AN ABSTRACT OF THE THESIS OF

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Title: Installation, Calibration and Independent Verification of the Performance of a Primary Water Radioactivity Monitor in a One Megawatt TRIGA Research Reactor

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Routine health physics measurements at research reactors commonly include monitoring the reactor water for radioactivity. One location where water monitoring is of particular importance is the reactor primary water system. The importance of monitoring this system is attached to the fact that primary water radioactivity concentrations are highly correlated with the potential for spreading radioactive contamination throughout the reactor facility and to the potential for occupational radionuclide uptake. In addition, however, such measurements also provide evidence of fuel cladding integrity and can be used to indicate the condition of ion exchange resins, filter media and containment of in-core experiments.

Installation and calibration procedures for a primary water radioactivity monitor (PWM) at a one megawatt TRIGA research reactor are described. The resulting PWM calibration, which was best fit to a power function expression, showed [gross water radioactivity concentration in $\mu Ci\ ml^{-1}$ (excluding ^{3}H)] = [(1.11×10^{-7})(net $cpm^{1.0222}$)]. This relationship was established by injecting solutions containing known concentrations of ^{56}Mn and ^{24}Na into the counting chamber of the PWM, while the chamber was isolated to prevent water flow. The 847 keV gamma peak from ^{56}Mn and the 1369 keV gamma peak from ^{24}Na cover essentially the entire range of gamma energies from radionuclides present in the primary water system during normal reactor operation. This range is also representative of gamma energies which would be present during abnormal situations, such as during the release of fission products from fuel elements.

The standard reference for the PWM calibration was a lithium-drifted germanium semiconductor [Ge(Li)] detection system, which was used to establish the ^{56}Mn and ^{24}Na concentrations in the calibration solutions. This Ge(Li) system was calibrated using an NBS certified ^{152}Eu standard solution (NBS-Standard Reference Material 4370).

The accuracy of the PWM calibration was independently verified by comparing analytical results from identical reactor water samples which were divided and then counted with the Ge(Li) system and the PWM. The PWM results agreed with the Ge(Li) to within ±9 percent under the isolated static flow conditions (i.e., no water flowing through the monitoring chamber), but were higher than the Ge(Li) by

an average factor of approximately 2.26 when water was allowed to flow through the PWM. It is therefore concluded that unless new evidence can be produced verifying the higher response of the PWM while water is flowing through, operational application of the PWM should be corrected to reflect the actual concentrations measured by the Ge(Li) detection system. This will require a downward adjustment of the $\mu Ci\ ml^{-1}$ concentrations predicted by the previously stated calibration equation by a factor of 2.26, and will result in the following expression for actual PWM use:

$$\begin{array}{c}\text{Gross water}\\ \text{radioactivity concentration}\\ \text{in } \mu Ci\ ml^{-1}\\ \text{(excluding } {}^{3}H\text{)}\end{array} = \left[\frac{(1.11 \times 10^{-7})(\text{net cpm}^{1.0222})}{2.26}\right]$$

INSTALLATION, CALIBRATION AND INDEPENDENT VERIFICATION OF THE PERFORMANCE OF A PRIMARY WATER RADIOACTIVITY MONITOR IN A ONE MEGAWATT TRIGA RESEARCH REACTOR

by

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INTRODUCTION

Normally, research reactors include the measurement of radioactivity in the primary cooling water as one of the routine health physics surveillance programs. Such measurements are particularly important for evaluating an important source of potential radioactive contamination in the reactor facility, and also the potential for occupational radionuclide uptake. Furthermore, such measurements provide evidence of fuel cladding integrity and can be used to indicate the condition of certain water system components such as demineralizers and filters.

Presently, the radioactivity in the primary coolant water at the Oregon State University TRIGA Reactor (OSTR) is measured by an on-line monitoring system supplied as part of the original equipment package by the reactor vendor, and also by taking individual grab samples back to the laboratory for analysis. The latter method is presently much more satisfactory because of its accuracy and sensitivity, but requires individual samples for each analysis. Therefore, an on-line primary water monitor with greater sensitivity could be an operational advantage, if calibrated to provide the type of data reactor operations and radiation protection personnel require.

The primary water radioactivity monitor (PWM) used in this study employed a lead-shielded Geiger-Mueller (GM) tube that was immersed directly into primary water which was allowed to flow inside the shielded liquid counting chamber. The PWM was sensitive to

essentially all of predominant beta-gamma emitting radionuclides present in the primary coolant, with the one major exception being tritium.

The general classes of radionuclides potentially present in the OSTR primary water are in two categories: (1) induced radioactive nuclides produced by neutron activation of structural or experimental materials, corrosion products, dissolved gases (mainly those in dissolved atmospheric air) or other impurities in the primary water; and (2) fission products, released from failed fuel elements or fueled experiments.

An analysis of normal gamma emitting radioactivity in the primary water of the OSTR using a lithium-drifted germanium [Ge(Li)] detector and a multichannel analyzer identified three major gamma-emitting radionuclides. These were ^{24}Na, ^{56}Mn and ^{41}Ar. ^{24}Na is produced from the fast neutron (3.13 MeV threshold) activation of aluminum through the $^{27}Al(n,\alpha)^{24}Na$ reaction; ^{56}Mn from neutron interactions with ^{56}Fe in steel, through the $^{56}Fe(n,p)^{56}Mn$ reaction; and ^{41}Ar from conventional thermal neutron activation of ^{40}Ar dissolved in the water. One major radionuclide that will also be produced in the primary water system during normal operation is nitrogen-16, through the $^{16}O(n,p)^{16}N$ reaction. Because of its 7.4 second half-life and rapid decay to stable oxygen, the ^{16}N will not live long enough to attain a uniform concentration in the reactor water, and decayed before it could be counted on the Ge(Li) system.

In the event of fission product release, those appearing in the primary water would be mainly isotopes of the elements krypton and

xenon, plus bromines and iodines. The bromines and iodines are very soluble in water; however, the noble gases krypton and xenon partially escape from the water and are released into the room air, where they subsequently decay to their respective radioactive particulate daughters rubidium and cesium.

The major radionuclides regularly present in the OSTR's primary water during normal reactor operation have gamma energies between 847 keV and 1369 keV. Also, most of the projected fission product radionuclides that could be released into the primary water due to the failure of fuel element cladding or an in-core experiment have gamma energies within the above range. Therefore, it was considered acceptable to calibrate the response of the PWM for both normal and abnormal operations using ^{56}Mn (847 keV gamma peak) and ^{24}Na (1369 keV gamma peak).

As a result of the preceding, this study was conceived with the following objectives: (1) to identify appropriate performance objectives and characteristics for an on-line primary water monitor for use in a TRIGA research reactor; (2) to apply these considerations in the actual installation of a primary water monitor; (3) to establish a PWM calibration curve and calibration expression suitable for operational use under normal and abnormal operating conditions; (4) to verify independently the accuracy of the established PWM calibration curve and calibration expression based on a calibrated Ge(Li) system; and (5) to evaluate the significance of any variations in response between the PWM and the Ge(Li) system.

MATERIALS AND METHODS

At the Oregon State University TRIGA Reactor, the primary coolant water is recirculated through a closed system divided into two separate parts, (1) the purification system, and (2) the cooling system (Fig. 1). A small fraction of the primary water flow, about 6.31×10^{-4} $m^3 s^{-1}$ (10 gpm), passes through the water purification system, which includes a pump, a cartridge filter, a mixed bed demineralizer, a flowmeter, and a monitoring vessel containing probes for measuring temperature, radioactivity, and conductivity. The other fraction of the primary water flow, about 3.09×10^{-2} $m^3 s^{-1}$ (490 gpm), is pumped through the cooling system, which primarily involves passing the water through a heat exchanger. The reactor tank is also equipped with a diffuser nozzle, which is used to delay ^{16}N rising to the tank surface.

