CHAPTER: 2

INSTRUMENTS, MATERIALS AND TECHNIQUES

This chapter includes the details about the instruments, materials and experimental techniques used in research work to assess the indoor radon/thoron and their progeny concentration, attached/unattached fractions, radon/thoron exhalation rates in soil and radon concentration in water. The details about the construction, working principle of these devices has been discussed below.

2.1 SINGLE ENTRY PINHOLE DOSIMETER

For measurement of indoor radon/thoron gas concentration, single entry pinhole dosimeter indigenously instrument developed by BARC, Mumbai, India has been used. It consists of two chambers named as 'radon + thoron compartment' while the other chamber is known as 'radon compartment' (Figure 2.1). Each chamber has a length of 4.1 cm and a radius of 3.1 cm. These two chambers are segregated by a barrier or discriminating plate which is known as radon-thoron discriminating plate and has 4 pinholes with the diameter of each hole as 1mm. Both compartments are internally coated with metallic powder (nickel) to attain neutral electric field inside the compartments to ensure deposition of progeny formed by radon and thoron gases uniformly throughout the chamber. The minimum detection limit of single entry pinhole dosimeter is approximately 4 and 6 Bqm^{-3} for radon and thoron respectively, considering 4 $trackcm^{-2}$ as background tracks in LR-115 (Sapra et al., 2016).

The gas enters the 'radon + thoron chamber' through a glass fibre filter paper which is placed at the entry face. The radon and thoron gas enter first chamber and their progeny are filtered out by glass fibre filter paper. Only radon gas will diffuse to second chamber as thoron gas is cut-off by pinholes of discriminating plate so that only radon gas can reach to second compartment. The LR-115 Type-II films of size 3 cm × 3 cm were placed in first compartment, registers alpha tracks arising due to radon, thoron gases and the film placed in second compartment registers alpha tracks solely due to radon.

