

**Tritium Instrumentation for a Fusion  
Reactor Power Plant**

K. E. Shank  
C. E. Easterly

**OAK RIDGE NATIONAL LABORATORY**

OPERATED BY UNION CARBIDE CORPORATION FOR THE ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

## **DISCLAIMER**

**This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.**

## **DISCLAIMER**

**Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.**

Printed in the United States of America. Available from  
National Technical Information Service  
U.S. Department of Commerce  
5285 Port Royal Road, Springfield, Virginia 22161  
Price: Printed Copy \$4.50; Microfiche \$2.25

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Energy Research and Development Administration/United States Nuclear Regulatory Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

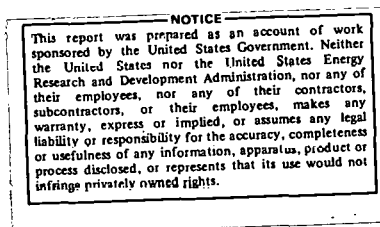
Contract No. W-7405-eng-26

HEALTH PHYSICS DIVISION  
FUSION TECHNOLOGY GROUP

TRITIUM INSTRUMENTATION FOR A  
FUSION REACTOR POWER PLANT

K. E. Shank  
C. E. Easterly

Date Published: September 1976



OAK RIDGE NATIONAL LABORATORY  
Oak Ridge, Tennessee 37830  
operated by  
UNION CARBIDE CORPORATION  
for the  
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

THIS PAGE  
WAS INTENTIONALLY  
LEFT BLANK

## TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT . . . . .	v
1. INTRODUCTION . . . . .	1
2. ON-SITE TRITIUM INSTRUMENTATION . . . . .	4
2.1 Stack Sampling . . . . .	4
2.2 Process Stream and Reactor Coolant Monitoring . . . . .	6
2.3 Occupational Protection . . . . .	8
2.3.1 Real-Time Air Monitoring . . . . .	8
2.3.1.1 Ionization Chambers . . . . .	8
2.3.1.2 Proportional Counters . . . . .	12
2.3.1.3 Scintillation Systems . . . . .	13
2.3.2 Tritium Collection and Conversion Systems . . . . .	14
2.3.3 Surface Monitoring . . . . .	17
2.3.4 Personnel Monitoring . . . . .	20
3. OFF-SITE TRITIUM INSTRUMENTATION . . . . .	23
3.1 Water Sampling . . . . .	23
3.2 Air Sampling . . . . .	24
3.3 Biological Sampling . . . . .	25
4. ASSESSMENT OF INSTRUMENTATION . . . . .	27
4.1 Ion Chamber and Proportional Counter . . . . .	27
4.2 Scintillation Systems . . . . .	30
4.3 Integrating Systems . . . . .	32
5. INSTRUMENTATION FOR A CTR . . . . .	33
6. CONCLUSIONS . . . . .	38
REFERENCES . . . . .	40

THIS PAGE  
WAS INTENTIONALLY  
LEFT BLANK



TRITIUM INSTRUMENTATION FOR A  
FUSION REACTOR POWER PLANT\*

K. E. Shank and C. E. Easterly

Health Physics Division  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

ABSTRACT

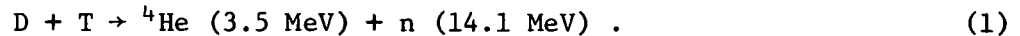
A review of tritium instrumentation is presented. This includes a discussion of currently available in-plant instrumentation and methods required for sampling stacks, monitoring process streams and reactor coolants, analyzing occupational work areas for air and surface contamination, and personnel monitoring. Off-site instrumentation and collection techniques are also presented. Conclusions are made concerning the adequacy of existing instrumentation in relation to the monitoring needs for fusion reactors.

---

\*Research sponsored by the Energy Research and Development Administration under contract with Union Carbide Corporation.

## 1. INTRODUCTION

Due to recent advances in the fusion research and development program, it is desirable to begin examining tritium monitoring and instrumentation needs for a controlled thermonuclear reactor (CTR). Tritium will likely be the principal potential source of ionizing radiation inside the plant, and it will be the dominant radionuclide of concern in maintaining occupational and public exposures as-low-as reasonably achievable during normal operations.<sup>1,2</sup> Regardless of the approach taken to attain fusion power, tritium will be used as fuel by first generation CTR's in the following reaction:



This reaction is the most feasible alternative for first generation reactors because of its relatively low threshold temperature and high energy gain.<sup>2</sup> For D-T fueled reactors, tritium breeding will be necessary to supply fuel for the reactor. This will be accomplished by allowing neutrons from the above reaction to interact with a lithium blanket surrounding the plasma. The following tritium-producing reactions occur:<sup>3</sup>



Large quantities of tritium will be present in various parts of a CTR including the fuel storage and processing system, the fuel injection system, the plasma recovery system, and the breeding blanket and recovery system. Various reactor designs call for 0.4 to 4 kg of tritium (1 g = 9600 Ci) in the blanket and coolant, and a total tritium inventory in

the plant of 2 to 15 kg.<sup>4</sup> (For comparison, tritium appears naturally in the environment at an estimated total worldwide level of 7 kg.<sup>5</sup>)

Tritium may be released during normal operation because of its physical characteristics. Tritium permeates most materials, especially at elevated temperatures, and can diffuse from the blanket-coolant system by three routes: (1) diffusion from the blanket into the plasma vacuum chamber (a recovery system will collect this tritium and recycle it as fuel); (2) diffusion outward through all reactor materials into the reactor hall; and (3) diffusion from the coolant system into the steam system. To help prevent escape by the second route, components will be enclosed within a second containment system which will be maintained at a negative pressure. In the third route, almost all of the tritium will be oxidized, and small amounts of HTO will, subsequently, be lost to the environment. In addition, the radionuclide can leak from other tritium-containing components.

Tritium is a radionuclide which emits a beta particle of only 18.6 keV maximum energy. For this reason, the external radiation hazard to an occupational worker is negligible. Tritiated water, however, is an internal hazard, and it can readily enter the body through the skin.<sup>6</sup> The maximum permissible body burden for tritium is 1 mCi, and the average biological half-life for ingested tritiated water is 9.5 days.<sup>7</sup> The  $(MPC)_w$  for occupational exposure to HTO is  $0.1 \mu\text{Ci}/\text{cm}^3$ . The  $(MPC)_a$  for HTO is  $5 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ , with the  $(MPC)_a$  for tritium gas being a factor of 400 higher.<sup>8</sup>

Fortunately, much of the tritium instrumentation needed for fusion reactors has already been developed in the fission reactor industry.

There has been 20 to 30 years experience in detecting tritium in both the oxide and gaseous forms and in combination with other radionuclides.

## 2. ON-SITE TRITIUM INSTRUMENTATION

The currently available on-site instrumentation for tritium monitoring is discussed in this section. On-site instrumentation includes stack sampling, monitoring process streams and reactor coolants, analyzing occupational work areas for air and surface contamination, and personnel monitoring.

### 2.1 Stack Sampling

Ventilation air from process areas is monitored as it is released through tall stacks, and the most commonly used instrument for this is the Kanne chamber.<sup>9</sup> The Kanne chamber was developed in the early 1940's for monitoring radioactive gases in the stack effluent from the first experimental graphite pile at Oak Ridge National Laboratory (then Clinton Laboratories).<sup>10</sup> The instrument has proven to be the most reliable, accurate, and versatile instrument for monitoring tritium in air.<sup>11</sup> Air to be monitored is drawn continuously through an electrostatic precipitator or ion trap into a large ionization chamber. A typical system includes an intake or sampling line, filter, flowmeter, Kanne chamber, air pump, and exhaust line. The current produced by ionization within the chamber is measured by an electrometer.

The Savannah River Plant is a large tritium-handling facility, and at this site, stack sampling is accomplished with either an aluminum chamber with an active volume of 51.5 liters or a stainless steel chamber with an active volume of 18.5 liters.<sup>12</sup> Air flow through each chamber is approximately 3 cfm. The amplifiers are logarithmic, and the range of the monitoring system for tritium in air is  $1 \times 10^{-5}$   $\mu\text{Ci}/\text{cm}^3$  to

5  $\mu\text{Ci}/\text{cm}^3$ . Except in the case of inordinately high tritium concentrations, or when a large fraction of the tritium present is in the oxide form, there is no problem with residual internal contamination. If this does occur, decontamination can usually be accomplished by purging the chamber for several hours with heated, desiccated air, or the chamber can be disassembled for decontamination. The instruments are sensitive to unusual particulate air loading, external gamma radiation, and other radioactive gases. These effects, however, are reduced by filtering the air intake, by locating the chambers in areas of low gamma background, and by using moisture traps between two chambers in series, respectively. Since the stack monitor provides a continuous record of tritium concentrations, simple integration of the data on the recorder chart indicates the total stack release.