The primary water radioactivity monitor (PWM) used in this study employed a Tracerlab MWP-4 Liquid Monitoring System. The system consisted of the MW-4A shielded counting chamber, MD-12C 250 mm (10-inch) long Geiger-Mueller detector[1], MM-5B linear ratemeter, and a Varian strip chart recorder. The shielded liquid counting chamber and Geiger detector were located in room D104A of the Radiation Center, on the water purification system skid, and were connected through a cable to the ratemeter, power supply and recorder located in the reactor control room (D302).

[1]Amperex Electronic Corporation, Model 912NB3, halogen-quenched, Geiger-Mueller detector with a 178 mm (7 inch) long active counting length and 40 mg cm^{-2} wall thickness.

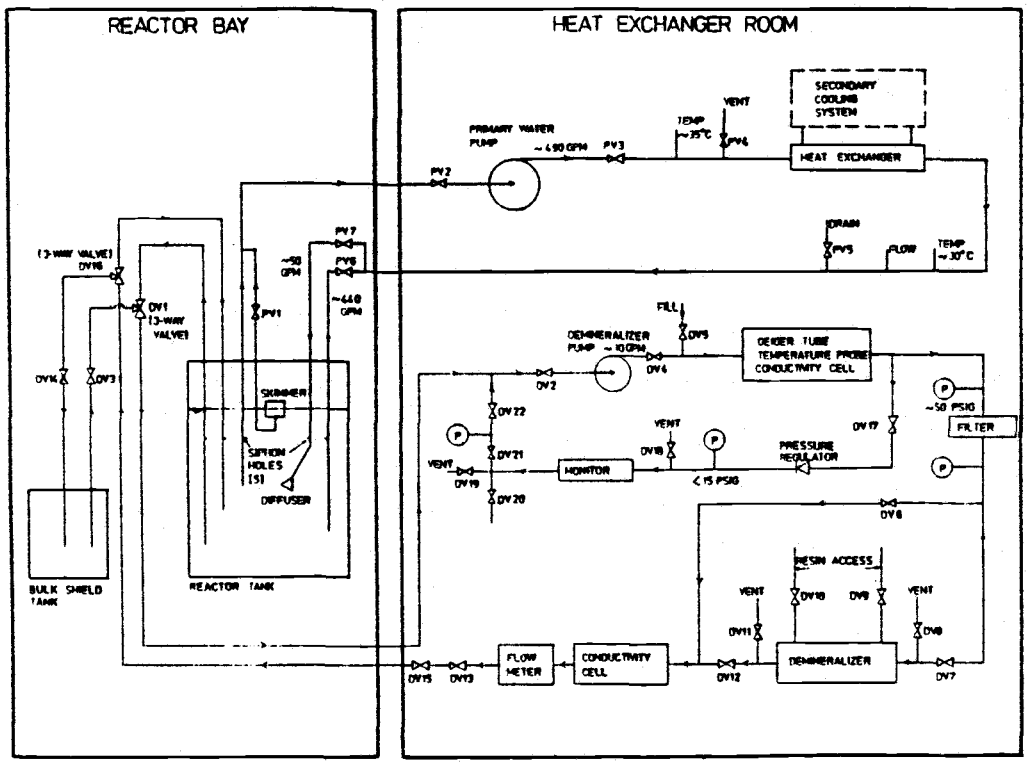

Fig. 1. General schematic of OSTR water cooling and purification system.

The design of the PWM was such that the GM tube was immersed directly into the liquid to be monitored inside the shielded liquid counting chamber. Inlet and outlet pipes from the counting chamber facilitated connection into the primary water system, and radioactivity detected by the Geiger tube was displayed in the reactor control room on the ratemeter and recorder. The ratemeter had four linear scales: 0 - 200, 2,000, 20,000 and 200,000 (200, 2K, 20K and 200K) counts per minute (cpm). The operating voltage for the Geiger detector was set at 900 volts, determined by plotting high voltage versus count rate and selecting a voltage about 50 volts above the lower end of the counting plateau (Fig. 2). The GM tube operating voltage and the 3,600 cpm signal for testing the ratemeter response were both verified every time the PWM was used.

All water radioactivity concentrations analyzed in this study were standardized by or compared to results obtained with a calibrated Ge(Li) based detection system. In this regard, it was assumed that the concentrations measured by the Ge(Li) detection system were correct. The Ge(Li) detection system used throughout the entire study consisted of a nominal 70 cm^3 Princeton Gamma-Tech lithium-drifted germanium semiconductor detector, a Nuclear Data Corporation ND 660 multichannel analyzer which is controlled by an LSI-11 microprocessor, and a Digital Equipment Corporation Decwriter II teletype.

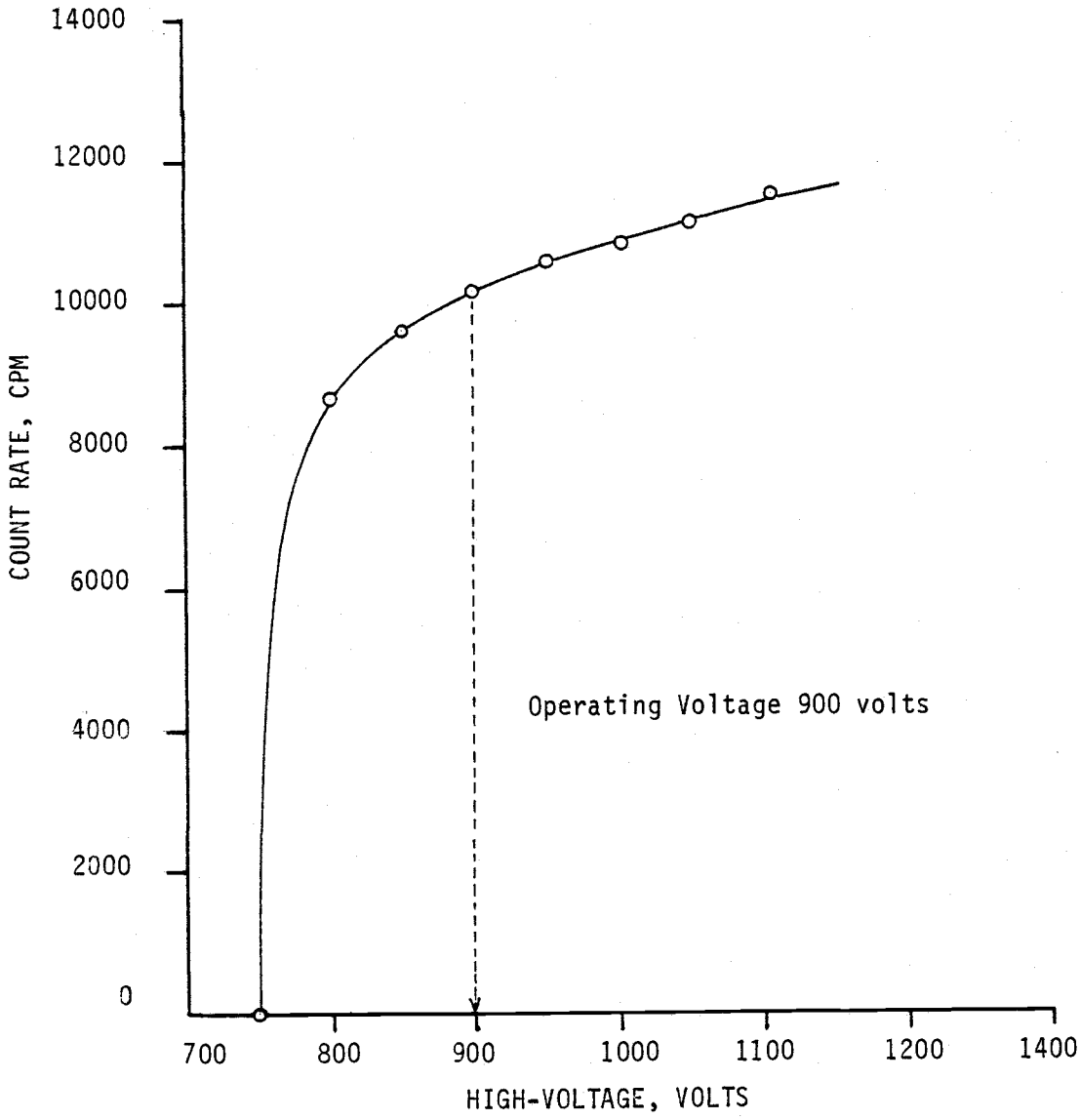

Fig. 2. Determination of Geiger tube operating voltage for the primary water monitor.