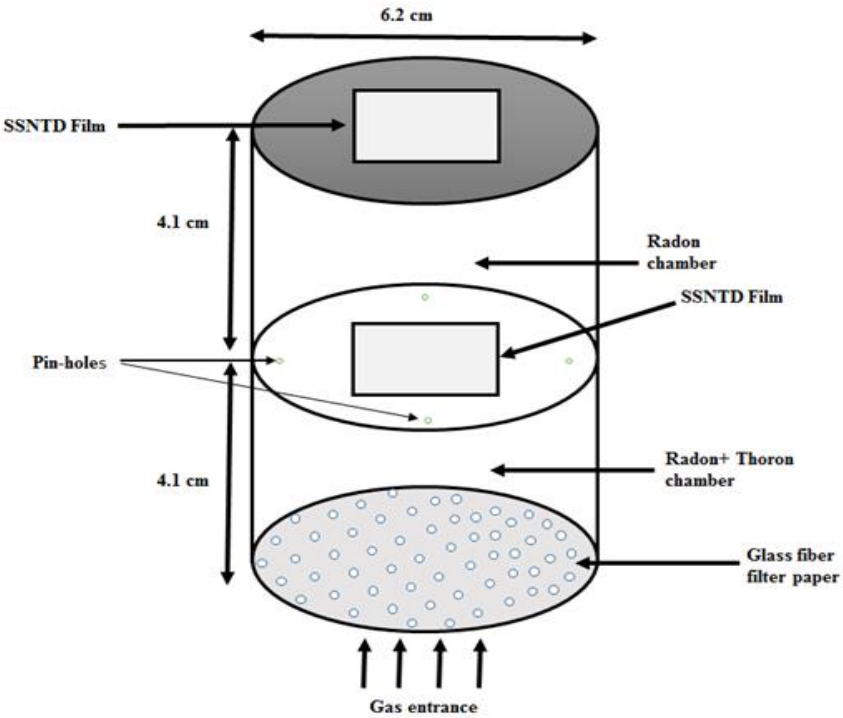


Figure 2.1: Single entry pin hole dosimeter

2.2 DEPOSITION BASED PROGENY SENSORS (DRPS/DTPS)

The deposition based direct radon and thoron progeny sensors (DRPS and DTPS) (Figure 2.2 A & B) has been used to estimate the equilibrium equivalent concentration of radon and thoron progeny in the dwellings. DRPS/DTPS have been developed by BARC, Mumbai, India, for estimating the time integrated progeny deposition fluxes in the environment (Mishra and Mayya, 2008). These are made up of solid state nuclear track detectors (LR-115) mounted with absorbers of appropriate thickness to degrade the energy of alpha particles emitted by progeny below 4 MeV for DRPS and DTPS separately. DTPS element is made up of detector LR-115 (2.5 cm × 2.5 cm) films mounted with 50 µm aluminized mylar as an absorber while the DRPS has an absorber combination comprising of aluminized mylar with 25 µm thickness and cellulose nitrate with thickness of 12 µm. The advantage of this sensor is that there is no interference from gas and thus it gives a direct estimation of the airborne concentration of the alpha- emitter progeny (Mishra et al., 2008; Singh et al., 2015). The minimum detection limit is 0.1 Bqm^{-3} for DTPS and 1.0 Bqm^{-3} for DRPS respectively (Mishra et al., 2014).

The basic principle of operation of these sensors is that the LR-115 detector detects the alpha particles emitted from the deposited progeny atoms in the range of 1.4 to 4.8 MeV. In DTPS, LR-115 type-II films selectively detect only 8.78 MeV α-particles emitted from Po-214; while DRPS detect mainly 7.67 MeV α-particles from Po-214. The tracks recorded in the exposed LR-115 film is related to equilibrium equivalent progeny concentration (EEC) using the sensitivity factor. The sensitivity factor is expressed as track density registered for one day exposure to an environment containing 1 Bqm^{-3} of EERC or EETC.