At Lawrence Livermore Laboratory, air is drawn continuously through 1-liter chambers at 1.5 cfm, and concentrations from  $10^{-4}$   $\mu\text{Ci}/\text{cm}^3$  to  $10^3$   $\mu\text{Ci}/\text{cm}^3$  can be detected.<sup>13</sup> The ionization chamber output is converted proportionately to pulses which are then fed to a counting system. One scaler, called the accumulator, is used for long-term recording of activity released from a stack. This determines the total tritium released during a given period and eliminates the hand-integration of the logarithmic curves and conversion of ampere-seconds to curies. Another scaler (one for each monitor) also accumulates pulses, but it is reset automatically every 15 min by an external timer after a printer prints the total count from the scaler. If the total count during any 15-min period exceeds preset limits, a high- or low-level alarm is activated. The low-level alarm activates a bell in the building, and the

high-level alarm activates an evacuation alarm and sends a signal to the Emergency Control Center.<sup>14</sup>

## 2.2 Process Stream and Reactor Coolant Monitoring

Anthony<sup>15</sup> has developed ionization chambers for measuring tritium in process streams. These ion chambers are used to monitor continuously the streams and indicate process irregularities immediately. Three types of ion chambers were designed for monitoring tritium gas in process lines and tanks in the range of 0.01 to 100% tritium. These ion chambers have active volumes of 5.5, 20, and 930 cm<sup>3</sup> and are designed to monitor tritium in hydrogen at pressures between 50 and 760 mm Hg. The 930-cm<sup>3</sup> chamber is used for both in-line and in-tank monitoring. The chambers are made of stainless steel and have glass insulators. The accuracy of the chambers and associated instrumentation is claimed to be within  $\pm 10\%$ .

In the LMFBR program, a continuous monitor to detect tritium in the sodium coolant has been developed.<sup>16</sup> The sensor for this monitor consists of a thin nickel membrane which permits the diffusion of tritium into an argon sweep gas containing 1 to 5% hydrogen. This type of instrument is probably similar to what will be required for monitoring the presence of tritium in potassium, which is a promising primary coolant for a fusion reactor.<sup>17,18</sup>

The monitoring of tritium in secondary and ternary reactor coolants is important in order to determine the amount of radioactive material leaving the plant. Many solid scintillation detectors have been developed for monitoring flowing water. Muramatsu et al.<sup>19</sup> were the first to

develop a scintillation probe for continuous monitoring of tritiated water. Moghissi and associates<sup>20</sup> later developed a detector that is cylindrical in shape and contains parallel 3-mm-diam plexiglass rods coated with anthracene. The rods are viewed by opposing photomultiplier tubes, and the response for tritiated water is approximately 10 cpm for each  $\mu\text{Ci/liter}$ .

The detection of tritium in flowing water has also been accomplished using plastic scintillators. One concept which has been used consists of 37 parallel sheets of 0.013-cm scintillator mounted between two disks of Lucite.<sup>21</sup> Two photomultiplier tubes are used to view the cell, and their outputs are counted in coincidence to reduce the background. Osborne<sup>22,23</sup> has fabricated plastic scintillator systems, which are capable of measuring down to  $10^{-3} \mu\text{Ci/cm}^3$  of tritium in water with a response time less than 1 min. One detector is a 4.6-cm-long cylinder with a 2.7-cm diam; it has a scintillation area of  $240 \text{ cm}^2$  and a counting efficiency of 26%. The second detector is a  $5 \times 5 \times 5 \text{ cm}$  cube, with a scintillation area of  $3000 \text{ cm}^2$ . This instrument has a counting efficiency of 12%, and when used as an effluent monitor, the detector is housed inside 10 cm of lead.

Reactor effluents are usually dirty, and filtration down to 5- or 10- $\mu\text{m}$  pore size is necessary. Chlorine, which is normally used to prevent bacterial growth in the water system, interferes with the measurement of tritium through chemiluminescence and is removed by an activated charcoal filter. To reduce interference from other radionuclides, ionic products can be removed by ion exchange, and gaseous nuclides can be purged from the water by sparging with air prior to passing the sample through the detector.



Ting and Little<sup>24</sup> have built a liquid scintillation detector for the continuous monitoring of secondary reactor coolants. They developed a system that mixes a toluene-based liquid scintillator (or dioxane-based scintillator if very rapid mixing is required) with the water sample in a mixing chamber. The mixed solution flows through a light trap, a liquid scintillation counter, and then out as waste. The liquid scintillator flows continuously into a vial. The water sample enters the vial by dripping gravity flow, and it is stirred for thorough mixing twice before exit. The time response is very good, and the total response peak is over in about 4 min. The efficiency for tritium detection is 32%, and the system is capable of detecting  $6 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$ . The device was successfully field-tested at the San Onofre Nuclear Power Plant where it measured tritium levels in the secondary coolant which were about half the  $(\text{MPC})_w$ , with a few-percent precision.<sup>25</sup>

## 2.3 Occupational Protection

Radiation protection for the radiation worker involves monitoring the reactor halls and working areas for airborne tritium (as water vapor and tritium gas), with both real-time and integrating instruments. It also includes instrumentation for monitoring the presence of surface contamination and for personnel monitoring.

### 2.3.1 Real-Time Air Monitoring

#### 2.3.1.1 Ionization Chambers

Monitoring the occupational work areas for airborne tritium is important in protecting the occupational worker from radiation hazards. A Kanne system at the Savannah River Plant permits the sampling of air

from widely separated work areas individually or from many work areas simultaneously.<sup>26</sup> By using electrically- or manually-operated valves, samples can be selected from one or more points in a work area, or the size of the area being monitored by one system can be changed. A selector switch at the Kanne amplifier allows the output signal to be transmitted to a remote meter in the area being sampled so that workers will have visual indication of the tritium concentration or the signal may be sent outside the work area to be used by the Health Physics Inspector. Audible alarms and flashing lights are added to the system to alert personnel when predetermined concentrations of tritium are reached. Recorders are also included when a record of air activity in a work area is needed. Tritium concentrations from  $10^{-5}$   $\mu\text{Ci}/\text{cm}^3$  to 5  $\mu\text{Ci}/\text{cm}^3$  of air can be measured.<sup>27</sup>

A description of instrumentation installed in the reactor halls of a nuclear plant for measuring tritium in air is given by James.<sup>28</sup> The instruments described are air ionization units that measure from  $10^{-5}$  to  $10^{-2}$   $\mu\text{Ci}/\text{cm}^3$ . Twelve remote sampling points are connected to each unit by polythene tubing. The delay time between a release near a sampling point and the first indication on the monitor has been measured as approximately 45 sec for 31 m of tubing. Two standard radioactive gas monitors, which have 5-cm lead shields around them, are used in each unit. The system has proved adequate for all operational requirements, and remote monitoring can be used in the event of evacuation from the reactor buildings.

The first portable tritium monitoring instrument was the Los Alamos Model 101 Sniffer; however, this monitor was sensitive to spurious

ionizations.<sup>29</sup> One of the first tritium monitors that considered this problem was the detector developed by Tracerlab Incorporated.<sup>30</sup> They made a tritium flow monitor in which air is drawn through a filter, removing the particulate matter and some ions. The air then passes into a sweeper (which is essentially a parallel-plate ionization chamber) where the remaining ions are removed from the air. Anthony<sup>31</sup> improved Tracerlab's model with the addition of a gamma-compensation chamber. It accurately detects tritium concentrations in the range  $4 \times 10^{-5}$  to  $1.6 \times 10^{-2}$   $\mu\text{Ci}/\text{cm}^3$ . It has a dc amplifier, two ionization chambers (sample and compensating chambers), a deionizer, and a battery-operated motor and blower. Air is drawn through a sample chamber which produces a current proportional to the gamma field and the tritium in the air sample. The compensating chamber is connected so that its current subtracts from the sample-chamber current. The difference current, proportional to tritium activity, is amplified and displayed on the meter. The total weight of the monitor is 7.3 kg, and the overall dimensions are  $13 \times 25 \times 41$  cm.

Another monitor developed at Los Alamos to replace the Model 101 Sniffer was the Model 200 AC/DC air sampler.<sup>32</sup> The Model 200 AC/DC is essentially the same electronically as the Model 101 but can be operated as either a portable unit (DC) or an in-place monitor (AC). The rechargeable batteries provide a big advantage in lower cost and lighter weight, and the replacement of a selector sensitivity switch with a four-decade logarithmic response circuit eliminates range changing. Only a qualitative detection of radioactive gases in the atmosphere is obtained, and low-level concentrations of less than  $50 \times 10^{-6}$   $\mu\text{Ci}/\text{cm}^3$  of tritium water vapor are not detectable with this air sampler.

At Chalk River National Laboratory, a considerable effort has been undertaken in the development of ionization chambers (along with other instrumentation) for tritium monitoring in the heavy-water reactor industry.<sup>33-36</sup> Cowper and Simpson<sup>33</sup> developed gamma-compensating ionization chambers where the chambers consisted of the halves of a cylinder divided on an axial plane. However, because of the spatial separation of the two chambers, the compensation was dependent on the gamma-field direction. Osborne and Cowper<sup>34</sup> later developed concentric chambers in which a sealed cylindrical ionization chamber is mounted inside a cylindrical sampling chamber in order to compensate for inhomogeneities in gamma fields. The effective volume of the outer chamber is equal to that of the sealed chamber, and only the difference in current from the pair of chambers is measured. With this arrangement, approximately 98% of the gamma contribution to the ionization current in the outer chamber can be cancelled out. Using this compensation technique, an area monitor was developed (AEP 10101) that uses a 40-liter ionization chamber with an air-flow rate of 36 liters/min.<sup>34</sup> For this detector, the most sensitive range is 0-1.5 (MPC)<sub>a</sub>, and the least sensitive range is 0-50,000 (MPC)<sub>a</sub>. Chart recorder and meter outputs are provided, together with an alarm on all ranges. The instrument output reaches 90% of its final reading in response to a large discrete input of HTO in air in 2 min, and recovery to 1% after such a pulse of HTO takes 25 min.