A counting efficiency calibration for the Ge(Li) detector and analyzer system was performed using gamma energy peaks between 121.8 keV and 1408.0 keV as shown in Table 1. The gamma energy peaks were obtained from an NBS certified Europium-152 liquid standard (NBS-SRM 4370)[1], which was diluted to 500 ml with distilled water and sealed into a marinelli beaker. A plot of gamma energy versus Ge(Li) system counting efficiency (Fig. 3) based on the ^{152}Eu standard in the marinelli beaker was then used to determine the Ge(Li) system counting efficiency for identically prepared water samples containing ^{56}Mn or ^{24}Na, and reactor primary water containing both of these radionuclides plus ^{41}Ar.

The first step in this study was measurement of the radioactivity in the reactor primary water. This measurement was intended to determine the concentration and identity of key radionuclides (except ^{3}H) that are routinely present in the primary water during normal reactor operation, and to establish which of these are major contributors to the overall radioactivity concentration during operation. This was achieved by taking samples of the primary water, placing 500 ml volumes into marinelli beakers, and counting these with the Ge(Li) detector. Table 2 gives typical radionuclides and their equilibrium concentrations immediately after the reactor had run for about four hours at one megawatt. Further Ge(Li) analysis of such

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[1] Europium-152 Radioactivity Standard from National Bureau of Standards (Standard Reference Material 4370).

Table 1. Ge(Li)detection system counting efficiency for ^{152}Eu standard source (NBS-SRM 4370)

E_γ (keV)	Photon Abundance per Disintegration	Gamma Disintegration Rate[1] (dps)	Observed Gamma Count Rate (cps)	Gamma Counting Efficiency
121.8	0.28370	9000	189.1	0.0210
244.7	0.07510	2383	39.6	0.0166
344.3	0.26580	8433	99.0	0.0117
411.1	0.02234	709	7.0	0.0098
444.0	0.03121	990	9.2	0.0093
778.9	0.12960	4112	22.1	0.0054
964.0	0.14620	4638	22.5	0.0049
1112.1	0.13560	4302	18.6	0.0043
1408.0	0.20850	6615	23.5	0.0036

[1] Gamma disintegration rate as of 0924 PST January 20, 1981.

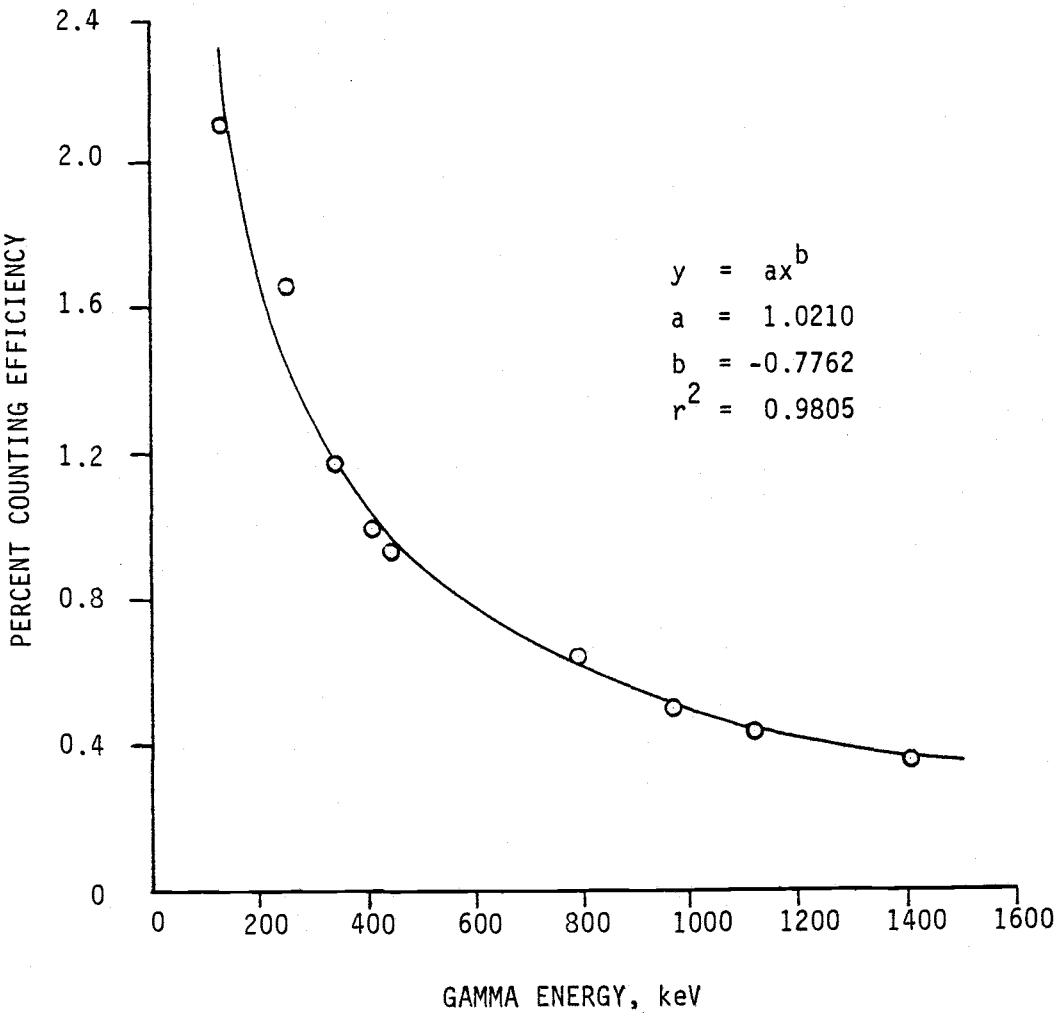

Fig. 3. Ge(Li) detector counting efficiency as a function of gamma energy.

Table 2. Predominant radionuclides[1] and their typical equilibrium concentrations at one megawatt in OSTR primary water.

Radionuclide	Half-life (Hours)	Typical Equilibrium Concentration[2,3] (μCi ml^{-1})
Argon-41	1.83	6.16×10^{-4}
Manganese-56	2.576	1.15×10^{-4}
Sodium-24	15.0	1.40×10^{-4}

[1] Tritium is excluded here because it was not detectable with the instrumentation used in this study. Its normal concentration in the primary water ranges between 3.0×10^{-5} and 3.65×10^{-4} μCi ml^{-1}.

[2] Concentration established after steady state operation for four hours at one megawatt.

[3] These particular concentrations were obtained on a Friday. It was observed that ^{56}Mn typically had a higher concentration by a factor of 1.24 than ^{24}Na when measurements were performed following similar operations on Monday or earlier in the week. This reflects the buildup of the longer lived ^{24}Na relative to ^{56}Mn during the course of the week's operation.

water samples after about 24 hours decay resulted in the identification of additional radionuclides at much lower concentrations, as given in Table 3. As mentioned earlier, ^{56}Mn and ^{24}Na were used for the PWM calibration primarily because their gamma energies span essentially the entire range of gamma energies expected from radionuclides present in the reactor primary water system during normal and abnormal situations. However, they were also considered to be desirable for calibration purposes because their relatively short half-lives created observably different radioactivity concentrations within comparatively short time increments, and thereby provided more data points for a calibration curve.

