NRC Job Code V6060: Extended In-Situ and Real Time Monitoring

Task 4: Detection and Monitoring of Leaks at Nuclear power Plants External to Structures

Argonne National Laboratory

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ACRONYMS AND ABBREVIATIONS

ANL	Argonne National Laboratory
BIXS	Beta-ray Induced X-ray Spectrometry
DTS	Distributed Temperature Sensing
EIC	Electret Ion Chamber
EIT	Electrical Impedance Tomography
EM	Electromagnetic
EPRI	Electric Power Research Institute
ERT	Electrical Resistance Tomography
GPR	Ground Penetrating Radar
LSC	Liquid Scintillator Counter
MERIT	Magneto-Electrical Resistivity Imaging Technique
NPP	Nuclear Power Plant
NRC	Nuclear Regulatory Commission
NRR	Nuclear Regulatory Research
SSC	Structures, Systems and Components
TDR	Time-Domain Reflectometry
TLD	Thermoluminescence Dosimeter

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1.0 Introduction

In support of Task 4 of the NRC study on compliance with 10 CFR part 20.1406, minimization of contamination, Argonne National Laboratory (ANL) conducted a one-year scoping study, in concert with a parallel study performed by NRC/NRR staff, on monitoring for leaks at nuclear power plants (NPPs) external to structures. The objective of this task-4 study is to identify and assess those sensors and monitoring techniques for early detection of abnormal radioactive releases from the engineered facility structures, systems and components (SSCs) to the surrounding underground environment in existing NPPs and planned new reactors. As such, methods of interest include: 1) detection of anomalous water content of soils surrounding SSCs, 2) radionuclides contained in the leaking water, and 3) secondary signals such as temperature.

ANL work scope includes mainly to (1) identify, in concert with the nuclear industry, the sensors and techniques that have most promise to detect radionuclides and/or associated chemical releases from SSCs of existing NPPs and (2) review and provide comments on the results of the NRC/NRR staff scoping study to identify candidate technologies.

This report constitutes the ANL deliverable of the task-4 study. It covers a survey of sensor technologies and leak detection methods currently applied to leak monitoring at NPPs. The survey also provides a technology evaluation that identifies their strength and deficiency based on their detection speed, sensitivity, range and reliability. Emerging advanced technologies that are potentially capable of locating releases, identifying the radionuclides, and estimating their concentrations and distributions are also included in the report along with suggestions of required further research and development.

2.0 Sources of Leaks at NPPs

A report on lessons-learned review of nuclear incidents involving an inadvertent release of radioactive liquids to the environment was documented in 2006 by the NRC chartered Task Force[1]. Sources of releases and radionuclides detected are summarized in Table1 along with a recent incident at the Vermont Yankee power plant. Most leaks were attributed to fuel transfer systems, spent fuel pools, buried piping, and storage tanks.

Electric Power Research Institute (EPRI) also provided the essential technical guidance to nuclear power plants on groundwater protection. In a recent EPRI report[2], potential ongoing radionuclide releases were identified and grouped into release sources of (1) high concentration low leak rate and (2) low concentration high leak rate. The group one includes the spent fuel pool, the refueling water storage tank, sumps for valve penetration rooms, chemistry sampling lines for primary systems, both lined and unlined concrete pits within the primary or secondary side of the plant, building construction joints below grade, and the radwaste discharge pipeline. The second group composes of secondary cooling water tanks and pipelines, secondary side steam condensate, unlined retention basins, and other water transport mechanisms such as foundation drains, roof drains, and storm drains.

Earlier documented events on groundwater contamination[3] by EPRI include:

- In early 1960s, in the Yankee atomic electric company's reactor in Rowe Massachusetts, a large quantity of contaminated water in *the* ion exchange pit leaked through a defect in the concrete wall for a period of several years. Initial detected tritium concentration in Sherman Spring was a few million picocuries per liter (pCi/L). Other events that might also cause the contamination were subsurface liquid radioactive waste system pipe breaks and spills and spent fuel pool leakage. Monitoring techniques involved well drilling, sampling, and chemical analysis.
- Connecticut Yankee plant, 545MWe pressurized water reactor operated from 1968 to 1996. Identified events that contributed to groundwater contamination were (a) refueling water storage tank leak, (2) drumming room water discharge line failure, (3) spent fuel pool and (4) other leakage events. In 1998, 31 monitoring wells at various depths were installed to collect samples for analysis of all gamma emitting radionuclides and tritium.

The release incidents were identified by means of detecting radionuclides, primarily the tritium concentration, and the anomalous water content. Anomalous water content in vadose zone and groundwater may be confirmed from changes in temperature and pressure, conductivity and resistivity of groundwater, and anomalous pH or other chemical indicators.

A recent report[4] on radiological effluents released by U.S. commercial nuclear power plants from 1995 – 2005 indicated that the releases had been level over the time period and public doses were well below 1 % of the regulatory limits.