Portable monitors having 1.2-liter or 0.3-liter ionization chambers have also been developed at Chalk River.<sup>34</sup> The most sensitive ranges on the portable monitors are 0-10 (MPC)<sub>a</sub> and 0-45 (MPC)<sub>a</sub>, respectively, and both can be switched over two decades with five ranges. Sampling rates

are high enough for the response times of the monitors to be less than 1 min. An ion trap and filter unit are attached beneath the mounting board, and a battery-operated air pump is attached to the carrying frame.

Commercially, Johnston Laboratories<sup>37</sup> sells both stationary and portable tritium monitoring instruments. The 955B model is their stationary high sensitivity instrument, and it measures  $10^{-5}$   $\mu\text{Ci}/\text{cm}^3$  full scale. The reproducibility of the instrument is  $\pm 2\%$ , and it has an accuracy of 10% of full scale. The instrument has a pump displacement of 10 liters/min, and the filter system removes particles, ions, and smoke. The 1055B model is portable and has a sensitivity of  $5 \times 10^{-5}$   $\mu\text{Ci}/\text{cm}^3$  full scale. This monitor can operate on line voltage, or it can be run 6 hr continuously on an internal rechargeable Ni/Cd battery. Both instruments have gamma compensation up to 5 mR/hr.

#### 2.3.1.2 Proportional Counters

Proportional counters can also be used for the detection of tritium. The use of proportional counters to detect tritium was initiated by Driver,<sup>38</sup> who developed an air monitor that mixes methane with the air before counting; the instrument is capable of detecting a tritium concentration of  $10^{-2}$   $\mu\text{Ci}/\text{cm}^3$  in air. This sensitivity was later increased by Ehret<sup>39</sup> to  $10^{-5}$   $\mu\text{Ci}/\text{cm}^3$  [ $0.02 \text{ (MPC)}_a$ ] using counters with less than 1-liter volume. The above counters, however, are of the flow-through type which may make them impractical for some monitoring situations.

A thin window proportional counter designed for general monitoring has been described by Block and associates.<sup>40-42</sup> They developed a

large-area, thin-window gas-flow proportional counter that can measure  $5 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$  of tritium oxide in the presence of 3 mR/hr of a uniform gamma radiation field. It does not require an external pump, since diffusion of gas in the atmosphere surrounding the counter provides the sample to be counted; tritium penetrates through inherent cracks and capillaries of a polymer window. There is also no exit port since the counting gas is allowed to diffuse through the window at slight positive pressure. The system uses two almost identical counters; one counter has an aluminized Mylar window that is infinitely thick for tritium betas and has a very low HTO permeation rate, while the other has a Formvar ( $\text{C}_5\text{H}_8\text{O}_2$ ) window sufficiently thin to allow adequate detection of tritium by penetration and permeation. Both counters detect background with essentially equal efficiency, but only the thin-windowed counter detects tritium. Therefore, the difference between the outputs is the tritium contribution. The system design prevents response to other radioactive gases. The monitor is  $2 \times 9 \times 22$  cm and has a counting volume of  $200 \text{ cm}^3$ .

#### 2.3.1.3 Scintillation Systems

Plastic scintillators can be used to detect tritium in air. Sannes and Banville<sup>43</sup> have developed a plastic scintillation monitor in which the beta rays from tritium are detected by two photomultiplier tubes mounted on each end of the scintillator detector. A tunnel diode coincidence circuit and a three-channel pulse-height selector are employed to analyze the photomultiplier pulses. Detection efficiencies of 20% have been observed with this system. Many of the other plastic scintillators described previously for measuring tritium in water effluents can

also be used to monitor for tritium in air. In particular, the detector developed by Osborne,<sup>22</sup> that consists of sixty  $5 \times 5$ -cm plates, has a response of 150 cpm per  $10^{-6} \mu\text{Ci}/\text{cm}^3$  [ $750 \text{ cpm}/(\text{MPC})_a$ ].

An inorganic scintillator for detecting tritium in air has been developed at Lawrence Livermore Laboratory.<sup>44</sup> This system uses a single crystal of  $\text{CaF}_2(\text{Eu})$ , which has a diameter of 4.4 cm and a thickness of 0.025 cm. The inorganic scintillator is nonhygroscopic, can be used as a windowless detector, and does not suffer from memory effects commonly observed with organic scintillators. However, the instrument has a minimum sensitivity of  $160 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ .

### 2.3.2 Tritium Collection and Conversion Systems

Since most direct monitoring instruments have difficulty in measuring tritium concentrations close to the MPC level and have interference problems from gaseous radionuclides, more sensitive techniques involving collection and conversion systems have been developed.<sup>45-51</sup> Unfortunately, many of these instruments cannot monitor tritium on a real-time basis. In collection systems, tritium is collected by pumping air through a desiccant or by bubbling the air sample through ethylene glycol, water, or a cold-trap configuration that condenses the sample. The HTO is then absorbed onto the desiccant or into the counting liquid, and the sample is counted by liquid scintillation. Air that is pumped through a desiccant such as Drierite, silica gel or molecular sieve will lose almost quantitatively its water vapor to the desiccant as long as saturation is not approached. The vapor can be retrieved by distillation, or if a lower sensitivity is tolerable, the water vapor can be retrieved by mixing the desiccant with distilled water and letting the solution sit

until displacement has occurred. Oxidation of HT through palladium or other oxidizing beds and trapping of the HTO onto an absorbent such as molecular sieve is frequently used. In integrating systems, therefore, individual analyses of the air sample stream for HTO and HT can be done by sequentially cold-trapping to obtain the oxide fraction and then oxidizing and trapping the HT fraction.

The cooled condensation trap yields high sensitivity since aliquots of the condensed vapor can be obtained without dilution. However, care must be exercised to ensure complete collection. If incomplete but constant collection can be tolerated, the water bubbler may prove to be satisfactory. [Condensation traps measure down to  $10^{-4}$  (MPC)<sub>a</sub>, while bubblers measure down to  $10^{-2}$  (MPC)<sub>a</sub>.] The bubbler does necessitate dilution, but the lower sensitivity may be offset by the greater convenience.

Valentine<sup>48</sup> describes a bubbler system that consists of a 125 cm<sup>3</sup> of distilled water as the collecting medium. A low-volume gas pump and flow meter combination is used to provide a known air flow through the system. The tritiated water from the bubbler is analyzed by liquid scintillation counting. The HTO concentration in the air is calculated from the volume of air pumped through the bubbler, the volume of bubbler water, and the HTO concentration in that water. The bubbler collector was tested under varying conditions, and the collection efficiency for tritiated water vapor was consistently greater than 90% and was less than 0.1% for tritium gas.

Another bubbler tested<sup>49</sup> experienced an average efficiency of 98%. This technique employed a gas wash bottle filled with ordinary distilled



water through which air was bubbled at a constant flow rate. The method proved to be rapid, reliable, and sensitive and was proven under a variety of field conditions. The minimum detectable tritium oxide air concentration that can reliably be measured in a 5-min sampling period is  $1 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$ .

An instrument developed for the continuous measurement of tritium (HTO) in air in the vicinity of a heavy-water reactor is discussed by Gibson and Burt.<sup>45</sup> In this instrument, water is condensed continuously from the air, and the tritium activity is measured in a flow cell filled with anthracene crystals; condensed water is pumped through the cell at the rate of  $10 \text{ cm}^3/\text{hr}$ . The instrument can continuously measure a tritium level of  $0.4 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$  [ $0.08 (\text{MPC})_a$ ]. In addition, it can be operated in a high gamma-ray background, and the instrument does not respond to radioactive gases such as  $^{41}\text{Ar}$ . Contamination of the cell is rapidly removed following a large release of tritium activity by flushing the cell with distilled water. The long-term buildup of activity and a loss of efficiency limits the life of the flow cell to 6 months.

At Chalk River National Laboratory<sup>51</sup> an  $\text{HTO}_v/\text{water}$  exchanger is used with plastic or liquid scintillator as the detector for tritiated water in a single sample-type liquid scintillation counter. The device was designed to measure tritium in the presence of radioactive noble gases, and is used for continuous measurement, with the concentration of tritium in the water leaving the exchanger sufficiently proportional to the concentration in the sampled air. The plastic scintillator consists of an array of 0.13-mm-thick sheets of scintillator. One such plastic scintillator, developed by Osborne, fits in a cell designed to be

interchangeable with the standard liquid scintillation counter vial. Flow rates of 4000 cm<sup>3</sup>/min for air and 4 cm<sup>3</sup>/min for water are used. The response is 50 cpm per (MPC)<sub>a</sub> with a time constant of 4 min and a background of 14 cpm. When a liquid scintillator is used as the detector, the processing system becomes far more complicated, but the sensitivity to tritium and discrimination against noble gases are improved by an order of magnitude. The water flow is 0.02 cm<sup>3</sup>/min with the flow of the liquid scintillator at 0.2 cm<sup>3</sup>/min. The response with liquid scintillation is 200 cpm per (MPC)<sub>a</sub> with a time constant of 2.5 min and a background of 20 cpm. The discrimination factor against Argon-41 is at least 2000. It is worth noting that both systems can be used either as a water or an air monitor.