To accomplish the PWM calibration, the following steps were carried out: (1) ^{56}Mn and ^{24}Na radionuclides were produced using the rabbit facility at the OSTR; (2) the radionuclides were then diluted with distilled water to make concentrations suitable for counting on the 2K, 20K and 200K cpm scales of the PWM; (3) a 500 ml fraction of the diluted water sample was next counted on the Ge(Li) detector to establish its concentration in $\mu Ci\ ml^{-1}$; (4) a 900 ml fraction of the same water sample was placed into the liquid counting chamber of the PWM and the decay was followed on the recorder in terms of cpm; (5) the net sample counting rates determined from the recorder were then correlated to $\mu Ci\ ml^{-1}$ concentrations in the PWM counting chamber by using the $\mu Ci\ ml^{-1}$ data obtained from the counts of the sample with the Ge(Li) system. Corrections for Ge(Li) counting times were taken into consideration when establishing concentrations

Table 3. Radionuclides detectable in OSTR primary water after 24 hours of decay.

Radionuclide	Half-life	Prominent Gamma Energy and Abundance (keV)	Concentration After 24 hours of Decay ($\mu Ci\ ml^{-1}$)
Argon-41	1.83 hours	1293 (99%)	2.20×10^{-8}
Cobalt-58	71.3 days	810 (99%)	1.68×10^{-7}
Cobalt-60	5.26 years	1173 (100%), 1332 (100%)	9.00×10^{-8}
Chromium-51	27.8 days	320 (9%)	1.64×10^{-6}
Manganese-54	303 days	835 (100%)	1.90×10^{-7}
Manganese-56	2.576 hours	847 (99%), 1811 (29%)	1.69×10^{-7}
Sodium-24	15.0 hours	1369 (100%), 2754 (100%)	3.36×10^{-5}
Hydrogen-3 (Tritium)	12.262 years	none	*

* Tritium is routinely present in the TRIGA primary water, but is not detectable by gamma counting.

for the PWM from Ge(Li) data. Details pertaining to the various steps listed for the calibration process are given below.

^{56}Mn and ^{24}Na were produced by irradiating one ml of $Mn(NO_3)_2$ or $Na(NO_3)$ solution, respectively. The irradiations were performed in the rabbit system at the OSTR, for two minutes each, while the reactor power was at one megawatt. After irradiation, the radionuclides were first diluted with distilled water to make a total volume of 1500 ml. A 500 ml fraction of this was then sealed into a marinelli beaker and another fraction (about 900 ml) was placed into the PWM counting chamber.

The solution in the marinelli beaker was immediately counted with the Ge(Li) detector to establish an initial reference $\mu Ci\ ml^{-1}$ concentration for the radionuclide involved. All Ge(Li) counts were designed to collect not less than 10,000 net counts for the photopeaks of interest. This was usually achieved by counting the samples for 600 to 1800 seconds, depending upon the concentration of the radionuclide. All counts were automatically divided by the analyzer's live counting time to arrive at the average net counting rate for the sample. Expressions (1) and (2) were used with the Ge(Li) results to obtain the radioactivity concentration in $\mu Ci ml^{-1}$ for each sample at an earlier reference time t_0, the time at which the PWM began to record activity from the other portion of the same radioactive water sample.

$$[\text{Conc.}]_{t_c} = \frac{\bar{c}}{(\text{Eff.})(3.7 \times 10^4 \frac{\text{dps}}{\mu\text{Ci}})(500\ \text{ml})} \qquad (1)$$

$$[\text{Conc.}]_{t_0} = \frac{([\text{Conc.}]_{t_c})(\lambda t_c)}{(1 - e^{-\lambda t_c})(e^{-\lambda t_1})} \quad (2)$$

Where:

$[\text{Conc.}]_{t_c}$ = average concentration measured during counting time (μCi ml^{-1})

$[\text{Conc.}]t_0$ = concentration at a reference time t_0 (μCi ml^{-1})

$\bar{c}$ = measured count rate (cps)

Eff = Ge(Li) detector counting efficiency for the particular gamma energy peak

t_0 = reference time when PWM counting of a specific sample began (this time was always earlier than the actual Ge(Li) counting of a water sample)

t_c = time of counting (s)

t_1 = elapsed time from the earlier start of PWM sample counting (at t_0) to the beginning of sample counting with the Ge(Li) system (s)

At least five Ge(Li) measurements were made for each water sample. The arithmetic mean of the derived concentrations, corrected for times t_c and t_1, was then used to represent the concentrations at time t_0. Concentrations at any time t later than t_0 were calculated by using the standard decay expression

$$[\text{Conc.}]_t = [\text{Conc.}]_{t_0} e^{-\lambda t} \quad (3)$$

Where:

$[\text{Conc.}]_t$ = concentration at any time t after t_0 (μCi ml^{-1})

$[\text{Conc.}]_{t_0}$ = concentration at reference time t_0 when PWM sample counting started (μCi ml^{-1})

λ = radionuclide decay constant (s^{-1})

t = decay time from reference time t_0 to any time t later (s)

The response of the PWM was displayed in the reactor control room on two indicators, a ratemeter and a chart recorder. For this particular study, the chart recorder was used to follow the sample counting rate as the radionuclide in the PWM counting chamber decayed with time. Calibration runs for each of the two radionuclides, ^{56}Mn and ^{24}Na, were carried out over the three ratemeter scales deemed to be of value for reactor operations, 0 - 2K, 20K, and 200K counts per minute. The 0 - 200 cpm scale was considered to be too low for operational use, but was checked with small instrument counting sources to establish a consistency of response with the higher ranges.

Two calibration runs were carried out for each radionuclide. While the results from the two separate runs were very close, the net counting rates (cpm) and corresponding concentrations ($\mu Ci\ ml^{-1}$) for each of the two radionuclides were obtained by averaging the data from the two runs.

To obtain the net counting rate due to radioactivity in the PWM counting chamber it was necessary to consider correcting for three sources of radiation background. These were the natural background, background due to radiation shine from the reactor (particularly at higher power levels), and the radiation background from a nearby filter cartridge and demineralizer tank. The count rate contribution from these three sources was measured by placing nonradioactive water (city water) into the PWM counting chamber. The PWM system was then allowed to record the total radiation background at three different reactor power levels, 250 kW, 500 kW and 1000 kW. Background data

were collected for a minimum of four hours at each power level. Subsequent net counting rate values were then obtained by using the total background contribution at a particular power level. The basic net cpm expression is shown below:

$$\text{PWM net cpm} = \text{PWM gross cpm} - \left\{ \begin{matrix} \text{natural} \\ \text{background} \\ \text{cpm} \end{matrix} + \begin{matrix} \text{shine} \\ \text{cpm} \end{matrix} + \begin{matrix} \text{filter and} \\ \text{demineralizer} \\ \text{cpm} \end{matrix} \right\} \quad (4)$$

Two different PWM operating modes were evaluated during the independent verification of accuracy for the PWM calibration. The two modes involved a static flow (no water flowing through the counting chamber) and a dynamic flow (water moving through the counting chamber) condition. The static flow tests were conducted by taking reactor primary water samples and using them to fill the PWM counting chamber while the chamber was isolated from the primary water system. The PWM response was displayed in the control room and recorded by the PWM chart recorder. A 500 ml volume of the same reactor water sample that was used to fill the PWM counting chamber was sealed in a marinelli beaker and analyzed with the Ge(Li) detection system. The Ge(Li) counting results were corrected according to expressions (1), (2) and (3) to provide water concentrations which would be compared to those being predicted by the PWM, based on the previously established PWM calibration curve.

Prior to the dynamic flow tests, the PWM counting chamber was connected to the reactor primary water system as shown in Fig. 1. The connection required the installation of a pressure regulator to reduce the primary system water pressure on the PWM GM tube to no

more than 1.03×10^5 N m^{-2} (15 psi). In addition, two pressure gauges and three valves were installed to improve the ability to control flow and pressure. Two of the valves, the inlet (DV17) and outlet (DV21) valves, allowed primary water to pass through the PWM counting chamber when opened, while a third valve (DV20), a drain valve, provided a location downstream of the counting chamber for collecting primary water samples (Fig. 1).