Table 1 Inadvertent release of radioactive liquids at U.S. nuclear power plants

Nuclear Power Plant	Date of Release Discovery	Source of Release	Radionuclides Detected
Braidwood	March 2005	Vacuum breaker valves on the circulating water blow down line	Tritium
Byron	February 2006	Vacuum breaker valves on the circulating water blow down line	Tritium
Callaway	June 2006	Vacuum breaker valves on the circulating water blow down line	Tritium, cobalt-58, Cobalt-60, cesium-134, cesium-137
Dresden	August 2004, January 2006	Non-safety related HPCI suction and return line	Tritium
Hatch	December 1986	Fuel transfer canal due to operator action	Tritium
Indian Point	August 2005	Unit 1 and unit 2 spent fuel pools	Tritium, nickel-63, cesium-137, strontium-90, and cobalt-60
Oyster Creek	September 1996	Condensing onto property after a gaseous release	Tritium
Palo Verde	March 2006	Rain condensate transfer system due to operator action	Tritium
Perry	March 2006	Feedwater venturi	Tritium
Point Beach	1999	Retention pond	Tritium, cesium-137
Seabrook	June 1999	Spent fuel pool	Tritium
Salem	September 2002	Spent fuel pool	Tritium
Three Mile Island	May 2006	Condensate storage tank	Tritium
Watts Bar	August 2002	Effluent release pipe and SFP transfer tube sleeve	Tritium and mixed fission products
Vermont Yankee	January 2011	Steam pipes inside the Advanced Off-Gas pipe tunnel	Tritium

3.0 Detection Methods

In this section, we compiled the detection methods covering mainly in-situ and real-time measurement or monitoring techniques. Sampling techniques followed by laboratory analyses will not be included. The survey focuses mainly on detection of radionuclides such as cesium-137, β-decay tritium, and water content or moisture in soil.

3.1 Detection of Radionuclides

Table 2 lists the major radionuclides present in the liquid releases from nuclear power plant to the environment. Their primary mode of decay is β-decay; emissions of γ-ray are also associated with some of the nuclides. Commonly used radiation detectors applied to field measurements have been reviewed[5,6]. In general, radiation detectors can be categorized into four classes: (1) scintillation detectors, (2) solid-state detectors, (3) gas-filled detectors, and (4) passive integrating detectors.

Table 2 Radionuclides present in the liquid release

Radionuclide	Mode of decay/ Energy MeV	Half life Years	γ Emission Energy MeV
Tritium	β-Decay/0.0186	12.3	--
Cobalt-60	β-Decay/0.315	5.26	1.33
Nickel-63	β-Decay/0.0659	92.	--
Iodine-129	β-Decay/0.15	1.7×10^7	0.04
Cesium-134	β-Decay/0.658	2.05	0.605
Cesium-137	β-Decay/0.514	30.	0.662
Strontium-90	β-Decay/0.546	28.1	--

A *scintillation* detector contains a solid or liquid medium that undergoes electronic transitions to excited states upon interactions with radiations. The excited states decay rapidly by emitting photons that are in turn captured by a photomultiplier and converted into electrical signals. The common scintillation material is NaI(Tl) for gamma detection.

A *solid-state* detector is basically a semiconductor sensor that creates electron-hole pairs upon absorption of radiations. A charged electrode then collects the current *and produces the signal. The design and operating conditions of a specific solid*-state detector determines the types of radiation to be detected. Typical solid-state detectors are built on germanium and silicon materials. A high-purity germanium detector provides a better energy resolution (2 KeV) than an NaI detector (50 KeV) in detecting gamma rays.

A *gas-filled* detector measures the ion-pairs produced by radiation interaction with gas. The most commonly used fill gases are air or argon gas with a small amount of methane (typically 10% by mass) and argon or helium gas with a small amount of halogen such as chlorine or bromine. Gas-filled detectors based on the region of gas amplification can be categorized as ionization, proportional, or Geiger-Mueller detector.

A *passive integrating detector* stores the absorbed radiation energy over the exposure time and provides an integrated signal to a laboratory or hand-held reader. Representatives of this type of detector are thermoluminescence dosimeters (TLDs) and electret ion chambers (EICs). TLDs are basically inorganic crystals imbedded with impurities such as CaF_2:Mn, CaF_2:Dy, $CaSO_4$:Mn, and Al_2O_3:C. The impurities in the crystalline lattice trap the free electrons and holes resulted from absorption of radiation energy and thus lock the excitation energy in the crystal. When the exposed TLD is heated in a reader, the stored energy will be released optically and detected by the reader. An EIC consists of a stable electret, typically a charged Teflon disk, mounted inside a small electrically charged plastic chamber. The ions produced inside this air filled chamber cause a reduction of surface charge of the electret. The change in voltage across the electret is then measured with a surface potential voltmeter.

Table 3 summarizes the reported detector types and their sensitivities and typical applications.