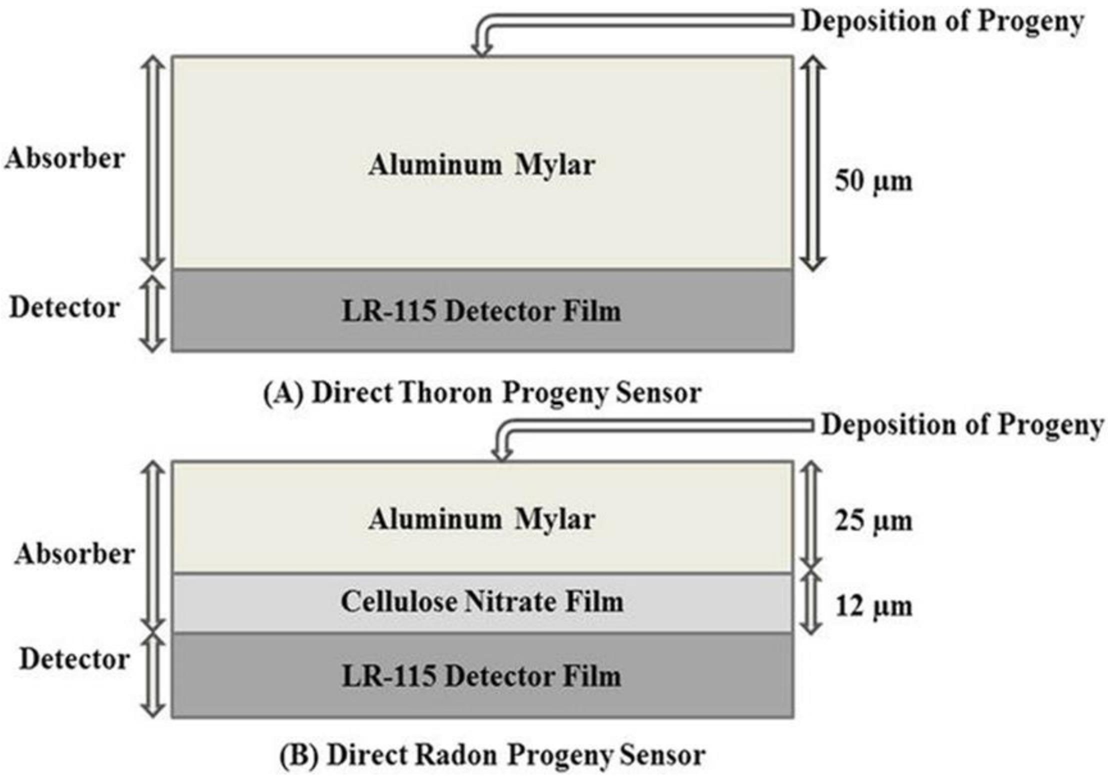


Figure 2.2: (A) Direct thoron progeny sensors (DTPS) and (B) Direct radon progeny sensors (DRPS)

2.3 WIREMESH DIRECT RADON/THORON PROGENY SENSORS (DRPS/DTPS)

Wire-mesh capped deposition progeny sensors for radon and thoron has been used to measure the attached/unattached fractions of radon/thoron in the dwellings. These sensors are the combination of DTPS/DRPS and a screen of 200 mesh- type wire screen (79 mesh cm^{-1}, wire diameter; 0.005 cm) (Mayya et al., 2010). The distance between the wire mesh and DTPS/DRPS is 1 cm. The minimum detection limit for wiremesh capped DTPS is 0.3 Bqm^{-3} and for wiremesh capped DRPS is 3 Bqm^{-3} (Mishra et al., 2014).

Attached/unattached fractions available in the air when these pass through the wiremesh. These sensors help in separating the fine fractions, as the fractions of unattached progeny atoms gets trapped into the screen of mesh. Hence, only deposition of attached or coarse fractions of progeny takes place on the nuclear track detector inside progeny sensor (Figure 2.3). The LR-115 time integrated passive nuclear track detector, mounted with an absorbers of thickness 50 μm aluminized mylar has been used for thoron progeny which detects only 8.78 MeV alpha particles which are emitted from Po-212. LR-115 with an absorber of thickness 37 μm (25 μm aluminized mylar and 12 μm cellulose nitrate of LR-115) has been used for radon progeny which detects mainly 7.67 MeV alpha particles which are emitted from Po-214 (Sapra et al., 2016; Mehra et al., 2016). The registered alpha particles on detector will be the measure of attached fraction of progeny activity concentration.

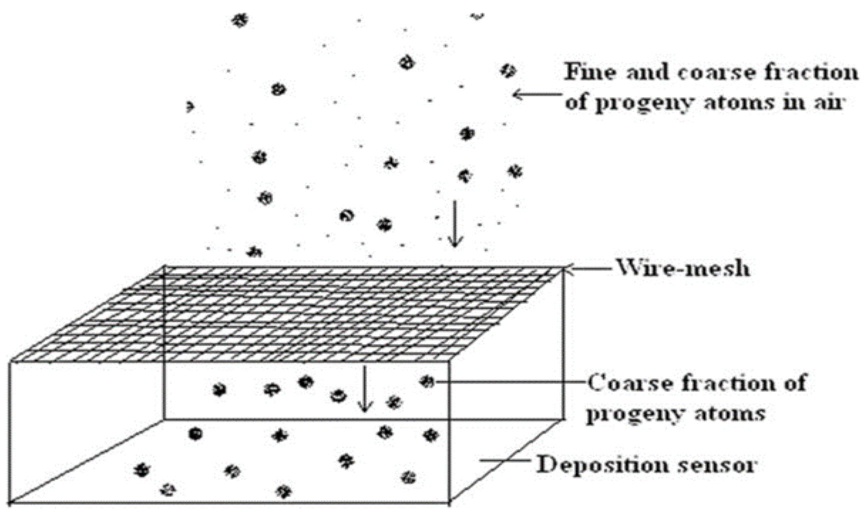


Figure 2.3: Coarse (attached) and fine (unattached) fractions of radon and thoron in wire-mesh capped DRPS/DTPS (Sapra et al., 2016)