### 2.3.3 Surface Monitoring

Tritium will be present on the walls of the containment vessel and associated reactor systems due to permeation and adsorption. Since this will be a source of occupational exposure, surface monitoring will need to be performed routinely at CTR's. Surface monitoring for tritium has been performed with the use of GM tubes,<sup>52</sup> ionization counters,<sup>53</sup> proportional counters,<sup>38,42</sup> smears measured in liquid scintillation systems,<sup>13,54</sup> plastic scintillators,<sup>55</sup> and exoelectron dosimeters.<sup>56</sup>

One of the first instruments used for tritium surface monitoring was the Geiger tube developed by Karraker<sup>52</sup> at Savannah River Laboratory. It is a windowless Geiger tube with the counting gas (98% helium, 2% isobutane) flowing through it. The normal detection limit of a single-tube probe is 800 dpm, and the counting efficiency for tritium is about

10%. Experience with this instrument, however, has shown it to be cumbersome and its results nonreproducible.

Further development at Savannah River produced an open-end ionization chamber for tritium detection.<sup>53</sup> The chamber is operated as a dosimeter to integrate the ionization current. The charged chamber is exposed to the contaminated surface and the reduction in the potential difference of the chamber is read with an electrometer. In this way it is possible to measure 12,000 dpm of tritium on a surface of about 50 cm<sup>2</sup>, with a signal-to-noise ratio of 10:1.

Proportional counters, due to their prime advantage of high sensitivity, have also been developed to measure tritium surface contamination. Driver<sup>38</sup> described a surface monitor that uses a 90% argon-10% methane counting mixture. With a counting system adjusted to accept pulses down to 10 mV amplitude, and the voltage on the  $2.5 \times 10^{-3}$ -cm-diam collector wire set just above the breakdown potential, the monitor will count virtually all ionizing particles entering the chamber volume.

Block and associates<sup>42</sup> developed a proportional counter, described previously, which can also be adapted for surface monitoring. With some minor modifications, the detector is compatible with an Eberline PAC-4G alpha survey meter for contamination readout. The propane counting gas flow is set at a flow rate of about 30 cm<sup>3</sup>/min so that a bottle containing  $8.5 \times 10^4$  cm<sup>3</sup> would operate the system for about 46 hr. The counter has a window thickness of approximately 50 µg/cm<sup>2</sup>. With 10% efficiency, the instrument is able to detect approximately  $2 \times 10^{-3}$  µCi per probe area (about 100 cm<sup>2</sup>). This detection system has been found to be useful in tritium contamination surveys at Lawrence Livermore Laboratory.

The most common approach used to check for surface contamination is through the use of smear tests. The standard smear is a 5-cm-diam circle of filter paper-type material. Usually, the smear is counted in a liquid scintillation counter; however, it can also be counted in a windowless flow proportional counter. Porter and Slaback<sup>54</sup> state that VM-1 and VM-100 are the best overall swipe materials for tritium smearing. The smear technique can be made more sensitive and reliable if the paper disk is wet in an organic liquid such as ethylene glycol before wiping.

At many facilities handling tritium, surface surveys are conducted at some scheduled frequency unless a release occurs requiring an immediate survey. If a swipe removes 10% of the surface contamination from 100 cm<sup>2</sup>, and a 2% counting efficiency is assumed, then  $10^{-4}$   $\mu\text{Ci}/\text{cm}^2$  will give rise to about 400 net counts in 10 min in a liquid scintillation counter. This technique is quite adequate at facilities having a surface contamination limit of  $10^3$  to  $10^4$  dpm/100 cm<sup>2</sup>.

Hughes Whitlock offers a tritium surface monitor which is available commercially.<sup>55</sup> A light-tight seal is made with the surface, and a vacuum pump evacuates above the area to be measured. A 0.125-mm plastic scintillator is used in the instrument for beta detection, and a contamination level of  $10^{-4}$   $\mu\text{Ci}/\text{cm}^2$  is measured with less than 2% error in 10 seconds.

Studies of solid-state detector capabilities indicate the feasibility of using thermally stimulated exoelectron emission (TSEE) dosimeters as tritium detectors. The exoelectron dosimeter can measure weakly penetrating radiations, such as tritium beta rays, due to the shallow surface region within which the exoelectrons originate. Exoelectron dosimeters

made of ceramic BeO have been tested at the Oak Ridge National Laboratory.<sup>56</sup> A tritium contaminated surface can be monitored by placing the BeO disk face down in direct contact with the surface. An exposure time of 100 seconds to an activity of  $10^{-3}$   $\mu\text{Ci}/\text{cm}^2$  produces a signal-to-noise ratio of 10:1.

#### 2.3.4 Personnel Monitoring

The standard personnel monitoring device at nuclear facilities continues to be the film dosimeter; however, the paper wrapping around films is too thick to allow penetration by tritium beta particles. The HTO can diffuse through the paper, but the amount of fogging depends on how long the HTO remains in contact with the emulsion before the film is developed; this causes difficulty in interpreting the fogging in terms of dose. A film badge for measuring HTO exposures of about 100 (MPC)<sub>a</sub> - hours or more has been developed,<sup>57</sup> but the dosimeter is  $5 \times 10^4$  times more sensitive for tritium as HTO than as HT. The film fogging due to HTO may also make it impossible to measure any dose from other sources. The film badge for tritium personnel monitoring would probably be useful only if facilities for measuring tritium in urine were not available.

Urinalysis is more sensitive and accurate than a film badge for measuring tritium exposures, and it is more applicable because tritium represents a hazard only if it is taken into the body. Various descriptions of urinalysis techniques have been given.<sup>42,58-63</sup> Presently, almost all of the urinalysis is performed with liquid scintillation,

though techniques involving ionization chambers,<sup>61</sup> GM counters,<sup>60</sup> and proportional counters<sup>42</sup> can also be used.

For tritium analysis in the field, the T449 Radiological Urinalysis Kit<sup>62</sup> or the proportional counter described by Block and associates<sup>42</sup> can be used. The T449 makes measurements down to 10  $\mu\text{Ci/liter}$  in aqueous solutions and makes the determination in 5 min. A disposable cartridge uses the following reaction of water and calcium carbide to form acetylene as the counting gas:



The gas is liberated from the urine at a controlled rate and is analyzed by the flow-through chamber for tritium content. The instrument weighs 8.2 kg and is enclosed by a  $23 \times 36 \times 25$  cm external carrying case. The instrument is adaptable for field operation and is presently installed in the Lawrence Livermore Laboratory emergency response trailer.

Liquid scintillation counting of urine samples is the most common procedure used at tritium-handling facilities. In the past, the routine assay of urine samples involved an elapse of hours or days between submission of a sample and the returned information. Osborne,<sup>58,59</sup> however, developed an automatic analyzer to determine almost instantaneously the amount of tritium in urine. This instrument is currently being used at Chalk River National Laboratory and at the heavy-water power reactor sites in Canada. The automatic tritium-in-urine analyzer can be used in the same manner as a hand-and-foot monitor for periodic checks. The instrument is extremely easy to operate; a person voids into a urinal and a  $0.3 \text{ cm}^3$  sample is collected and mixed with  $5 \text{ cm}^3$  of

liquid scintillator (Nuclear Enterprises, NE220). The mixture is pneumatically transferred into a counting cell located between photomultipliers. The counting rate is displayed on a chart recorder that is calibrated directly in  $\mu\text{Ci/liter}$  of urine and maximum permissible body burdens (MPBB). A slot in the recorder case allows the user to sign the chart by his recorded body burden. The analyzer can measure as little as 1  $\mu\text{Ci/liter}$ , and the total time for the assay is less than 2 min. Performance of the instrument is automatically checked by processing standard and background samples. In addition to urine, any aqueous sample, such as a bubbler sample, may be assayed by this method.

### 3. OFF-SITE TRITIUM INSTRUMENTATION

The off-site requirements for tritium monitoring involve collecting water, air, and biological samples in a reactor's vicinity. Since the concentrations are low and the sampling locations are unattended, integrating systems are required to monitor the air. Proportional samplers, which collect a proportional fraction of the effluent being released, are used for water collection. In all reported cases, the samples are analyzed by liquid scintillation counting.

#### 3.1 Water Sampling

At tritium facilities, water samples are collected continuously by proportional samplers. The liquid samples are then brought back to the laboratory to be analyzed by liquid scintillation counting. Numerous journal articles<sup>64-69</sup> and books<sup>70,71</sup> have been written about liquid scintillation counting systems. Several important facts dealing with liquid scintillation systems and the counting of samples are reported by Moghissi et al.<sup>68</sup> They state that plastic vials are preferable to glass vials and that the solution which optimizes counting efficiency is 6-7 g of PPO (2,5-diphenyloxazole), 1.2-1.5 g of bis-MSB [p-bis-(o-methylstyryl)-benzene], and 120 g of naphthalene per liter of dioxane. In this solution, the optimum water content is about 1 ml water per 4 ml solution. The minimum detectable activity at a confidence level of one sigma of statistical error for 1-min counting time is about  $10^{-6}$   $\mu\text{Ci}/\text{cm}^3$ . Lieberman and Moghissi<sup>69</sup> state that emulsions should be used when more sensitive counting is needed. By using emulsions, a minimum sensitivity of  $0.2 \times 10^{-6}$   $\mu\text{Ci}/\text{cm}^3$  can be achieved.