Table3. Common detectors for beta and gamma ray detection

Detector Type	Detector Configuration	Applications		Sensitivity[#]	Remark
		Beta	Gamma		
Scintillation	NaI(Tl) up to 5cm x 5cm hand-held	--	Surface scanning	1 – 5 μR/hr or 200-1,000 cpm	Energy sensitive with the greatest response around 100-120 keV
	CsI or NaI(Tl) thin crystal	--	Scanning	--	Detection of low-energy radiation
	Plastic scintillator	Contamination measurement	Dose equivalent rate	--	Large area coverage
Solid state	Germanium semiconductor	--	Field γ spectrometry, multichannel analyzer	50keV with P-type, 10keV with N-type	Liquid nitrogen cooling, high voltage operation, long counting time (up to 1000 min)
Gas filled	Proportional	<0.1 mg/cm^2 window; probe area 50 to 1,000 cm^2	Surface scanning and surface contamination measurements	Beta efficiency ranges from 5% to 35%, <1% for gamma	Require a supply of appropriate fill gas.
	Ionization	1-7 mg/cm^2 window	Handheld ionization chamber	0.05 mR/hr for γ. For beta, 10,000 Bq/m^2 @ 1 hr and 500 Bq/m^2 @ 24 hrs.	Exposure rate measurements.
	Geiger - Mueller	< 2 mg/cm^2 window; probe area 10 to 100 cm^2	Pancake (<2 mg/cm^2 window) or side window (~ 30 mg/cm^2)	Around 0.1 mR/hr in rate meter mode or 0.01 mR/hr for integrate mode for gamma detection. ~10% better for beta detection	Relatively poor sensitivity
Passive integrating	TLD	Secondary radiation detected	Left in the field for a period of a day and read in the laboratory on a calibrated reader.	Standard TLD sensitivity at 100 mrem/y, New Al_2O_3 TLD at 0.01 mrem/y.	Sensitive to visible light, direct sunlight, fluorescent light, excessive heat and humidity.
	EIC	7 mg/cm^2 window, or window-less, window area 50-180 cm^2, volume 50-1,000 ml	7 mg/cm^2 window, or window-less, window area 50-180 cm^2, volume 50-1,000 ml	In integrate mode, the sensitivity can be as low as 0.05 mR/hr.	Usable in high humidity and temperature.

μR/hr = 10^{-6} rontgen per hour (1 röntgen = 2.58×10^{-4} Coulomb/kg); cpm = counts per minute; 1becquerel (Bq) = 27 pCi.

Overall, mobile gamma-ray spectrometers with NaI(Tl) detectors are commonly utilized to characterize soil contamination with radionuclides, such as ^{238}U, ^{226}Ra, and ^{232}Th. An example of this type mobile system is the one used at the DOE's Fernald Closure Project. The mobile system includes a radiation tracking system (a full-size farm tractor), a radiation scanning system, a utility vehicle, and an excavation monitoring system. To make the detection accountable, there are several measurement uncertainties[7] that need to be resolved. They are uncertainties in calibration, soil profile of radionuclides, net count rates, corrections due to radon and moisture, and background radiations.

3.2 Detection of Tritium

Tritium is produced in nuclear power plants from mostly nuclear reactions of boron absorbing neutrons; smaller amounts can also be produced from the splitting of U-235 in the reactor core or when other chemicals ($_3Li^6$, $_5B^{10}$ or heavy water) in the coolant water absorb neutrons. Tritium can bond with oxygen to form tritiated water, different from heavy water but chemically identical to normal water. Nuclear power plants routinely and safely release dilute concentrations of tritiated water; normally the resulting radiation level in groundwater is less than EPA drinking water tritium standard, 20,000 pCi/L. Monitoring tritium level in groundwater may therefore provide the detection of leaks from for example the underground piping. Vermont Yankee Nuclear Power Station recently reported detection of more tritium contamination in their deeper groundwater monitoring wells at a level of 500,000 pCi/L in one of their wells. The groundwater contamination was found to be resulting from corroded, buried piping leaks. Similar problems have been identified, as reported in a recent news[8] that radioactive tritium has been detected in the groundwater at more than 48 commercial U.S. nuclear power sites. It becomes apparent that close monitoring of underground tritium level is one of the most important safety requirements at NPPs.

There are two laboratory methods[9] recommended for tritium activity measurement in aqueous samples. The basic principle is the same but differs in chemical treatments of water samples. The first method, ISO 9698, 1989, adds sodium thiosulfate to the water sample to convert iodine to iodide and sodium carbonate to make the sample alkaline. The second method, APHA-AWWA-WEF, 19th edition 1995, recommend sodium peroxide and potassium permanganate to mix with the water sample. The standard analysis procedure[10] is described as follows.

Ground water was purified by double distillation under atmospheric pressure after addition of $KMnO_4$ and Na_2O_2. Purified water of 10ml was mixed with 10ml liquid scintillation cocktail in a Teflon vial. After cooling for 4 d in the liquid scintillation counter at 13°C in dark, tritium was measured for 500 min (50 min x 10 repeats) for each sample. The counting system was calibrated with a tritium standard. The quenching was corrected by the external standard channel ratio method using Ra-226 source. The chemiluminescence contribution was subtracted from the total counting rate to obtain a net counting rate ascribable to tritium only. The background counting rate was determined with a background sample prepared using a tritium-free ground water, collected from a deep well. The typical counting efficiency for tritium was 27-28%, background level of 0.9 ± 0.02 cpm, detection limit or sensitivity for

tritium to be 1.7 Bq/L (46 pCi/L). Typical solvents used for LSC are toluene, pseudocumene, and PXE (phenyl xylylethane). Scintillators, typically phosphors, provide the conversion of captured energy to the emission of light to be measured. Two types of scintillators are required in the cocktail, a primary (e.g., PPO, 2,5-diphenyloxazole with emission wavelength 357 nm) and a secondary (e.g., Bis-MSB, 1,4-bis[2-methylstyryl]benzene with emission wavelength at 420 nm.