2.4 LR-115 TYPE -II FILMS (SOLID STATE NUCLEAR TRACK DETECTOR)

Solid state nuclear track detectors (SSNTDs) are insulating materials which can be found in nature or may be synthesised in lab (Iyer et al., 1972; Fleischer et al., 1975).

Inorganic crystals, glassware, and polymers are among the materials used to make these detectors. The low-energy alpha particles released by radon and certain radon offspring are recorded by solid track detectors such as allyl diglycol carbonate (CR-39), bisphenol-A polycarbonate (Lexan, Makrofol), and cellulose nitrate (LR-115, CN 85). As a result, such solids are suitable for use as radon integrating detectors. These are especially well suited for determining long-term average radon levels in a simple, durable, and dependable method. Because of their simplicity, good geometry, low cost, and ability to obtain an integral signal after long-term measurements, nuclear track detectors are widely used in a variety of scientific areas like searching for super-heavy materials, magnet monopoles, high and low energy nuclear physics, geology, and dosimetry and radon measurement etc. (Banjanac et al., 2006).

In the present study, the cellulose nitrate (CN) plastic detector, commercially known as LR-115 Type-II with chemical composition $C_6H_8O_9N_2$, manufactured by Kodak Pathé, France, has been used to detect the alpha particles generated by radon, thoron, and their progeny (Figure 2.4). The LR-115 Type II plastic track detector is used to detect alpha particles with energy ranging from 1.7 to 4.8 MeV. (Abu-Jarad et al., 1980; Jonson, 1981). As a result, any extra alpha particles emitted by radon daughters will not be detected on the surface of the LR-115 Type II plastic track detector since their alpha energies are greater than the detector's upper threshold energy (6.0 and 7.68 MeV from 218Po and 214Po, respectively). The advantages of LR-115 Type II detectors are unaffected by humidity, low temperature, mild heating, or light (Durrani and Ili, 1997). LR-115 films are unaffected by electrons and radiations in the electromagnetic spectrum (such as gamma rays, X-rays, ultraviolet or infra-red rays).

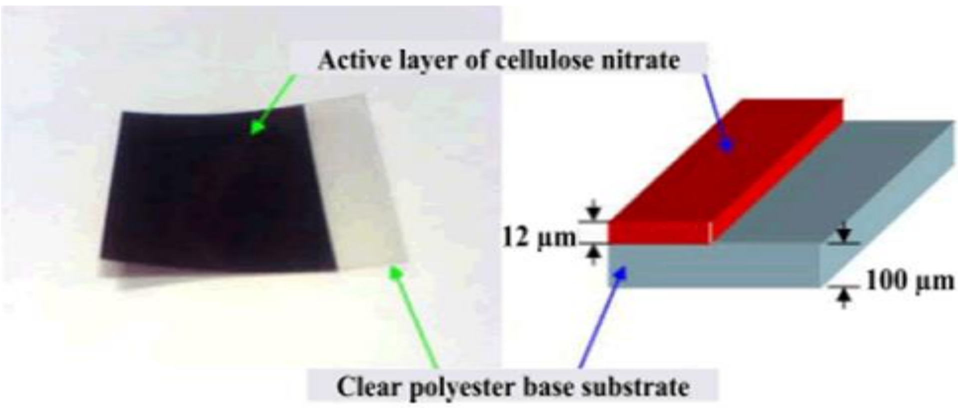


Figure 2.4: LR-115 type-II film

Alpha particles emitted from radon/thoron and their progeny are detected by LR-115 type-II films. When the energetic alpha particles pass through such insulating materials (LR-115 type-II), then tracks are formed. The energy of these alpha particles transfer to the medium and electrons leaves a narrow trail of damage along its route, ranging from 1 to 10 nm in diameter. Energy transmission is influenced by the type of particle, its mass, energy, density, and the composition of the detector material. "Latent tracks" appear in the form of holes as shown in the Figure 2.5 and are also termed as tracks. Because of their small size, latent tracks are not visible optically but can only be viewed with transmission electron microscopes or equivalents at extremely high magnification.