The major problem associated with liquid scintillation systems is quenching, which has considerable variation among scintillation solutions. The most frequently used calibration method for tritium counting is the internal standard, in which a standard activity is used interchangeably with the unknown samples. The United States Bureau of Standards has tritiated-water and tritiated-toluene standards.<sup>66</sup>

### 3.2 Air Sampling

At Savannah River,<sup>12</sup> tritium is removed from air by the use of a desiccant (silica gel) for routine sampling. Air is drawn through a silica-gel column at a continuous rate of 100 cm<sup>3</sup>/min by an aquarium aerator pump. The columns are replaced weekly, and the water is removed from the silica gel by distillation before analysis. The concentration of tritium oxide in air is estimated from the tritium content of the sampled moisture and the absolute humidity of the atmosphere. The minimum sensitivity of this procedure is  $4 \times 10^{-11}$   $\mu\text{Ci}/\text{cm}^3$ .

At Mound Laboratory,<sup>50</sup> a rugged ultrasensitive tritium field air sampler capable of simultaneously differentiating tritium oxide from the elemental tritium in the air has been developed. In the collection system, ambient air is passed through a bottle containing silica gel which strips the HTO vapor from the air stream. The air then passes through a second bottle containing silica gel coated with palladium black, and the elemental tritium is catalytically converted to tritiated water. To assure the collection of sufficient HTO in this bottle for subsequent tritium analysis, tritium-free supplemental hydrogen is introduced into the dry air stream just prior to entering the catalytic

collection bottle. The recovered fractions are counted by liquid scintillation spectrometry. The detection sensitivity approaches tritium background levels. The theoretical sensitivity of this sampling system is  $1.6 \times 10^{-12}$   $\mu\text{Ci}/\text{cm}^3$  of air at the 95% confidence level for liquid scintillation counting with 20-cpm counter background, a 21% counter efficiency, and a 100-min sample counting time.

Integrating systems using molecular sieve and freeze-out samplers are also used for environmental air sampling.<sup>46,47,72</sup> Ostlund<sup>46</sup> described a system in which tritium gas is extracted from the air by catalytic combustion on palladium at ambient temperature, with the resultant water being absorbed on a molecular sieve carrying the palladium. In freeze-out methods, the sampler condenses water vapor from the surrounding air. However, the "cold finger" method may be unwieldy for field use, because of pump requirements, and a device such as a "cold strip"<sup>47</sup> may be more applicable for a condensation instrument in the field. This apparatus consists of a metallic strip partially immersed in a liquid nitrogen bath; water droplets and carbon dioxide from the atmosphere condense on the upper part of the cold strip. In the laboratory, the ice is melted, and the solution is analyzed by liquid scintillation counting.

### 3.3 Biological Sampling

From the area surrounding a tritium-handling facility, samples of milk, food, crops, soil, vegetation, and animal tissue are analyzed for tritium content. The free water of the samples is separated by freeze-drying and analyzed separately from the organically-bound tritium. The

organically-bound tritium is converted to water by burning the dried tissue in a furnace in an oxygen atmosphere over copper oxide or by using a commercially available sample oxidizer. All samples are measured for tritium content with liquid scintillation counting.

#### 4. ASSESSMENT OF INSTRUMENTATION

##### 4.1 Ion Chamber and Proportional Counter

The ion chamber is used extensively for the measurement of airborne tritium. It offers the benefits of simplicity and economy and requires only an electrically polarized ionization chamber, an electrometer, and a method for passing the sample through the chamber. However, such an instrument can give erroneous results because of cigarette smoke, aerosols, ions, moisture and condensation, absorption and memory effects, clogged filters, incorrect instrument calibration, ambient gamma-ray field changes, radon, and fission product gases.<sup>73</sup> To compensate for cigarette smoke, aerosols, and ions, a micron-pore-size filter is used to remove all the larger particles. Those particles that pass through the filter are removed by an electrostatic precipitator. Generally, both the filter and electrostatic precipitator must be used, since neither is sufficient alone.

The materials used in the air stream and ion chamber must be carefully selected. If improper materials are used, tritium can be absorbed, causing a low reading, and later reemitted, causing a "memory effect." Polythene and Teflon are good materials to use, whereas, gum rubber and polyvinyl chloride tubing are very retentive for HTO. Also, when flushing the chamber, a dry gas is much less effective than a gas at a high relative humidity, because in the latter case surface tritium activity is exchanged.

Calibration of ion chambers is also very important. Waters<sup>73</sup> states that a tritium measuring instrument should be calibrated frequently

with tritium gas and not by gamma rays or internally generated ions. The gamma-ray check indicates that the ion chamber is working, but an air hose may have fallen off or developed leaks so that the air sample could not be drawn through the chamber for measurement. The most accurate calibration is obtained when the instrument operates in a closed loop so that the tritium concentration is constant, and a steady meter reading is obtained independent of air-flow rate and instrumental time constants. Johnston Laboratories<sup>37</sup> sells a tritium calibrator that has an accuracy of  $\pm 10\%$ , reproducibility of  $\pm 2\%$ , and a calibration time of 3 to 5 min.

Around the fusion reactor there will be a flux of neutrons creating gaseous activation products of argon, nitrogen, and oxygen. Unfortunately, tritium ionization monitors will respond to these radionuclides, and relatively low concentrations of the gases will indicate a false high concentration of tritium. For example, 1 (MPC)<sub>a</sub> of argon-41 produces the same ionization in air as about 50 (MPC)<sub>a</sub> of HTO.<sup>36</sup> This is a problem which must be solved since a CTR reactor hall will contain sizable quantities of activation products in the air.

A problem to fusion plants will be the effect that magnetic fields have on ionization chambers. Strong magnetic fields are used to confine the high-temperature plasmas in many fusion reactor concepts, such as the tokamak design. For the 5000 MW(t) UWMAK-1 concept,<sup>74</sup> a toroidal field in the order of 10,000 gauss will be present in the area near the reactor, and strong poloidal fields as high as 70 to 450 gauss may be present outside the reactor area in the transport and hot cell regions. (For comparison, the earth's magnetic field is about 1/2 gauss.) The

effects of the strong magnetic fields on ionization collection efficiencies and on readout meters require documentation.

Tritium inventories for present CTR designs vary between 2 and 15 kg,<sup>4</sup> with much of the tritium in storage; appropriate instrumentation will be needed to monitor for leakage from these facilities. At Savannah River, an ion chamber has been developed<sup>75</sup> for use as a leak detector to ensure the integrity of storage bottles containing concentrated tritium gas. It can detect tritium leaking at a rate of  $10^{-13}$  cm<sup>3</sup>/sec over a gas-collecting period of 16 hr; this corresponds to a total tritium leakage of 0.015  $\mu$ Ci. The instrument is simple, inexpensive, and easy to maintain. This detector, along with other instrumentation discussed in the review, should be adequate to check for leakage.

Proportional counters are also used to monitor for tritium; they have an advantage over ion chambers in that energy discrimination is possible by electronic discrimination on the output pulse height, whose size is proportional to the energy deposited. The primary disadvantage of the gas proportional counter is the continual consumption of counting gas, which is discarded after being used in the assay of tritiated samples.

The thin window proportional counter,<sup>42</sup> which utilizes a polymer film, has an inherent advantage. For many polymer films, the permeation rate of HTO is greater than that for HT by one to two orders of magnitude. This is of advantage for a tritium monitor in that the  $(MPC)_a$  is two orders of magnitude greater for HT than for HTO, and hence the instrument response may be approximately related to dose.

## 4.2 Scintillation Systems

The inorganic scintillator system developed by Prevo and Santomassimo<sup>44</sup> for detecting tritium in air appears to offer the advantage of simplicity in that only a single photomultiplier tube, an amplifier, and a single channel analyzer is required. The sensitivity of this system, however, is lower than other scintillation systems. The minimum detectable activity is  $1.60 \times 10^{-4} \mu\text{Ci}/\text{cm}^3$  [ $32 \text{ (MPC)}_a$  for HTO], and hence this detector, used alone, is not adequate for monitoring work areas.

Solid organic scintillators<sup>20,22,43</sup> (anthracene and plastic) have also been used for air monitoring, but with limited success. One problem is that when these systems are used for monitoring tritiated water vapor, adsorption of water on the scintillator surface increases the counting rate above that from tritium gas at the same air concentration. The counting rate for a given concentration of tritium in air will also vary with the relative humidity of the air, with the highest count rate occurring at very low humidities. In addition, anthracene will oxidize over time, resulting in a loss of sensitivity.

Anthracene cells are satisfactory in many laboratory applications, but in industrial applications, such as liquid effluent monitoring, they have not been very successful, since they are easily fouled by retention of material suspended in the flowing sample. In addition, the anthracene is subject to relocation and erosion. Plastic scintillators are better in this respect, but plastic surfaces tend to deteriorate over a period of time because of sedimentation, bacterial growth, and discoloration. Discoloration of the scintillator surface or accumulation of slimes on the scintillation surface leads to loss in sensitivity and retention of

activity. The latter point has proved to be a practical problem in monitoring effluents from fission reactors, and a periodic flushing of the detector with a chelating agent is required.<sup>23</sup> Plastic scintillators may, therefore, not be acceptable for liquid effluent monitoring, although when specific activity is high and the water sample is pure, this method can be used satisfactorily.