Tritium emits beta particles with a maximum energy of 18.54 keV and an average energy of 5.69 keV with a half-life of 12.35 years. Methods to detect beta particles have been covered in Section 3.1; they are generally based on either ionization or scintillation. Since the penetration range of the tritium beta ray is less than 6 mm in air and 6μm in water, non-contact type detection is nearly impossible. Conventional ionization detectors such as Geiger counter have limited in-situ applications. New sensor technology focuses on solid-state detectors. For example, a microfabricated beta-particle detector[11] with dual cavities for energy spectroscopy was recently developed and tested for measuring beta radiation produced from U-238 (0.8 MeV), Sr-90 (0.546 MeV), Co-60 (up to 0.314 MeV) and Tl-204 (max. 0.776 MeV). This sensor may not be applicable to detection of tritium because of low energy of the beta particles emitted by tritium.

Scintillators for beta detectors can be organic or inorganic molecules in a form of liquid, solid or plastics. A liquid scintillation counter is widely used for evaluation of tritium concentration in liquid samples. The liquid scintillators are mainly aromatic organics with dissolved phosphores. The widely used commercial instrument is Quantulus-1220 by Perkin Elmer, which has tritium detection efficiency up to 27%. This type of detector is again limited to laboratory analysis that requires sampling and specific analysis environment. Solid scintillators are mainly used for tritium gas counting, particularly of mixtures containing tritiated water. Common problems encountered are the contamination problem resulting from either tritium exchange with organic scintillators or absorption of tritiated water by hygroscopic crystals such as NaI(Tl). Since a solid scintillation counter relies on surface detection with the detection limit around 1 μCi/l, it can have a much faster time response. However, the system requires a high surface-to-volume ratio and prefiltration of water. It is also vulnerable to fouling and subject to memory effects from tritium retention on the solid scintillant. These problems are partially overcome by using inorganic scintillators with low water solubility such as europium-doped calcium-fluoride (CaF_2[Eu])[12]. Potentially, this type of solid scintillators may be developed into real-time applications.

Plastic scintillators have been developed for tritium gas counting. A plastic scintillator consists of a solid solution of organic scintillating molecules in a polymerized solvent. It can be easily shaped and fabricated. The scintillation emission of a typical plastic scintillator has a maximum around 400 nm with a relatively large light output [typically 25-30% of NaI(Tl)] and a short decay time, on the order of a nanosecond. This makes the material well suited for fast timing measurements. However, plastic scintillators are also sensitive to other charged particles. Different types of plastic scintillator are commercially available (for example, OKEN/NE Technology). Further development in this area may lead to a practical in-situ detector.

In-situ real-time nondestructive detection systems have not been well developed. Two recent developments that adopt rather different technical approaches are briefly described as follows:

β-ray induced X-ray spectrometry (BIXS)[13] -- It is a nondestructive measurement technique for in-situ measurements of tritium concentration distribution in bulk of the metallic materials. The technique is based on the phenomena that emission of β-ray from tritium absorbed in a material gives rise to X-rays, i.e., bremsstrahlung and characteristic X-rays. The former is a continuous spectrum of which the shape is dependent on the depth profile of tritium but independent on the total amount of tritium in the bulk. The latter X-rays depend on both. However, the characteristic X-rays from carbon materials are low energy (0.277 keV) and the stopping power of carbon materials to β-ray is small. To measure the total tritium relies on the intensity of the characteristic X-rays of argon gas (K_α :2.96 keV, K_β : 3.19 keV). At present, this technique is mainly for surface analysis.

Fiber-optic radiation sensor for tritium detection[14] – The sensor is composed of an inorganic scintillator, a plastic optical fiber bundle and a light-measuring device. The fiber bundle and the scintillator are connected using an optical epoxy (DP-100 plus, 3M). Figure 1 shows the basic design of such a system. It detects tritium in real time using a photomultiplier tube. Generated scintillation photons are of 455-550 nm in wavelength. Normally, optical fiber is not influenced by temperature, pressure or electromagnetic waves and it enables long-distance signal transmission and real-time detection. Three types of inorganic scintillators given in Table 4 were tested and Gd_2O_2S:Tb was recommended. Again, the system has not been tested in an actual environment.