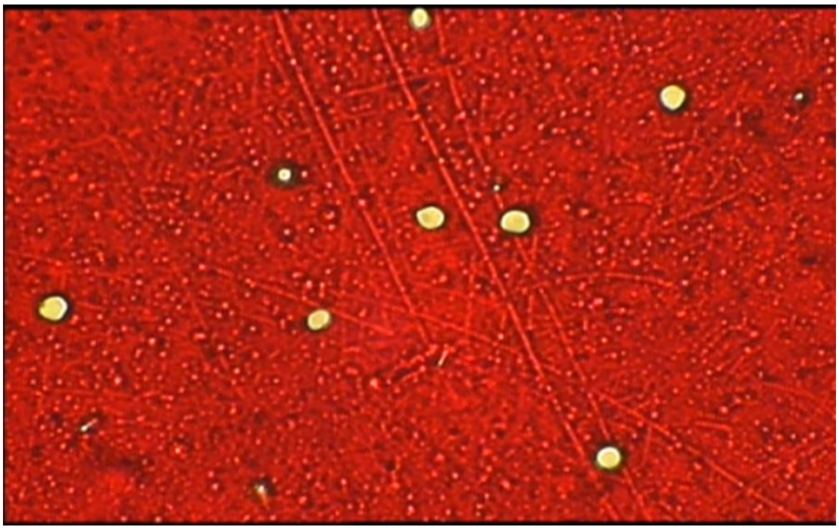


Figure 2.5: Tracks formed on LR-115 type-II films

2.5 CONSTANT TEMPERATURE BATH UNIT

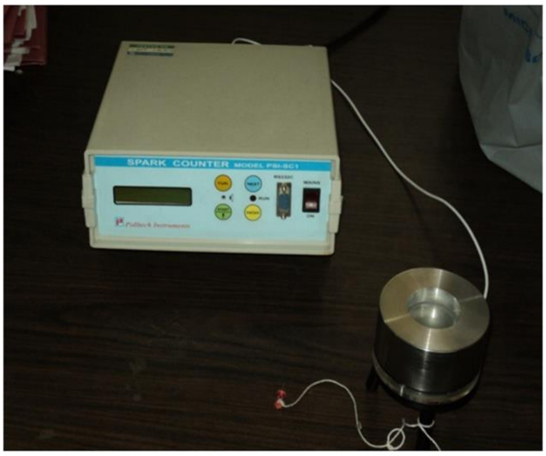
Tracks formed by alpha particles on LR-115 films are normally imperceptible to the naked eye. Hence chemical etching is done to make these tracks visible under optical microscope or to count with spark counter. Constant temperature bath unit (model: PSI-CTB1, manufactured by Polltech Pvt. Ltd., India) to precisely adjust the temperature of the etchant solution has been used in the present study for chemical etching (Figure 2.6). Several chemical etchants like NaOH, KOH are routinely used, depending on the etching conditions for different detectors, particle sensitivity and the critical angle of etching.

Figure 2.6: Constant temperature bath unit in Radiation Physics lab, department of Physics, MRSPTU, Bathinda

Before etching the retrieved LR-115 films, the bulk etch rate of the etching bath was standardised using unused films. The 2.5 N NaOH solutions were made at room temperature, and retrieved films were placed into the etching bath unit with cartridges and etched at 60°C for 90 minutes (Eppan and Mayya, 2004; Sahoo et al., 2013). After etching the films were cleaned using distilled water and then left to dry for further counting of tracks.