For solid scintillators, the conversion of tritium gas to a liquid prior to counting has been more successful than counting the gas directly. Tritium as tritiated water vapor can be measured by continuously condensing the sampled vapor as described by Gibson and Burt.<sup>45</sup> An inherent disadvantage of the condenser method, however, is that the sensitivity in terms of concentration in air is dependent upon the humidity of the air since the flow cell measures concentration in water. This is of no consequence if the air is maintained at a fixed humidity, but for monitoring in an uncontrolled environment, the effect of varying humidity must be properly accounted for. Another system for continuous monitoring developed by Osborne<sup>51</sup> does not rely on the condensation method. Here an  $\text{HTO}_v/\text{water}$  exchanger is used in which the concentration of tritium in the water leaving the exchanger is sufficiently proportional to the concentration in the sampled air. In addition, the radioactive gases are discriminated against in that, relative to  $\text{HTO}$ , they are only slightly soluble in water.

The use of liquid scintillation allows several orders of magnitude more sensitive detection of tritium, has greater freedom from contamination, and offers better opportunity to discriminate against other radionuclides than solid scintillation techniques. For batch samples,



as in the off-site surveillance, liquid scintillation counting will be used exclusively, and these techniques are well established. The liquid scintillation system is, however, more difficult to adapt to continuous monitoring, because of the problems of metering and mixing the scintillator and sample.

#### 4.3 Integrating Systems

Integrating systems must be used for an accurate determination of low levels of tritium in the air at a fusion plant. These use desiccants, bubblers, or cold traps to collect HTO. The bubbler is not as efficient as silica gel, but preparation for counting is less complicated. Additionally, the use of silica gel (or other desiccant) presents a "memory effect" that must be taken into consideration because complete moisture desorption of silica gel is not easily attainable.

## 5. INSTRUMENTATION FOR A CTR

Tritium will most likely be the only radionuclide released in significant quantities during the normal operation of a CTR. The CTR complex will be quite large (the reactor hall alone may be 50 m in diameter<sup>76</sup>) and will include many components with diverse functions. Tritium will be present in one form or another within most of the subsystems; however, for most systems, neither the loss rate nor the form of tritium to be released is yet known. The rate of conversion from HT to tritiated water and to tritiated organic materials<sup>77</sup> is also unknown for the type of environment into which it will be released. Nonetheless, much tritium monitoring development has taken place at national laboratories, production facilities, and at heavy-water reactor sites, and a significant body of knowledge concerning tritium monitoring already exists.

Gamma-compensating ion chambers will likely be used at fusion power plants as detectors for stack sampling and for monitoring accidental releases where high concentrations of tritium are involved. It is important that the chambers be gamma compensated, since 1 mR/hr gamma field gives a reading of 10 (MPC)<sub>a</sub> for HTO.<sup>36</sup> For fixed locations, such as near the reactor, the gamma-ray flux may vary in intensity, but not in relative spatial distribution. In this case, side-by-side compensating chambers<sup>31,33</sup> are satisfactory, and lead shields can be used. Where portability in monitoring is required within the reactor building, the concentric chambers concept developed by Osborne and Cowper<sup>34</sup> should be used to compensate for inhomogeneities in the gamma fields. In a CTR plant, it will usually be better to keep a monitor fixed and use a long

flexible sniffer hose to investigate remote areas, rather than using a portable monitor.

In order to accurately record tritium levels, it will be necessary to employ instruments that are more sensitive than ion chambers. Tritium collecting systems will be needed at fusion plants to integrate the cumulative tritium release over a convenient period of time. Cooled condensation traps yield very high sensitivity, and the air can be analyzed for both HTO and HT by sequentially cold-trapping to obtain the oxide fraction and then oxidizing and trapping the HT fraction. Weekly composite samples for liquid scintillation counting will give good data of the amount of HTO and HT in the air.

Scintillation methods appear to be the only practical approach for the assay of tritium in liquids at CTR's. Solid and liquid scintillation methods have been used, and both are discussed in the review. Large surface areas are generally needed for solid scintillators, since the range of a tritium beta is so short. Liquid scintillation, in which the scintillator is intimately mixed with the tritiated liquid, possesses an obvious advantage because of the ideal  $4\pi$  geometry. For either scintillation method, however, when monitoring reactor effluents that contain activated corrosion products and other radionuclides, the counting system should be preceded by a filter or trap which removes other contaminants by ion exchange, adsorption, or, in the case of dissolved gases, purging. The system developed by Ting and Little<sup>24</sup> shows the most promise for monitoring tritium in flowing water effluents from fusion reactors. Its main drawback is the cost of the scintillation liquid; the annual cost of scintillator liquid can be in the range

of \$9,000 to \$15,000 (\$3 to \$5 per liter) when used at the rate of 7 ml per minute.

Due to the amount of tritium in a fusion reactor, large quantities of tritium will adhere to and be absorbed in metal surfaces. Fraas<sup>78</sup> has estimated that the niobium blanket structure for a fusion reactor alone will absorb approximately 21 g ( $2.1 \times 10^5$  Ci) of tritium. Surfaces within the reactor hall and other working areas are expected to be major sources of occupational exposure. At present, the most common method for determining surface contamination is by using smears. The smear indicates the amount of removable tritium on the surface and is strictly a qualitative technique. While the smears indicate the potential hazard from removable tritium, precise information cannot be obtained about the amount of tritium occluded in the metal itself. This is an advantage that an exoelectron dosimeter has over the smear technique. This dosimeter, however, cannot be used where temperatures are much above 100°C. The reproducibility and humidity problems that have plagued TSEE dosimeters in the past must be resolved before they can be used practically. The monitor by Hughes Whitlock<sup>55</sup> appears to be suitable for CTR application; however, for correct operation the surface to be monitored should be smooth, impervious, and free from scratches or cracks.

The proportional counter developed by Block and associates<sup>42</sup> could be used to check for surface contamination at a fusion reactor plant. This counter is compensated against gamma radiation and other radioactive gases up to 5 mR/hr, and it indicates the amount of tritium escaping from the surface by the permeation of tritium through a plastic film.

It was noted that HTO has a much greater permeation rate than HT, and, therefore, the counter is more sensitive to the oxide form. In addition, further sensitivity (to HT and HTO equally) was attempted by trying to make the window sufficiently thin to allow the penetration of tritium betas. Memory effects for this type of instrument are not yet documented.

Uptake of tritium by operating personnel can be determined readily by urinalysis. Each worker in the fusion plant will, in a way, function as a dosimeter. The urinalysis, in addition to providing dosimetric information, will also function as a sensitive indicator of changes in the occupational environment at exposure levels well below regulatory limits. The automatic analyzer developed by Osborne<sup>59</sup> should be particularly useful for CTR application.

The sampling techniques for environmental analysis of tritium are well established. For water analysis, the preferred sampling locations are upstream from the reactor outside of its influence, at the reactor off-site water discharge area, at the first drinking water intake downstream from the reactor, and at the largest drinking water intake point downstream.<sup>79</sup> Selected wells and rainwater should also be collected continuously at locations surrounding the plant. For off-site air monitoring, the silica-gel column used by Mound Laboratory<sup>50</sup> would be appropriate for CTR application. All samples will be counted by liquid scintillation systems using standard techniques.

The measurement of tritium levels in reactor systems, however, will require substantial instrumentation development. For example, sensitive tritium monitors exist for monitoring gas streams, such as helium coolant streams, but their use at the high pressures and in the presence of

large radiation levels has not been demonstrated. Additional research is also needed to accurately determine tritium concentrations in the blanket where very low tritium pressures may exist and in primary coolant systems. Prototype tritium monitors, which depend upon tritium diffusion across a metal membrane, have been developed for use in molten sodium.<sup>16</sup> Liquid lithium has a very great affinity for tritium, and diffusion through a membrane is inadequate at the expected tritium concentration.<sup>80</sup> Additional development work is required to allow the detection of tritium at the 1-100 ppm level in liquid lithium at 500°C. As engineering designs for future fusion devices evolve, tritium monitoring and control instrumentation should be reevaluated for applicability.

## CONCLUSIONS

Tritium monitoring instrumentation and techniques have been developed over the past 30 years. Adequate protection from the harmful effects of tritium associated with CTR's will be provided for the occupational worker and the general public by the proper use of these monitoring systems, along with the prudent application of health physics procedures and engineering techniques. This is not to say, however, that no tritium will be released or that the amount that will be released into the occupational areas or the external environment is known with any certainty at present.

The health physics instrumentation needs can be broken down into monitoring requirements for environmental protection (air, water, and biological samples) and for occupational protection (air, liquid, surface, and personnel monitoring). The environmental monitoring will consist of an integrating device such as a silica-gel column for air monitoring and a proportional sampler for water monitoring. Various biological samples will be collected and separated into organically-bound tritium and free tritium before measurement in a liquid scintillation system. Major additional efforts in instrumentation development are not considered to be required in the area of environmental monitoring for tritium.