Table 4 Inorganic scintillators applicable for fiber-optic radiation sensors

Inorganic Scintillator powder	Wavelength peak	Decay to 10%	Efficiency relative to NaI (%)
Gd_2O_2S:Tb	545 nm	1.5 ms	15
$Y_3Al_5O_{12}$:Ce	550 nm	70 ns	5
CsI:Tl	545 nm	5 μs	8.5

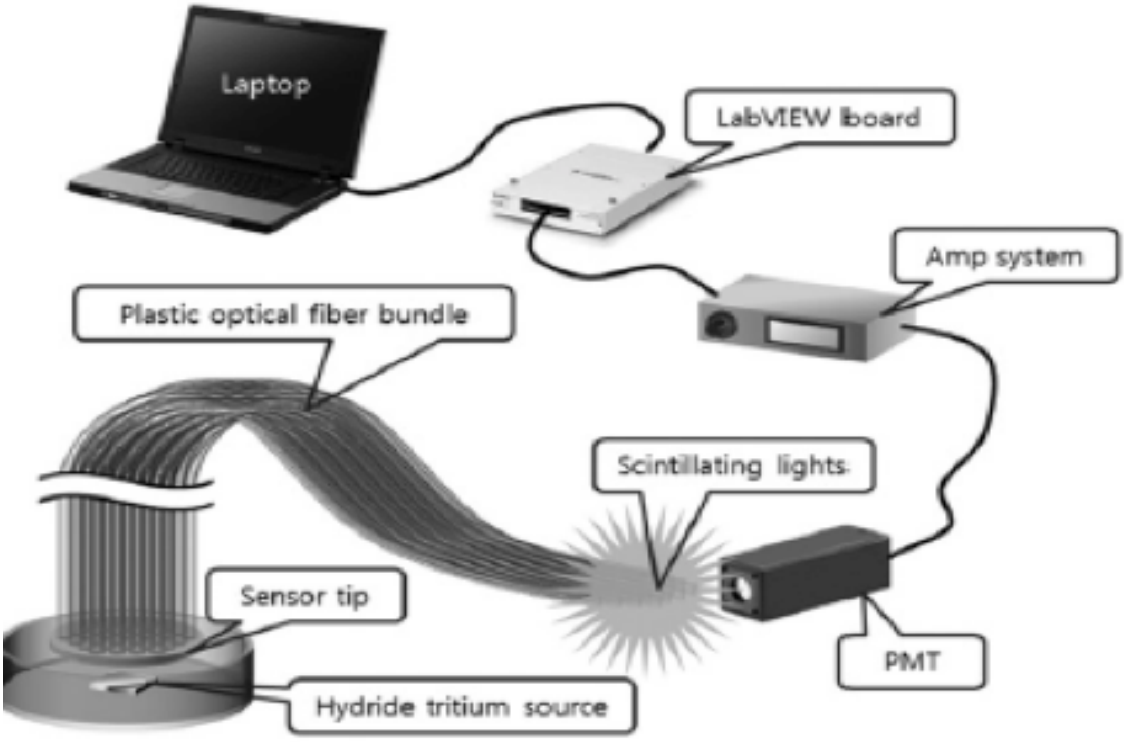

Figure 1: Fiber-optic tritium detection system

The state of the art of tritium analysis relies on mainly sampling technique followed by laboratory analysis. Limited field deployable systems have been demonstrated. Figure 2 shows such a system developed at Savannah River technology center[15]. Groundwater was collected through wells and purified. Samples of the purified water are mixed with liquid scintillators and then analyzed by optical sensors with bismuth germinate windows. Another detection technique recently reported[16] is a micro-fabricated Geiger counter using dual cavities for energy spectroscopy measurement. Figure 3 shows the design of the sensor. A portable system based on the anthracene scintillation method was also reported[17], which demonstrated a sensitivity of better than 5 nCi in approximately 3 min counting time.