The dynamic flow test for the PWM was carried out by allowing reactor primary water to flow through the monitor's counting chamber. After the water flow had been established, the PWM recorder in the reactor control room was started, and reactor primary water samples were taken from the drain pipe immediately downstream from the PWM sample chamber. The time of collection was recorded for specific samples. Each primary water sample (500 ml per sample) was then sealed into a marinelli beaker and measured with the Ge(Li) detection system. The concentrations obtained from the Ge(Li) analyses were then corrected for decay using expressions (1) and (2), to provide a basis for comparison to the $\mu Ci\, ml^{-1}$ concentrations being projected from the PWM net counting rate and calibration expression.

During the course of the experimental work, a water leak into the PWM preamplifier was incorrectly interpreted as a GM tube malfunction. Consequently, several different GM tubes of the exact same type[1] were used. Operational tests of different tubes with

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[1] Amperex Electronic Corporation, Model 912NB3.

respect to background, operating voltage, counting plateau, response to solutions of ^{56}Mn, and static and dynamic flow tests showed very little difference. More specifically, differences in PWM response due to different GM tubes was unobservable when compared to the normal counting variations present in the ratemeter and recorder. This was not only helpful for this study, but will also be valuable to know in the future when the current detector fails.

EXPERIMENTAL RESULTS AND DISCUSSION

The counting efficiency calibration performed on the Ge(Li) detection system utilized gamma energies from a Europium-152 NBS standard source as shown earlier in Table 1. The gamma counting efficiencies at the energies shown in Table 1 were plotted as displayed previously in Fig. 3, and the data were found to fit a power curve with the following parameters:

$$\text{Gamma Counting Efficiency} = 1.021\ (E_\gamma)^{-0.7762} \qquad (5)$$

Analysis of reactor primary water to determine radionuclides normally present and their respective concentrations was conducted using the Ge(Li) detection system. Typical concentrations for the three major radionuclides found after the reactor had been operating normally for about four hours at one megawatt have already been given in Table 2. As these three major radionuclides decayed away, additional radionuclides at lower concentrations were also identified and have been previously tabulated in Table 3.

Since it was felt that one of the key objectives of the PWM should be to provide useful data under abnormal as well as normal operating conditions, fission products expected to be released into the primary water following a fuel element cladding failure were also considered. Amendment 4 to the OSTR Safety Analysis Report (OSU75) contained information estimating the saturated activity levels for fission products in a FLIP (70% enriched) TRIGA fuel element, and the fraction of these which would be released into the reactor primary water system due to a cladding failure. Based on the data from this

Amendment to the Safety Analysis Report, it appears that a single element cladding failure would (in the worst case) release approximately 111 mCi of total halogens, including 94 mCi of total iodine isotopes, and 156 mCi of noble gas activity. Table 4 gives a listing of all the major fission products expected to be released into the OSTR primary water system following a fuel element cladding failure, based on a tabulation found in the OSTR Safety Analysis Report (Ri68), and work reported by Bouchey and Gage in 1970 (Bo70).

The fission product nuclides expected are mainly the gaseous radioisotopes of bromine, iodine, krypton and xenon. Bromine and iodine are very soluble in water and will largely remain in the reactor primary water system. The noble gases krypton and xenon will subsequently decay into their particulate daughters, rubidium and cesium, respectively, which will also partially remain in the primary water. Under the stated circumstances, the projected (OSU75) halogen concentration in the reactor pool (reactor primary water system) would be 5.89×10^{-3} μCi ml^{-1}.

As stated above, Tables 2, 3 and 4 summarize the major radionuclides present in the reactor primary water system during normal operation, and those expected to be present during a fuel element cladding failure. Most of the radionuclides identified have a prominent gamma energy between 847 keV and 1369 keV, while most of the others are close to falling within this energy range. Thus, it is possible to establish a reasonable calibration for the present PWM system for both normal and abnormal operations using ^{56}Mn (847 keV gamma peak) and ^{24}Na (1369 keV gamma peak). It is, of course,

Table 4. Major fission products expected to be released into the OSTR primary water system following a fuel element cladding failure.

Radionuclide	Half-life	Prominent Gamma Energy and Abundance (keV)
Bromine-87	55.6 seconds	1440 (100%)
Iodine-131	8.05 days	364 (82%)
Iodine-132	2.26 hours	668 (100%), 773 (89%), 955 (22%)
Iodine-133	20.3 hours	530 (90%)
Iodine-134	52.0 minutes	850 (95%), 890 (65%)
Iodine-135	6.68 hours	1140 (37%), 1280 (34%)
Krypton-89	3.18 minutes	230 (85%), 600 (100%), 880 (65%), 1120 (45%)
Krypton-90	33 seconds	120 (65%), 536 (48%), 1110 (48%)
Rubidium-89	15.4 minutes	1050 (75%), 1260 (54%)
Rubidium-90	2.91 minutes	830 (61%)
Xenon-133	5.27 days	81 (37%)
Xenon-135	9.14 hours	250 (91%)
Xenon-138	17.5 minutes	260 (100%), 1780 (66%)
Xenon-139	43 seconds	220 (100%), 300 (54%) 1150 (23%)
Cesium-138	32.2 minutes	1010 (25%), 1426 (73%)
Cesium-139	9.5 minutes	1280 (strong)

essentially certain that the PWM would overestimate the water radioactivity concentration under abnormal conditions, due to multiple photon emissions at high abundances from certain fission products. Since this response would be on the conservative side and would have the effect of increasing the system's overall sensitivity to undesirable situations, it was not considered worthwhile to derive a second calibration curve just for fission products. In addition, water samples could and would be immediately analyzed on the Ge(Li) system following any abnormalities indicated by the PWM.

The effect of reactor-contributed background on the PWM's readings was measured at three power levels; 250 kW, 500 kW and 1000 kW. The results of this study are given in Table 5, and include values for the three different power levels based on the operating time at each power level. These data revealed that the PWM total radiation background is dependent not only on the reactor power level, but also on the time the reactor has been operated at a given power level. However, since the background counting rate, with or without consideration of the reactor contribution, is small compared to the counting rates observed in the presence of normal primary water radioactivity concentrations, the study turned out to be mainly academic. Generally, background values used will have little impact on water concentrations predicted by the PWM. Also, power levels below 100 kW did not make a measurable contribution to the PWM background, and therefore the background at these lower powers should be assumed equal to the preoperational background.

Table 5. The influence of reactor power and operating time on the reactor contribution to the primary water monitor background counting rate.

Reactor Operating Power (P) (kW)	Reactor Operating Time (T) (Hours)	PWM Total Background Counting Rate[1] (cpm)	Net Reactor Background[2] Contribution (cpm)
0	0[3]	40	0
100	3	45	5
250	1	42	2
250	2	46	6
250	3	49	9
250	4	54	14
500	1	44	4
500	2	48	8
500	3	55	15
500	4	64	24
1000	1	60	20
1000	2	65	25
1000	3	72	32
1000	4	80	40
1000	5	88	48
1000	6	98	58

[1] All measurements here were performed by using the PWM 200 cpm scale.

[2] Net reactor background contribution = $[\text{PWM total background}]_{(P,T)} - [\text{PWM total background}]_{(0,0)}$.

[3] Before reactor startup in the morning.

The average net count rates from the PWM and the corresponding average concentrations measured with the Ge(Li) detection system for the specifically prepared ^{56}Mn and ^{24}Na solutions used in the static flow PWM calibration runs are given in Table 6 and Table 7. When the same data were plotted, as presented in Fig. 4, the PWM showed a slightly more sensitive response for ^{56}Mn (lower gamma energy) than for ^{24}Na (higher gamma energy). The resulting PWM calibration curve (Fig. 5) was obtained by performing a least squares fit of all static flow ^{56}Mn and ^{24}Na data. The result was the following power function expression with a coefficient of determination (r^2) of 0.9956:

$$\begin{array}{l}\text{Gross water}\\ \text{radioactivity}\\ \text{concentration}\\ \text{in } \mu Ci\ ml^{-1}\\ \text{(excluding } ^{3}H)\end{array} = 1.11 \times 10^{-7}(\text{net cpm}^{1.0222}) \qquad (6)$$

As mentioned earlier, the GM tube in the PWM was changed in response to a water leak in the preamplifier. As a result, it was deemed necessary to confirm the applicability of the established calibration expression with a different GM tube of exactly the same type. Verification of the calibration equation was carried out using a ^{56}Mn solution prepared as described previously. The results of the ^{56}Mn run with a different GM tube are shown in Table 8, and based on the previously established calibration expression, a different GM tube provided concentrations which averaged 1.01 of the values indicated by the Ge(Li) system for the ^{56}Mn solution. This confirmed that the current GM tube provided essentially the same degree of accuracy as the earlier tube.