In the area of occupational safety, ionization chambers permit measurements down to  $(MPC)_a$  levels of tritium, and integrating systems are able to measure down to  $10^{-4} (MPC)_a$ . Scintillation methods appear the most suitable for monitoring aqueous coolants and effluents at a

CTR, and detection down to  $10^{-6}$  (MPC)<sub>a</sub> is available with liquid scintillation systems. Surface monitoring can be performed either by taking smears or using a modified proportional counter, and established urinalysis techniques will be useful at a fusion power plant. Thus, most of the instrumentation needed for a CTR plant has been designed and fabricated. However, developments directed towards improving electronics and ease of utilization should be continued. In particular, some improvements in portable air monitors would provide a greater freedom in conducting health physics surveys. A real-time portable instrument that can detect (MPC)<sub>a</sub> levels of tritium in the presence of high levels of induced air activity, and a reliable portable instrument for the determination of air and surface contamination which can indicate on a real-time basis whether the tritium is in the elemental or oxidized form are among needed developments. There are also two general areas which require some research: The first is a need to improve the capability of detectors to recover quickly after exposure to high tritium levels and to reduce the longer term "memory effects"; the second is to ascertain whether the presence of magnetic fields will cause adverse operation of instruments to be used within the reactor building.



## REFERENCES

1. R. F. Post and F. L. Ribe, "Fusion Reactors as Future Energy Sources," *Science* 186, 397-407 (1974).
2. USAEC, *Fusion Power: An Assessment of Ultimate Potential*, WASH-1239 (1973).
3. V. A. Maroni, "Selected Problems of Tritium Management Related to CTR," in *A Short Course in Fusion Power*, NP-20040 (1972).
4. USAEC, *CTR Engineering Systems Study Review*, WASH-1278 (1973).
5. D. G. Jacobs, "Survey Paper on Tritium and Nuclear Power Technology," *Am. Nucl. Soc. Trans.* 14, 160 (1971).
6. W. R. Bush, *Assessing and Controlling the Hazard from Tritiated Water*, AECL-4150 (1972).
7. H. L. Butler and J. H. Leroy, "Observation of Biological Half-Life of Tritium," *Health Phys.* 11, 238-85 (1965).
8. ICRP, "Report of ICRP Committee II on Permissible Dose for Internal Radiation (1959)," *Health Phys.* 3, 1-380 (1960).
9. H. L. Kelley and C. R. Phillips, "Review of Tritium Monitoring Devices," in *Tritium*, ed. by A. A. Moghissi and M. W. Carter, CONF-710809 (1973).
10. W. L. Marter and C. M. Patterson, "Monitoring of Tritium in Gases, Liquids, and in the Environment," *Am. Nucl. Soc. Trans.* 14, 162-63 (1971).
11. J. E. Hoy, "Operational Experience with Kanne Ionization Chambers," *Health Phys.* 6, 203-10 (1961).
12. W. C. Reinig and E. L. Albenesius, "Control of Tritium Health Hazards at the Savannah River Plant," *Amer. Ind. Hyg. Assoc. J.* 24, 276-83 (1963).
13. W. J. Silver and M. H. Chew, *Health Physics Aspects of Tritium Control at the Lawrence Radiation Laboratory*, UCRL-71327 (1969).
14. W. J. Silver, E. G. Shapiro, N. A. Greenhouse, and K. M. Scown, *A Tritium Stack Monitor with a Wide Digital Range*, UCRL-73295 (1972).
15. J. D. Anthony, *Ionization Chambers for Measuring Tritium in Process Streams*, DP-747 (1962).

16. R. Kumar, A. F. Panek, D. J. Raye, and P. A. Nelson, *Continuous Monitors for Tritium in Sodium Coolant and Cover Gas of an LMFBF*, ANL-8079 (1974).
17. A. P. Fraas, *Comparative Study of the More Promising Combinations of Blanket Materials, Power Conversion Systems, and Tritium Recovery and Containment Systems for Fusion Reactors*, ORNL/TM-4999 (1975).
18. J. E. Draley and S. Greenberg, "Some Features of the Environmental Impact of a Fusion Reactor Power Plant," in *Technology of Controlled Thermonuclear Fusion Experiments and the Engineering Aspects of Fusion Reactors*, ed. by E. L. Draper, CONF-721111 (1974).
19. M. Maramatsu, A. Koyano, and N. Tokunaga, "A Scintillation Probe for Continuous Monitoring of Tritiated Water," *Nucl. Instr. Methods* 54, 325-26 (1967).
20. A. A. Moghissi, H. L. Kelley, C. R. Phillips, and J. E. Regnier, "A Tritium Monitor Based on Scintillation," *Nucl. Instr. Methods* 68, 159 (1969).
21. C. T. Prevo and V. C. Santomassimo, *The Detection of Tritiated Water Using a Plastic-Scintillator Flow Cell*, UCRL-50007-69-1 (1969).
22. R. V. Osborne, "Detector for Tritium in Water," *Nucl. Instr. Methods* 77, 170-72 (1970).
23. R. V. Osborne, *Monitoring Reactor Effluents for Tritium: Problems and Possibilities*, AECL-4054 (1971).
24. P. Ting and R. L. Little, "Continuous Monitoring of Aqueous Tritium Activity," in *Tritium*, ed. by A. A. Moghissi and M. W. Carter, CONF-710809 (1973).
25. P. Ting and M. K. Sullivan, *Reactor Coolant Monitoring with a Discrete Sampling Flow Cell-Liquid Scintillation System*, Report No. 558, Beckman Instruments, Fullerton, Calif. (1971).
26. E. C. Morris, *Remote Tritium Air Monitoring System*, DPSPU 67-30-1 (1967).
27. R. C. Cooley, *Controlling Tritium Hazards Around Heavy Water Moderated Reactors*, DPSPU 66-30-13 (1968).
28. B. T. James, *The Installed Tritium Monitoring System in the Dido and Pluto Reactor Halls*, AERE-M-1929 (1967).
29. B. C. Eutsler, G. L. Evans, R. D. Hiebert, R. N. Mitchell, C. Robbins, and R. J. Watts, *Instruments for the Monitoring of Tritium in the Atmosphere*, LA-1909 (1955).

30. J. Brinkerhoff, C. A. Ziegler, R. Bersin, and D. J. Chleck, "Continuous Air Monitor for  $H^3$ ," *Nucleonics* 17, 76-81 (1959).
31. J. D. Anthony, "Portable Tritium Monitor Has Gamma Compensation," *Nucleonics* 17, 110-14 (1959).
32. M. J. Engelke, *Model 200 AD/DC Air Sampler*, LA-3228-MS (1965).
33. G. Cowper, and S. D. Simpson, *A Monitor for Airborne Tritium*, AECL-1049 (1960).
34. R. V. Osborne and G. Cowper, *The Detection of Tritium in Air with Ionization Chambers*, AECL-2604 (1966).
35. R. V. Osborne, *Studies and Techniques in Tritium Health Physics at CRNL*, AECL-2699 (1967).
36. W. R. Bush, *Assessing and Controlling the Hazard from Tritiated Water*, AECL-4150 (1972).
37. Johnston Laboratories, Inc., 3 Industry Lane, Cockeysville, Maryland.
38. G. E. Driver, "Tritium Survey Instruments, *Rev. Sci. Instr.* 27, 300-03 (1956).
39. R. Ehret, "Proportional Flow Counters for Measurement of Tritium in Air," in *Assessment of Airborne Radioactivity*, STI/PUB/159 (1967).
40. S. Block, D. J. Hodgekins, and R. D. Taylor, "A Portable  $^3H$  Monitor Using a Large Area, Thin Window Proportional Counter," in *Hazards Control Progress Report No. 31*, UCRL-50007-68-2 (1968).
41. S. Block, O. M. Barlow, J. E. Dixon, and D. J. Hodgekins, "A Tritium Proportional Counter with Gamma Compensation," in *Hazards Control Progress Report No. 33*, UCRL-50007-69-1 (1969).
42. S. Block, D. Hodgekins, and O. Barlow, *Recent Techniques in Tritium Monitoring by Proportional Counters*, UCRL-51131 (1971).
43. F. Sannes and B. Banville, *A Portable Tritium-in-Air Monitor*, AECL-2283 (1965).
44. C. T. Prevo and V. C. Santomassimo, *Tritium Detection Using a  $CaF_2(Eu)$  Scintillator*, UCRL-50007-68-2 (1968).
45. J. A. B. Gibson and A. K. Burt, "A Method for Continuous Measurement of Tritiated Water in Air," *J. Nucl. Energy, Parts A/B*, 185-90 (1966).
46. H. G. Ostlund, "A Sampling System for Atmospheric HT and HTO," *IEEE Trans. Nucl. Sci.* 21, 510-12 (1974).