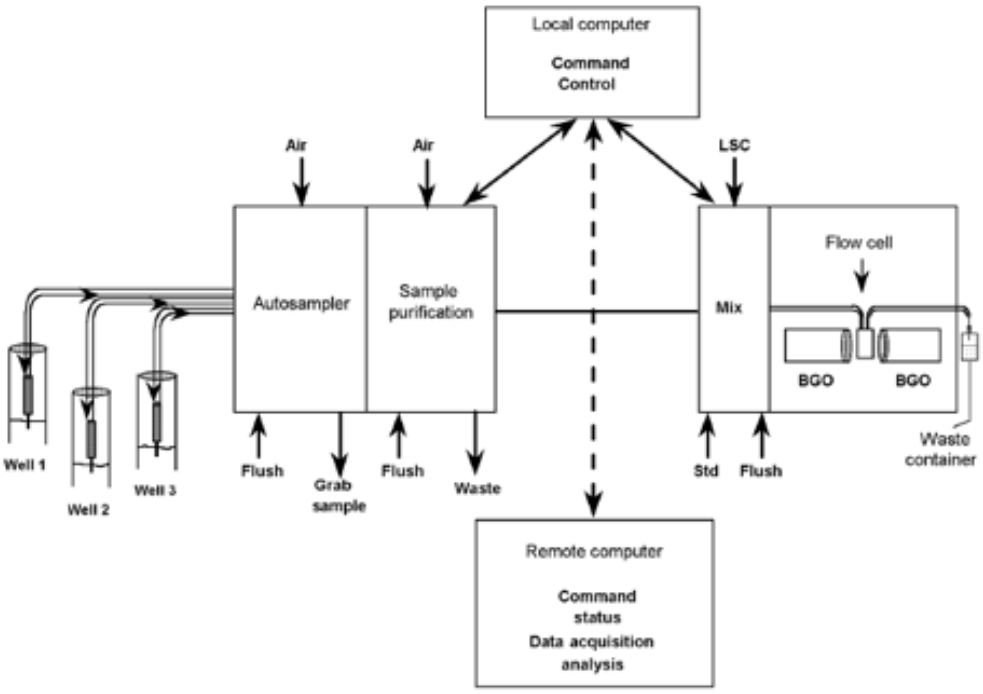

Figure 2: A field deployable tritium analysis system

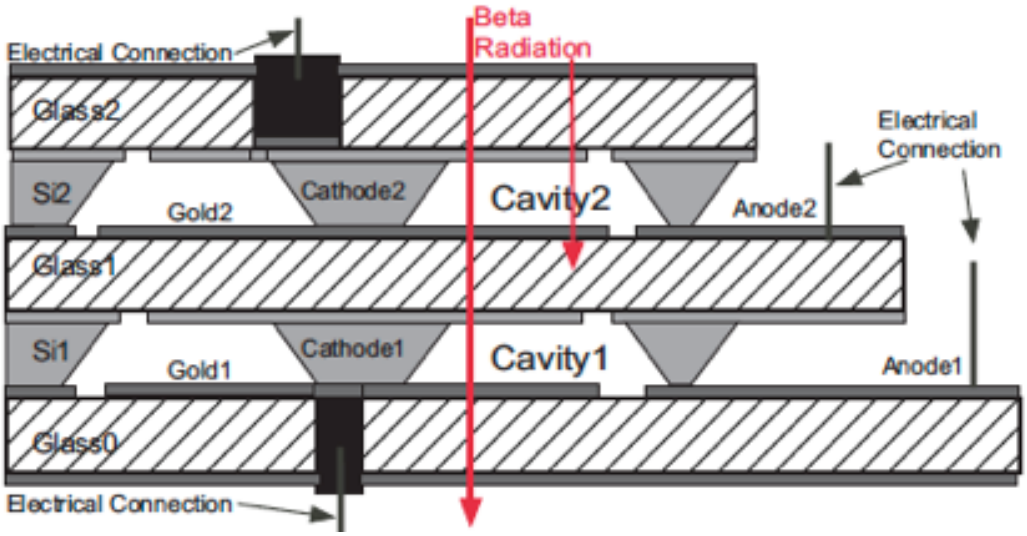

Figure 3: Microfabricated Geiger counter for beta particle detection

3.3 Moisture Detection Methods

Steam or water leaks in the primary system of pressurized and boiling water reactors have been closely monitored for structure and components integrity. Typical leak monitoring systems measure dew point temperature, accumulated condensate inside the recirculating air coolers, air temperature, water level in building sumps, and gully monitoring. Moisture or humidity detection is the primary leak detection method. Similarly, monitoring the moisture content in subsurface at a nuclear power plant can also be an effective leak detection method. The physical parameters directly related to the moisture content and sensor measurements include:

- Thermodynamic properties – pressure, temperature, and thermal conductivity of water,
- Electrical properties – conductivity and resistivity,
- Dielectric property,
- Nuclear magnetic resonance, and
- Radiation reactions – neutron absorption and gamma ray attenuation.

Many moisture detection methods have been developed and some of them being routinely used. They can be categorized based on their applications into point measurement, wide-area imaging, and remote sensing. Most sensing techniques can be used for point measurement and in-situ monitoring. Arrays of sensors are normally used to cover a wide area survey. Representative moisture sensors and sensing techniques are briefly described below.

Point Measurement Sensors:

- Tensiometer[18] – It consists of a water permeable ceramic cup and a pressure gage. The cup is placed in contact with the soil and the gage measures the soil matric potential. The meter is not affected by the osmotic potential of the soil solution. In general the response is slow and requires water refill to maintain bubble free. If the cup loses contact with the soil when for example an air pocket is created around the cup, the instrument will not function.
- Capacitive sensor[19]--The basic design of a capacitive sensor consists of a pair of electrodes forming a capacitor with the soil acting as the dielectric in between. With modern electronics, it is possible to measure very small capacitance changes in the order of 10^{-3} pF. However, the detection sensitivity is strongly depending on the gaps between sensor electrodes and soil. Different sensor geometries[20] have been examined to remedy the problem.
- Time-domain reflectometry (TDR) sensor[21] – The TDR technique is based on the measurement of the dielectric properties (or the apparent permittivity) of soil, which can be directly related to soil water content. An empirical relationship[22], Eq. 1, between the apparent dielectric constant, K_a, and volumetric soil water content, θ, was established and widely used.

$$\theta = -5.3\times10^{-2} + 2.92\times10^{-2}K_a - 5.5\times10^{-4}K_a^2 + 4.3\times10^{-6}K_a^3 \quad [1]$$

The basic design of a TDR probe is a long electromagnetic transmission line. The wave propagation velocity, V_P, is related to the speed of light, V, by $V_P = V / \sqrt{K_a}$.