Table 6. Observed net count rates for various ^{56}Mn concentrations in the primary water monitor under static flow conditions

PWM Scale & Observation Number	Average Observed Net Count Rate with the PWM (cpm)	Manganese-56 Concentration ($\mu Ci\ ml^{-1}$)
200K cpm Scale		
1.	99710	1.47×10^{-2}
2.	89705	1.27×10^{-2}
3.	79700	1.11×10^{-2}
4.	69690	9.36×10^{-3}
5.	59680	7.85×10^{-3}
6.	49670	6.31×10^{-3}
7.	39670	5.11×10^{-3}
8.	29670	3.71×10^{-3}
9.	24670	2.93×10^{-3}
10.	19675	2.32×10^{-3}
20K cpm Scale		
11.	15680	1.98×10^{-3}
12.	13685	1.72×10^{-3}
13.	11690	1.46×10^{-3}
14.	9695	1.19×10^{-3}
15.	8700	1.07×10^{-3}
16.	7700	9.44×10^{-4}
17.	6705	8.12×10^{-4}
18.	5705	6.86×10^{-4}
19.	4710	5.61×10^{-4}
20.	3715	4.36×10^{-4}
21.	3215	3.71×10^{-4}
22.	2720	3.09×10^{-4}
23.	2225	2.40×10^{-4}
24.	1730	1.77×10^{-4}
2K cpm Scale		
25.	1330	1.72×10^{-4}
26.	1140	1.46×10^{-4}
27.	942	1.22×10^{-4}*
28.	755	9.60×10^{-5}*
29.	658	8.37×10^{-5}
30.	560	7.20×10^{-5}
31.	465	5.93×10^{-5}
32.	430	5.22×10^{-5}
33.	380	4.61×10^{-5}
34.	332	4.04×10^{-5}
35.	282	3.45×10^{-5}
36.	232	2.82×10^{-5}
37.	185	2.19×10^{-5}
38.	140	1.57×10^{-5}

*Typical value for one megawatt equilibrium concentration in OSTR primary water.

Table 7. Observed net count rates for various ^{24}Na concentrations in the primary water monitor under static flow conditions

PWM Scale & Observation Number	Average Observed Net Count Rate with the PWM (cpm)	Sodium-24 Concentration ($\mu Ci\ ml^{-1}$)
200K cpm Scale		
1.	99765	1.77×10^{-2}
2.	89740	1.53×10^{-2}
3.	79748	1.34×10^{-2}
4	69775	1.15×10^{-2}
5.	59793	9.68×10^{-3}
6.	49810	7.96×10^{-3}
7.	39820	6.24×10^{-3}
8.	29830	4.52×10^{-3}
9.	24830	3.31×10^{-3}
10.	19830	2.47×10^{-3}
20K cpm Scale		
11.	15720	2.28×10^{-3}
12.	13730	1.93×10^{-3}
13.	11765	1.63×10^{-3}
14.	9830	1.52×10^{-3}
15.	8755	1.30×10^{-3}
16.	7755	1.10×10^{-3}
17.	6977	9.13×10^{-4}
18.	5790	7.67×10^{-4}
19.	4725	6.17×10^{-4}
20.	3750	4.75×10^{-4}
21.	3245	4.14×10^{-4}
22.	2725	3.46×10^{-4}
23.	2250	2.75×10^{-4}
24.	1750	2.18×10^{-4}
2K cpm Scale		
25.	1355	2.13×10^{-4}
26.	1155	1.80×10^{-4}
27.	975	1.46×10^{-4}**
28.	725	1.14×10^{-4}**
29.	640	9.66×10^{-5}
30.	575	8.25×10^{-5}
31.	440	6.56×10^{-5}
32.	390	5.96×10^{-5}
33.	370	5.28×10^{-5}
34.	325	4.48×10^{-5}
35.	290	3.56×10^{-5}
36.	210	2.53×10^{-5}
37.	165	2.11×10^{-5}
38.	120	1.47×10^{-5}

*Typical value for one megawatt equilibrium concentration in OSTR primary water.

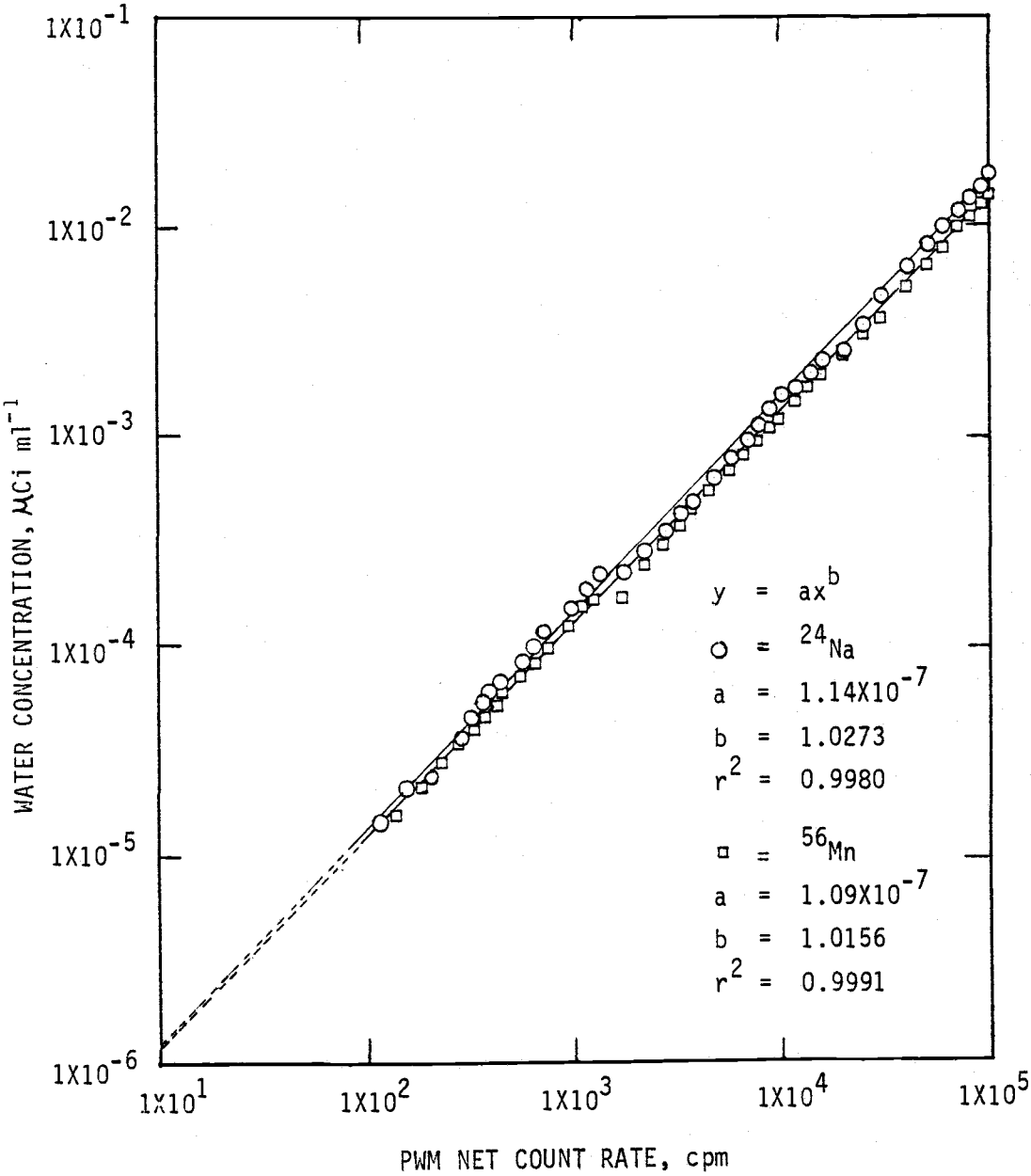

Fig. 4. Primary water monitor net counting rate in response to various water concentrations of ^{56}Mn and ^{24}Na under static flow conditions.