47. T. S. Iyengar, S. H. Sadarangani, S. Somasundaram, and P. K. Vaze, "A Cold Strip Apparatus for Sampling Tritium in Air," *Health Phys.* 11, 313-14 (1965).
48. A. M. Valentine, *An Investigation of a Bubbler Tritium System*, LA-3916 (1968).
49. D. McConnon, *The Use of Water as a Sampling Medium for Tritium Oxide*, BNWL-CC-547 (1970).
50. R. Brown, H. E. Meyer, B. Robinson, and W. E. Sheehan, *A Ruggedized Ultrasensitive Field Air Sampler for Differentially Determining Tritium Oxide and Gas in Ambient Air Atmosphere*, MLM-2015 (1973).
51. R. V. Osborne, *Development of a Monitor for Tritiated Water Vapour in the Presence of Noble Gases*, AECL-4303 (1972).
52. D. G. Karraker, *A Monitor for Surface Adsorbed Tritium*, DP-34 (1953).
53. D. W. Colvin, *A Simple Monitor for Tritium Contamination on Surfaces*, DP-242 (1957).
54. S. W. Porter and L. A. Slaback, *Measurement of Tritium Surface Contamination*, AFRRI SP66-13 (1966).
55. Hughes Whitlock, Ltd., Worcester, WR135ND, England.
56. R. B. Gammage and J. S. Cheka, "Exoelectron Emitting Dosimeters as Tritium Detectors," paper presented at 19th Annual Health Physics Society Meeting, Houston, Tex., July 8-11, 1974.
57. E. L. Geiger, "Tritium Film Badge," *Health Phys.* 14, 51-55 (1968).
58. R. V. Osborne, "Performance of an Automatic Analyser for Tritium in Urine," *Health Phys.* 18, 87-89 (1970).
59. R. V. Osborne, *Monitoring for Tritium at Low Levels*, AECL-2700 (1967).
60. J. E. S. Bradley and D. J. Bush, "A Simple Method for the Assay of Tritium in Water Samples," *Intern. J. Appl. Radiation Isotopes* 1, 233-34 (1956).
61. B. Eutsler, M. Robbins, and R. Hiebert, *A Portable Apparatus for the Determination of Tritium in Body Fluids and Aqueous Solutions*, LA-1894 (1955).
62. Sandia Laboratories, *A New Family of Tritium Monitoring Equipment*, SC-M-68-245 (1968).

63. A. A. Moghissi, R. Lieberman, M. W. Carter, and J. E. Regnier, "Improved Radiobioassay of Urine for Tritium," *Health Phys.* 17, 727-29 (1969).
64. C. P. Haigh, "Liquid Scintillation Counting for Tritium and Carbon-14," *Nucl. Power* 3, 585-87 (1958).
65. E. Jones, J. R. Mallard, and C. J. Peachey, "A Tritium Counter Designed for Routine Laboratory Use," *Phys. Med. Biol.* 4, 253-63 (1960).
66. S. B. Garfinkel, W. B. Mann, R. W. Medlock, and O. Yura, "The Calibration of the National Bureau of Standards' Tritiated-Toluene Standard of Radioactivity," *Intern. J. Appl. Radiation Isotopes* 16, 27-33 (1965).
67. A. A. Moghissi and M. W. Carter, "Internal Standard with Identical System Properties for Determination of Liquid Scintillation Counting Efficiency," *Anal. Chem.* 40, 812-14 (1968).
68. A. A. Moghissi, H. L. Kelley, J. E. Regnier, and M. W. Carter, "Low-Level Counting by Liquid Scintillation - I. Tritium Measurement in Homogeneous Systems," *Intern. J. Appl. Radiation Isotopes* 20, 145-56 (1969).
69. R. Lieberman and A. A. Moghissi, "Low Level Counting by Liquid Scintillation II. Applications of Emulsions in Tritium Counting," *Intern. J. Appl. Radiation Isotopes* 21, 319-27 (1970).
70. D. K. Horrocks and C. T. Peng (eds.), *Organic Scintillators and Liquid Scintillation Counting*, Academic Press, New York, 1971.
71. E. D. Bransome (ed.), *The Current Status of Liquid Scintillation Counting*, Grune and Stratton, New York, 1970.
72. J. W. Mullins, V. E. Andrews, and L. M. Dunn, "Techniques for Monitoring and Analysis of Environmental Tritium," *IEEE Trans. Nucl. Sci.* 19, 177-80 (1972).
73. J. R. Waters, "Precautions in the Measurement of Tritium Concentrations in Air when Using Flow-Through Ion Chambers," *Nucl. Instr. Methode* 117, 39-43 (1974).
74. B. Badger et al., *Wisconsin Tokamak Reactor Design*, UWFD-68 (1973).
75. D. W. Colvin, *A Simple Leak Detector for Tritium*, DP-198 (1957).
76. J. R. Young, B. F. Gore, D. D. Mahlum, J. A. Strand, R. C. Thompson, and J. K. Soldat, *Information for Requirements for Controlled Thermonuclear Reactor Environmental Impact Statements*, BNWL-1883 (1975).

77. R. G. Hickman, "Tritium Problems in Fusion Reactor Systems," in *Proc. of the Symp. on Tritium Technology Related to Fusion Reactor Systems*, ERDA-50, ed. by W. H. Smith, W. R. Wilkes, and L. J. Wittenberg (1975).
78. A. P. Fraas, *Conceptual Design of the Blanket and Shield Region and Related Systems for a Full-Scale Toroidal Fusion Reactor*, ORNL/TM-3069 (1973).
79. S. W. Porter, "The Monitoring of Tritium in the Aquatic Environment of Power Reactors," in *Tritium*, ed. by A. A. Moghissi and M. W. Carter, CONF-710809 (1973).
80. W. R. Wilkes, *Tritium Interactions of Potential Importance to Fusion Reactor Systems - Technology Requirements*, MLM-2292 (1976).

THIS PAGE  
WAS INTENTIONALLY  
LEFT BLANK

## INTERNAL DISTRIBUTION

- |                           |                                      |
|---------------------------|--------------------------------------|
| 1. S. I. Auerbach         | 31. J. E. Phillips                   |
| 2. J. A. Auxier           | 32. H. Postma                        |
| 3. J. T. Bell             | 33. J. W. Poston                     |
| 4. R. S. Booth            | 34. C. R. Richmond                   |
| 5. H. M. Butler           | 35. P. S. Rohwer                     |
| 6. J. F. Clarke           | 36. M. W. Rosenthal                  |
| 7. R. B. Cumming          | 37. T. H. Row                        |
| 8. R. C. Dahlman          | 38-52. K. E. Shank                   |
| 9-18. C. E. Easterly      | 53. R. L. Shoup                      |
| 19. J. W. Elwood          | 54. D. Steiner                       |
| 20. A. P. Fraas           | 55. F. J. Smith                      |
| 21. W. Fulkerson          | 56. J. B. Storer                     |
| 22. R. B. Gammage         | 57. E. G. Struxness                  |
| 23. W. R. Grimes          | 58. J. S. Watson                     |
| 24. F. F. Haywood         | 59. C. E. Wheeler                    |
| 25. J. A. Horak           | 60-62. Central Research Library      |
| 26. D. G. Jacobs          | 63. ORNL-Y-12 Library                |
| 27. G. D. Kerr            | 64-76. Laboratory Records Department |
| 28. T. W. Oakes           | 77. Laboratory Records, ORNL R.C.    |
| 29. P. Otaduy             | 78. ORNL Patent Office               |
| 30. P. Papagiannakopoulos |                                      |

## EXTERNAL DISTRIBUTION

79. J. Baublitz, Division of Magnetic Fusion Energy, ERDA, Washington, DC 20545
80. R. P. Blaunstein, Division of Biomedical and Environmental Research, ERDA, Washington, DC 20545
81. R. L. Butenhoff, Division of Biomedical and Environmental Research, ERDA, Washington, DC 20545
82. F. E. Coffman, Division of Magnetic Fusion Energy, ERDA, Washington, DC 20545
83. R. D. Cooper, Division of Biomedical and Environmental Research, ERDA, Washington, DC 20545
84. G. Cowper, Biology and Health Physics Division, Chalk River Nuclear Laboratories, Chalk River, Ontario
85. J. Davis, McDonnell Douglas Corporation, Box 516, St. Louis, Missouri 63166
86. F. Flakus, International Atomic Energy Agency, Kartner Ring 11, P.O. Box 590, A-1011 Vienna, Austria
87. P. Hancox, United Kingdom Atomic Energy Agency, Culham Laboratory, Abingdon, Berkshire, England
88. J. Hickey, Environmental Standards Branch, U.S. Nuclear Regulatory Commission, Washington DC 20555
89. A. B. Johnson, Battelle Pacific Northwest Laboratories, Richland, Washington 99352



90. C. J. Kershner, Mound Laboratory, Miamisburg, Ohio 45342
91. J. L. Kulcinski, Department of Nuclear Engineering, University of Wisconsin, Madison, Wisconsin 53706
92. P. H. Lamberger, Mound Laboratory, Miamisburg, Ohio 45342
93. D. S. Myers, Lawrence Livermore Laboratory, Livermore, California 94550
94. R. V. Osborne, Biology and Health Physics Division, Chalk River Nuclear Laboratories, Chalk River, Ontario
95. W. D. Patterson, Lawrence Livermore Laboratory, Livermore, California 94550
96. T. B. Rhinehammer, Mound Laboratory, Miamisburg, Ohio 45342
97. J. K. Soldat, Battelle Pacific Northwest Laboratories, Richland, Washington 99352
98. R. Wilson, Ontario Hydro, 620 University Ave., Toronto, Ontario
99. R. W. Wood, Division of Biomedical and Environmental Research, ERDA, Washington, DC 20545
100. J. R. Young, Battelle Pacific Northwest Laboratories, Richland, Washington 99352
101. Director, Research and Technical Support Division, ERDA-ORO
- 102-293. Given distribution as shown in TID-4500 under UC-20, Controlled Thermonuclear Processes and Plasma Physics