- Electrical resistance sensor[22]—The basic design is simply electrodes embedded in a porous material such as a block of gypsum. The resistance or conductance between the electrodes gives the moisture measurement. It is a simple and inexpensive sensor that can be left in the field for continuous monitoring. But the sensor suffers from hysteresis and the sensor block tends to degrade over time.
- Nuclear magnetic resonance sensor[23]—It is a borehole-logging tool being developed for oil/gas exploration. It measures the density of hydrogen nuclei in soil, which can be directly related to water content. The NMR probe as marketed by Halliburton Energy Services consists of a magnetic mandrel with a diameter of 4.5 to 6 in. and an electronic cartridge. However data interpretation is more complicated because the NMR signal is the sum of the signals coming from all the pores located in the measurement volume. More development is needed.
- Neutron moisture sensor[24]-- The neutron moisture sensor consists of a fast neutron source (^{241}Am-Be, and ^{241}AmF) and a neutron detector such as ^{3}He proportional counter, ^{6}Li(Eu) scintillator, and gold-foil activation sensor. The fast neutrons propagating in soil will be slow down by hydrogen and detected by the neutron detector that is sensitive to slow neutrons. Accurate calibration may not be easily achieved. The calibration equation may be represented by a linear regression given as $\theta = bn + a$, where θ is the free water content of the soil on a volumetric basis, n is the ratio of the neutron count in the soil (C_S) to the count in water (C_W), i.e. $n = C_S/C_W$.
- Thermal conductivity sensor[25] -- Thermal conductivity measurements are based on the rate at which heat is dissipated from the heating element and are directly affected by the amount of moisture in the soil. Water is a better heat conductor than air; more heat will be dissipated as the water content increases in the soil. The sensor probe has a simple design consisting of a heating element and a K-type thermocouple. The temperature rise detected by the probe is used to estimate the moisture content.

<u>Wide Range Measurement Techniques:</u>

- Electrical resistance tomography[26] (ERT) – ERT is a technique for imaging the subsurface electrical structure using conduction current. It uses a series of electrodes that inject a low frequency electrical current into the soil and measure the resulting electrical potential distribution. A 3-D inversion algorithm is used to produce the subsurface distribution image.

The technique has been proved to be a valuable tool for imaging solute transport processes in subsurface; quantitative measurement, particularly for detecting small quantity change, remains as a challenge.

- Electrical impedance tomography (EIT) and magneto-electrical resistivity imaging technique (MERIT)[27] – EIT is an extension of ERT; it measures the phase shift between voltage and current caused by induced polarization. Water in soil usually contains various ions; migration of bulk ions in the applied electrical field results the electrical conductance and also induces a magnetic field. Thus, EIT and MERIT are methods complement one another. However, the techniques are still in research stage.
- Distributed temperature sensing[28] (DTS) – The concept is based on the fact that soil moisture influences soil thermal properties. Therefore, monitoring temperature dynamics can yield information on changes in soil moisture content. The tool to measure the DTS is a fiber-optic cable. When laser light is sent through fiber-optic cable, a small fraction of energy undergoes inelastic scattering (Raman scattering) whereby reflected light contains frequencies higher (anti-Stokes signal) and lower (Stokes signal) than the transmitted laser light. The ration of the anti-Stokes and Stokes intensities provides a measure of the cable temperature. The spatial resolution is typically 1 m for cable up to 10 km long. The precision of the temperature measurement depends on laser intensity, detector sensitivity, and integration time. 0.1 K precision can be obtained using 1 km cable with 1 m resolution and 60 s integration time.

Remote Sensing Techniques

The widely cited remote sensing technique is the ground-penetrating radar (GPR) mehtod[29], a noninvasive technique capable of efficiently investigating intermediate to large areas. A GPR uses a number of antennae operating at 100 MHz to transmit and receive electromagnetic (EM) waves propagating in the soil. The EM wave propagation velocity depends on the apparent permittivity of the soil, which is strongly dependent on water content in the soil. But determining the exact sampling depth of the ground wave can be a difficult task. Different ground survey geometries are commonly used to determine the EM velocity of the subsurface. The techniques that have been field-tested and their detection resolutions were surveyed and reported[30]. They are reproduced in the table 5.

Table 5. Overview of moisture detection techniques

Technique	Property measured	Derived information	Resolution
Neutron probe	Hydrogen concentration	Moisture content, porosity, lithology	< 10 cm
Tensiometry/Lysimetry	Matric potential and chemical analysis	Matric potential, water content, hydraulic conductivity, water sample	Point
Electrical resistivity tomography	DC electrical resistivity	Bulk resistivity	>1 m
Crosshole radar	Dielectric permittivity	Moisture distribution, lithology, soil disturbances, buried materials	5-60 cm depending on frequency
Crosshole electromagnetic induction	Electrical conductivity and dielectric permittivity	Moisture distribution, shallow contaminant plumes, lithology	1.5 to > 4.5 m
High-resolution resistivity	DC electrical resistivity	Moisture, lithology, geologic structure, buried materials, shallow contaminant plumes	> 1 m
Time-domain reflectometry	Electrical conductivity and dielectric permittivity	Flow and transport, lithology	> 2 cm depending on probe length

4.0 Conclusions

This report covers a survey on sensor and monitoring techniques for early detection of abnormal radioactive releases from the engineered facility structures, systems and components (SSCs) to the surrounding underground environment in existing nuclear power plants (NPPs). Techniques being surveyed include those involving detection of radionuclides and subsurface moisture content. Among the released radionuclides, tritium is the common tracer indicating the abnormal release from a power plant. A separate survey on tritium detection is included. The survey focuses on in-situ, real-time monitoring techniques; conventional sampling techniques are not included.