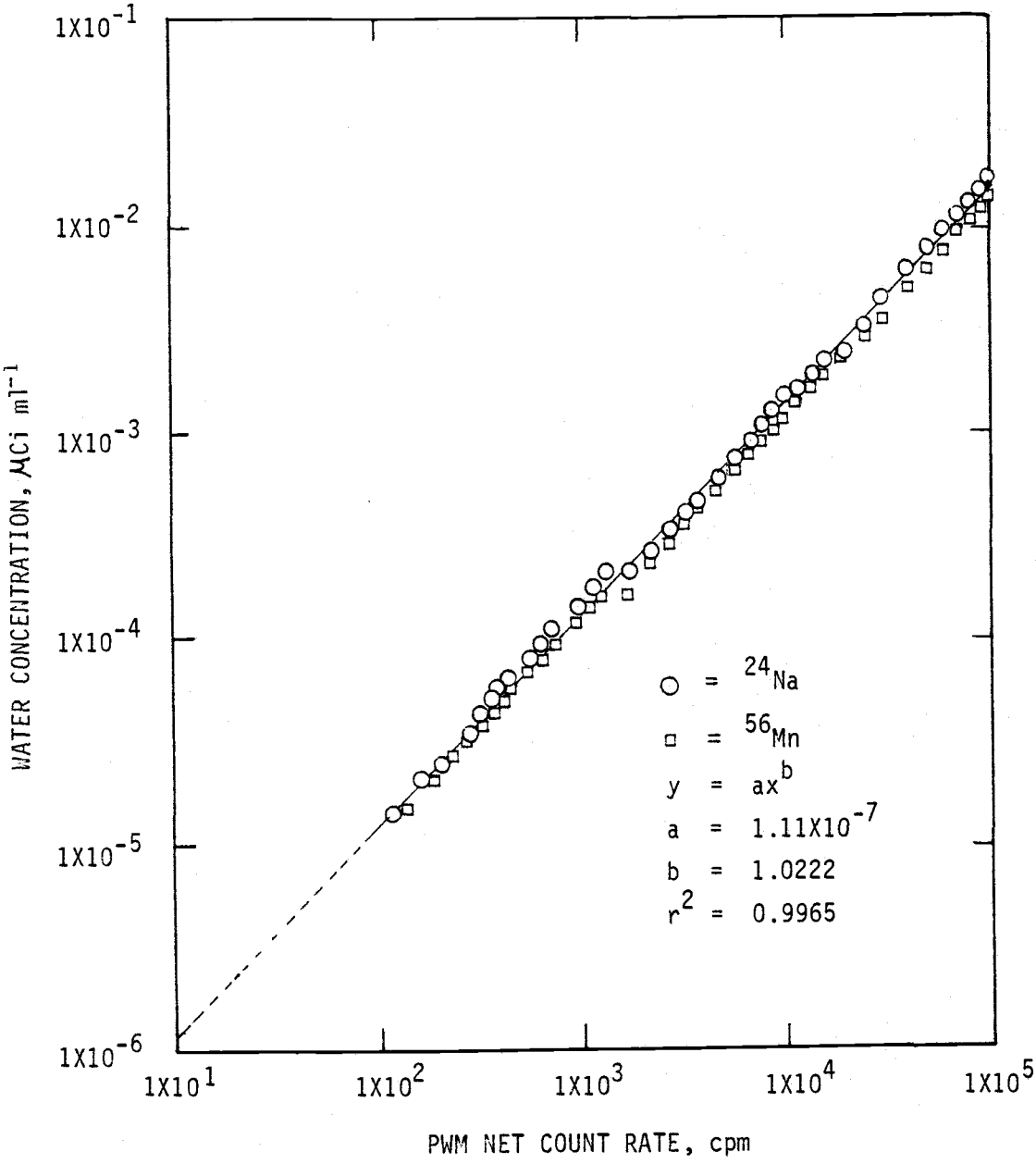

Fig. 5. PWM calibration curve based on predomonant radionuclides in the OSTR primary water (excluding ^{3}H): static flow conditions.

Table 8. A comparison of ^{56}Mn concentrations measured by the Ge(Li) system and the PWM equipped with a different GM tube of the same type under static flow conditions.

PWM Scale & Observation Number	Observed Net Count Rate With the PWM (cpm)	Manganese-56 Concentrations PWM ($\mu Ci\ ml^{-1}$)	Ge(Li) ($\mu Ci\ ml^{-1}$)	Ratio PWM:Ge(Li)
20K cpm Scale				
1.	15884	2.01×10^{-3}	1.97×10^{-3}	1.02
2.	11708	1.48×10^{-3}	1.51×10^{-3}	0.98
3.	8808	1.11×10^{-3}	1.15×10^{-3}	0.96
4.	6550	8.19×10^{-4}	8.80×10^{-4}	0.93
5.	5558	6.93×10^{-4}	6.73×10^{-4}	1.03
6.	4304	5.35×10^{-4}	5.14×10^{-4}	1.04
7.	3020	3.73×10^{-4}	3.93×10^{-4}	0.95
8.	2125	2.61×10^{-4}	3.00×10^{-4}	0.87
2K cpm Scale				
9.	1723	2.11×10^{-4}	2.29×10^{-4}	0.92
10.	1548	1.89×10^{-4}	1.75×10^{-4}	1.08
11.	1166	1.42×10^{-4}	1.34×10^{-4}	1.06
12.	828	1.00×10^{-4}	1.02×10^{-4}	0.98
13.	622	7.50×10^{-5}	7.82×10^{-5}	0.96
14.	507	6.09×10^{-5}	5.97×10^{-5}	1.02
15.	400	4.79×10^{-5}	4.56×10^{-5}	1.05
16.	320	3.82×10^{-5}	3.49×10^{-5}	1.09
17.	250	2.97×10^{-5}	2.66×10^{-5}	1.11
18.	180	2.15×10^{-5}	2.04×10^{-5}	1.06

The independent verification of the PWM calibration (using reactor primary water) was conducted under both static flow and dynamic flow conditions. For the static flow verification, two completely different samples of reactor primary water were used to obtain two separate comparisons of the relationship between the reactor primary water concentration measured by the Ge(Li) and predicted by the PWM. Table 9 gives the results of these comparisons and shows an average ratio of 1.01 between the predictions of the PWM and the Ge(Li) measurements.

Verification of the PWM's performance under dynamic flow conditions utilized 18 samples of primary water taken from the drain pipe for the PWM counting chamber, while reactor primary water was flowing through the chamber. A total of eight samples were taken while the reactor was operating between 80 kW and 500 kW, and another ten samples were taken while the reactor was operating at 1000 kW. These samples were measured using the Ge(Li) detection system and concentrations were compared to the concentrations indicated by the PWM. Table 10 gives the results of these comparisons and shows an average ratio for the predicted concentrations from the PWM to those measured by the Ge(Li) system of 2.14 between reactor powers of 80 kW and 500 kW, and 2.34 at 1 MW. The overall average ratio considering all reactor power levels was approximately 2.26.

