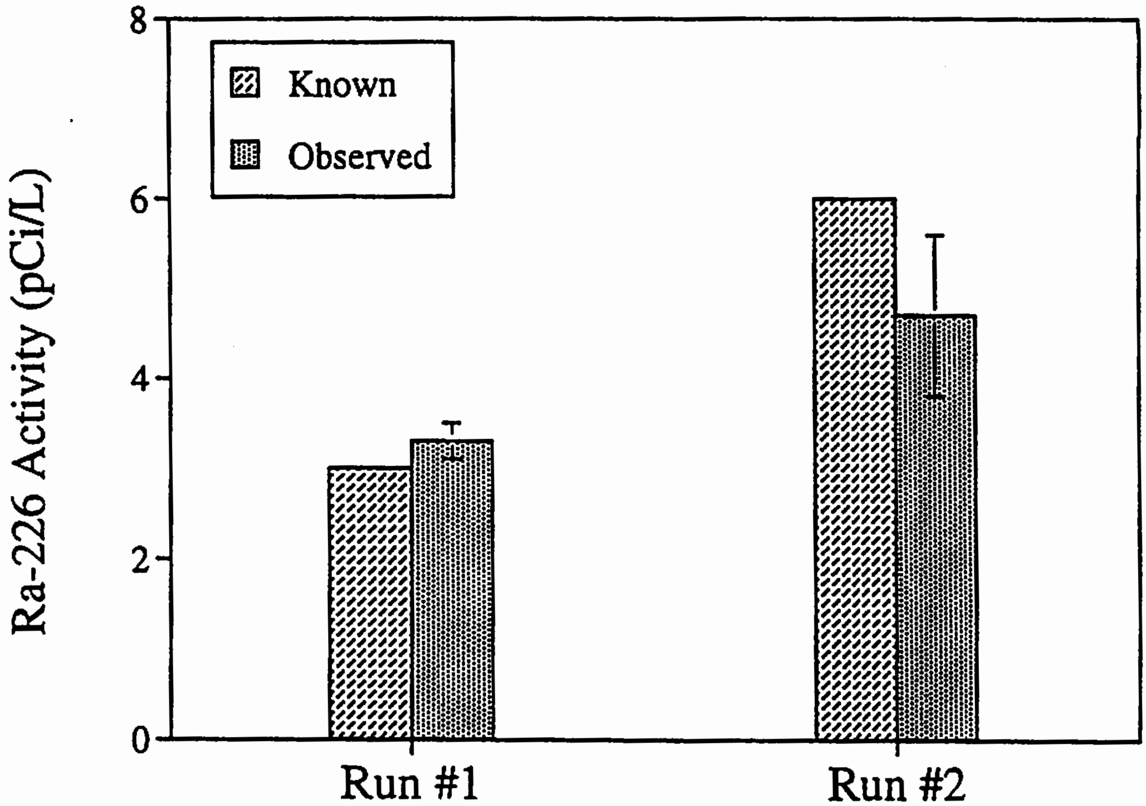
Figure 1
SCHEMATIC FOR MEASUREMENT OF DISSOLVED RADON IN WATER

Radon and Radium in water



- Water and air phase method
- Immersion method
- Possible to measure down tc 1 pCi/L of radium

Figure 2
SCHEMATIC OF MEASUREMENT OF RADON AND RADIUM BY IMMERSION METHOD



Ra-226 in water was analyzed using EIC's and compared to the known value. Each run is a average and standard deviation of four different sampling jars with the same stock solution.

Figure 3
EXPERIMENTAL RESULTS OF MEASUREMENT OF RADIUM IN WATER

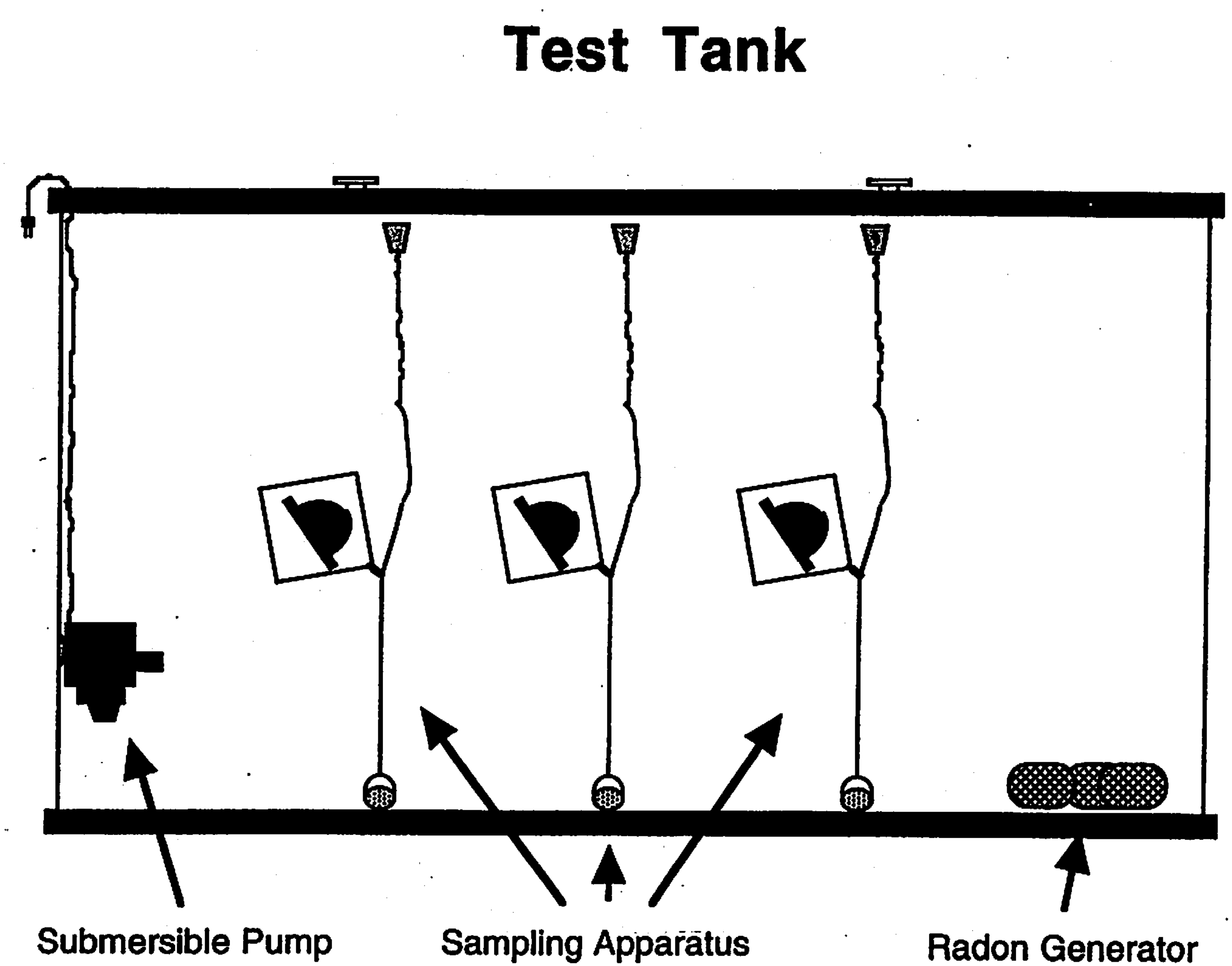


Figure 4
EXPERIMENTAL ARRANGEMENT FOR TESTING IMMERSION METHOD OF MEASURING RADON IN WATER