Detection of moisture content has been actively pursued. Many detection methods have been developed and applied; most techniques are still limited to point measurement. Development of wide-area monitoring methods remains as a research subject. Detecting anomalous water content and its distribution or profile in the vadose zone is a real challenge. Many difficulties need to overcome before a positive detection can be made, for example, how to discern a leak from background that itself can change rapidly.

Detection of tritium provides the ultimate confirmation of underground leaks. Development of an in-situ, fast response, and sensitive tritium sensor remains as an active research. The status of tritium detection methods can be summarized as below:

- Most are sampling techniques – Because penetration range of the tritium β-ray is < 6mm in air and < 6μm in water, non-contact type detection is nearly impossible.
- Ionization detectors apply mainly to gas-phase detection
  - High sensitivity and low background but requiring ionization chamber (may need a vacuum system).
  - Detection limit -- 0.3 nCi/L
- Scintillation detectors are routinely used for tritium detection.
  - Solid scintillation counting systems – Less efficient (<1%), detection limit around 1μCi/L, need of a high surface-to-volume ratio, requiring prefiltration of water, being vulnerable to fouling, and subject to memory effect.
  - Liquid scintillation counting systems – Detection efficiency ~ 9%, detection limit around 1nCi/L, Drawbacks are not being truly continuous, water filter fouling, production of tritiated liquid waste, and significant cost of organic solvents.
  - Plastic scintillation counting system – Detection efficiency <0.5%, subject to memory effect and free from organic scintillator waste.

To identify the radioactive releases or leaks due to failure of SSCs one must also recognize that there is a radiation background level around the power plant due to the normal releases to the environment under normal reactor operating conditions. Hence, in-situ sensor calibration is needed in order to produce a positive detection. Radiation detectors, beta or gamma, are close range detectors; arrays of such sensors are needed for mapping out the nuclide

distribution profile. To date, the effective tritium detection still relies on sampling of groundwater; in-situ monitoring is limited to point measurement.

The ideal detection methods should be in-situ, real-time, sufficiently robust to maintain their useful life over some years, and sufficiently sensitive that they provide an early signal of leaks. Furthermore, an effective sensor-array design and associated signal processing and detection algorithms need to be developed so that leaks can be positively detected and located.

5.0 Summary and Recommendations

Inadvertent release of radioactive liquids to the environment at nuclear power plants (NPPs) has occurred more frequent in recent years as nuclear power plants are aged. Detection and close monitoring of leaks at NPPs, in particular external to reactor structures, are inevitably needed to ensure the public safety. Presently, a leak is confirmed mainly by analyzing the tritium level using sampling techniques. To pinpoint the leak location remains as a time consuming and difficult task.

Nuclear and physical signatures that are directly related to structures and components leaks are (1) presence of radionuclides including tritium, cobalt-60, nickel-63, iodine-129, cesium-134, cesium-137, and strontium-90, (2) moisture level in the vadose zone, (3) underground temperature gradient caused by leaking hot water. Is there any other non-radioactive chemicals, such as boron and alkaline, which can also be used as leak signatures?

Radiation sensors that detect beta or gamma ray can be grouped into four types: (1) scintillation, (2) solid state, (3) gas-filled proportional and ionization, and (4) passive integrating. Scintillation and gas-filled sensors are well developed and employed. However they, like the other sensors, are limited to point measurement. One emerging technique that may cover a large area is the plastic scintillator. Overall, radiation detectors, particularly for tritium detection, are contact sensors because the penetration range of the tritium beta ray is only 6μm in water. All sensors require calibration to correct the background uncertainty.

Moisture and temperature sensors measure the changes of the physical properties including thermodynamic, electrical, nuclear magnetic properties. A great number of moisture detectors have been developed for geophysical survey and they can be directly applied to detection of water leaks at NPPs. Some of the techniques such as electrical resistance tomography, electrical impedance tomography, and magneto-electrical resistivity imaging technique have been developed for 2-D and 3-D imaging. However, all the techniques including radiation techniques using neutron and gamma sources suffer from the baseline uncertainty and lack of a reliable model to predict the underground heterogeneity and porosity.

In conclusion, a real-time in-situ subsurface leak detection and monitoring system does not exist. More sensor research and development (R&D) are needed. The following R&D are recommended:

1. Radiation sensor R&D – (a) Advance solid-state sensing techniques and array-sensor design, (b) Plastic scintillation sensor for wide area coverage, (c) Fiber-optic radiation sensor and (d) microfabricated beta-particle sensor.

2. Moisture and temperature sensor R&D – (a) Promising 2-D and 3-D imaging techniques coupled with in-situ water pathway calibration techniques such as tracer methods, (b) Sensor fusion approach that can simultaneously measure moisture, temperature, and tritium, and (c) Wide area leak detection system involving array moisture sensor design using the underground piping as an electrode.
3. Other sensor R&D – (a) Remote sensing such as use of radar or millimeter-wave to detect radiation induced plasma and (b) Acoustic/ultrasonic techniques to detect leak induced flow noise.
4. A multiple sensor system including acoustic, moisture, temperature, and radiation sensors.

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