With the apparent difference in predicted water concentration between the static flow and dynamic flow conditions, further experiments were performed to try to identify the cause. It was thought that perhaps the difference was due to ^{16}N concentrations that were

Table 9. A comparison of radioactivity concentrations in OSTR primary water as measured by the Ge(Li) detection system and the PWM under static flow conditions

Sample Number and Observation Number	Observed Net Count Rate With the PWM (cpm)	Indicated Concentration of Primary Water Sample		Ratio PWM:Ge(Li)
		PWM (μCi ml^{-1})	Ge(Li) (μCi ml^{-1})	
Primary Water Sample 1				
1.	3950	5.27×10^{-4}	4.27×10^{-4}	1.23
2.	2345	3.09×10^{-4}	2.93×10^{-4}	1.06
3.	1790	2.35×10^{-4}	2.30×10^{-4}	1.02
4.	1320	1.72×10^{-4}	1.75×10^{-4}	0.98
5.	1300	1.69×10^{-4}	1.72×10^{-4}	0.99
6.	1104	1.43×10^{-4}	1.45×10^{-4}	0.99
7.	906	1.17×10^{-4}	1.15×10^{-4}	1.02
8.	708	9.09×10^{-5}	8.76×10^{-5}	1.04
9.	510	6.50×10^{-5}	6.42×10^{-5}	1.01
10.	324	4.09×10^{-5}	4.24×10^{-5}	0.96
Average				1.03
Primary Water Sample 2				
1.	4918	6.59×10^{-4}	6.16×10^{-4}	1.07
2.	3916	5.22×10^{-4}	5.98×10^{-4}	0.87
3.	2934	3.89×10^{-4}	4.38×10^{-4}	0.89
4.	1940	2.55×10^{-4}	2.72×10^{-4}	0.94
Average				0.94
Overall Average for Samples 1 and 2				1.01

Table 10. A comparison of radioactivity concentrations in OSTR primary water as measured by the Ge(Li) detection system and the PWM under dynamic flow conditions

Reactor Power & Sample Number	Observed Net Count Rate With the PWM (cpm)	Indicated Concentration of Primary Water Sample		Ratio PWM:Ge(Li)
		PWM (μCi ml^{-1})	Ge(Li) (μCi ml^{-1})	
80 kW				
1.	1417	1.85×10^{-4}	9.01×10^{-5}	2.05
2.	1582	2.07×10^{-4}	9.66×10^{-5}	2.14
250 kW				
1.	2496	3.30×10^{-4}	1.53×10^{-4}	2.16
2.	3120	4.14×10^{-4}	2.20×10^{-4}	1.88
3.	4123	5.51×10^{-4}	2.53×10^{-4}	2.18
500 kW				
1.	4940	6.62×10^{-4}	3.04×10^{-4}	2.18
2.	6404	8.64×10^{-4}	3.47×10^{-4}	2.48
3.	7650	1.04×10^{-3}	5.06×10^{-4}	2.05
Average ratio at powers of 80 kW, 250 kW and 500 kW.....				2.14
1000 kW				
1.	7450	1.01×10^{-3}	4.22×10^{-4}	2.39
2.	10546	1.44×10^{-3}	5.24×10^{-4}	2.74
3.	13936	1.91×10^{-3}	8.71×10^{-4}	2.19
4.	15130	2.08×10^{-3}	8.84×10^{-4}	2.35
5.	15270	2.10×10^{-3}	1.01×10^{-3}	2.08
6.	10696	1.46×10^{-3}	5.91×10^{-4}	2.47
7.	13420	1.84×10^{-3}	7.77×10^{-4}	2.37
8.	15320	2.11×10^{-3}	1.02×10^{-3}	2.06
9.	15716	2.16×10^{-3}	9.46×10^{-4}	2.28
10.	13667	1.87×10^{-3}	7.23×10^{-4}	2.59
Average ratio at reactor power of 1000 kW.....				2.35
Overall average ratio.....				2.26

being detected by the PWM under dynamic conditions, but due to rapid ^{16}N decay, were not being counted by the Ge(Li) detection system. Experiments in which the PWM counting chamber was instantaneously isolated during dynamic flow did not show the rapid drop in counting rate to be expected if there was a significant concentration of ^{16}N in the water circulating through the PWM. In fact, no real explanation was found for the higher predictions of water radioactivity from the PWM during dynamic flow. It was concluded, however, that the increase seemed very consistent, and it was later hypothesized that physical characteristics associated with water flowing past the (rather long) GM tube might be causing vibrations leading to an electronic noise signal.

Since the PWM's response under dynamic conditions was consistently higher than the independently measured Ge(Li) concentrations, by an average factor of 2.26, it was considered necessary to modify the basic static flow calibration expression for the PWM to account for this disparity. As a result, it was decided to divide the existing expression by 2.26 to obtain concentrations under dynamic flow conditions which would be very close to those predicted by the Ge(Li) system. Thus, the final PWM calibration expression is:

$$\begin{array}{l}\text{Gross water}\\ \text{radioactivity}\\ \text{concentration}\\ \text{in } \mu\text{Ci ml}^{-1}\\ \text{(excluding } ^{3}\text{H)}\end{array} = \left[\frac{(1.11 \times 10^{-7})(\text{net cpm}^{1.0222})}{2.26}\right] \qquad (7)$$

CONCLUSIONS

This study primarily involved the development of a technique for calibrating and independently verifying the performance of a primary water radioactivity monitor at a TRIGA research reactor. The technique is quite simple and easily performed after the system has been installed. In addition, however, the work was to include the specification and application of simple performance objectives and the recognition of operational characteristics for a conventional on-line PWM. In this respect, it is concluded that (1) the system should provide useful data during both normal and abnormal situations; (2) that a single calibration based on radionuclides present during normal operations is acceptable, and slightly conservative, for abnormal conditions; and (3) that exchanging GM tubes of the exact same type does not appear to significantly change the basic calibration expression for the PWM system.

Two separate studies were carried out which compared reactor primary water concentrations predicted by the PWM to those measured independently with the Ge(Li) system. In the first cast, a reactor primary water sample was divided, and one part was counted on the Ge(Li) while the other part was placed into the PWM under static flow conditions. Agreement between the two predicted concentrations averaged 1.01 based on a ratio of PWM predicted concentrations to those predicted by the Ge(Li). In the second case, the reactor primary water was allowed to flow through the PWM under dynamic

conditions. In this operating mode, the PWM's predicted concentrations averaged 2.26 times higher than those measured independently by the Ge(Li) detection system. Thus, the final PWM calibration expression must be divided by a factor of 2.26 for operational application.

Since no definite explanation could be found for the PWM's different response during static and dynamic conditions, a further study is warranted to evaluate this inconsistency.

REFERENCES

Bo70 Bouchey, G., Gage, S., 1970, Detection and Location of Leaking TRIGA Fuel Element, in: First TRIGA Reactor Owners Conference Proceedings, Denver, Colorado, 19-20 Feb., 1970, pp 43-50 (Gulf-General Atomic Co.).

DHEW70 U.S. Department of Health, Education and Welfare, 1970, Radiological Health Handbook, (Washington, D.C.: U.S. Government Printing Office).

He56 Heath, R., 1956, Fission Product Monitoring in Reactor Coolant Water, Nucleonics 15(12),54.

Mo57 Moeller, D., 1957, Radionuclides in Reactor Cooling Water-Identification, Source, and Control, Oak Ridge National Laboratory, ORNL-2311.

OSU75 Oregon State University, 1975, Amendment No. 4 to the Safety Analysis Report for the Oregon State TRIGA Reactor (OSTR), (Corvallis: Oregon State University Radiation Center).

Pi80 Picket, B., 1980, Independent Determination of the Accuracy of the Stack Gas Monitor at the Oregon State University TRIGA Mark II Reactor, Research Report, Department of General Science, Oregon State University.

Ri68 Ringle, J., Anderson, T., and Johnson, A., 1968, Safety Analysis Report for the Oregon State University TRIGA Research Reactor, (Corvallis: Oregon State University Radiation Center).

Tr66 Tracerlab, 1966, MWP-4A Low Level Liquid Monitor, Operating and Maintenance Instructions, (Richmond: Tracerlab).