2.6 SPARK COUNTER

A spark counter (Model PSI–SC 1) a simple, accurate, and inexpensive piece of equipment (Figure 2.7a) has been used to count alpha tracks (holes) developed on the LR-115 type-II films. It counts the number of holes created on the LR-115 SSNTD films as a result of an etching process done after the film was exposed to radon/thoron and their progeny.



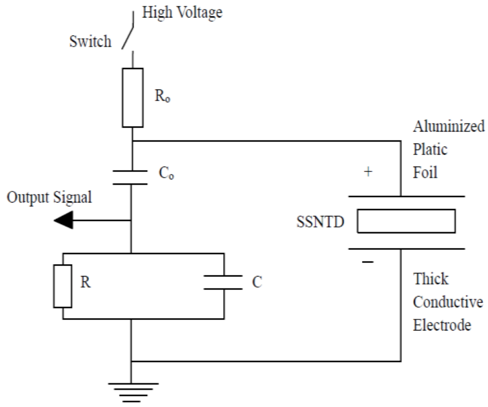


Figure 2.7: a) Spark counter. b) Schematic diagram of spark counter.

In the spark counter, a capacitor is formed by sandwiching a thin etched track detector (about 8-10 µm thick) between two electrodes which is generally made up of brass or stainless steel. Aluminized mylar is placed in such a way that shining or conducting face encounters the detector (etched LR-115 film). A heavyweight is placed on top to ensure good contact between the electrodes, the detector, and the aluminized film. When a high voltage is applied across the capacitor C (Figure 2.7b), an electrical discharge on spark occurs through a track-hole. A simple voltage pulse produced across a load can be counted using an electronic counter.

A spark passing through a track hole has enough energy to vaporise the tiny layer of aluminium coating (less than 1m thick) and generate a much larger hole in the aluminium electrode. After the short-circuit, the capacitor C is charged again, but a second spark in the same track hole is impossible due to the evaporation of the aluminium in the electrode. As a result, until all track holes have been counted, the spark moves from one track hole to the next at random. The number of sparks in the plastic track detector, and thus the number of track holes, is equal to the evaporated spots on the metal, which have a diameter of roughly 100 µm.

A spark counter's optimum operating voltage, identical to that of a Geiger tube, lies in the middle of the plateau curve (Malik and durrani, 1974). The operating applied voltage for track counting is typically between 400 and 600 V. A higher voltage must always be given to the track detector to punch out any tracks that have not been completely etched through.

2.7 SMART RnDuo (A PORTABLE MONITOR)

Smart RnDuo, a scintillation-based detector has been used to measure the radon concentration in water, radon/thoron exhalation rates in soil. It works on the principle of detecting alpha particles that strike the detector and cause scintillations inside the Lucas cell due to the ZnS:Ag coating. The Photo Multiplier Tube counts the scintillations and converts them to radon/thoron activity using an integrated algorithm (Figure 2.8). In a 0.5 m^3 calibration chamber at Bhabha Atomic Research Centre (BARC), Mumbai, India, the instrument is calibrated against standard radon-thoron sources (Model RN-1025 & TH-1025) acquired from Pylon Electronics Inc., Ottawa, Canada (Sahoo et al., 2013). The relative humidity is controlled between 10% and 99 percent, while the temperature is controlled between 20°C and 50°C (Jobbagy et al., 2017). The advantage of a portable monitor is that it is unaffected by humidity or the presence of traces of various gases in samples. With alpha detection proficiency of the scintillation, the instrument possesses radon/thoron sensitivity of 1.2 CPH $(Bqm^{-3})^{-1}$ and 0.8 CPH $(Bqm^{-3})^{-1}$, respectively (Gaware et al., 2011).

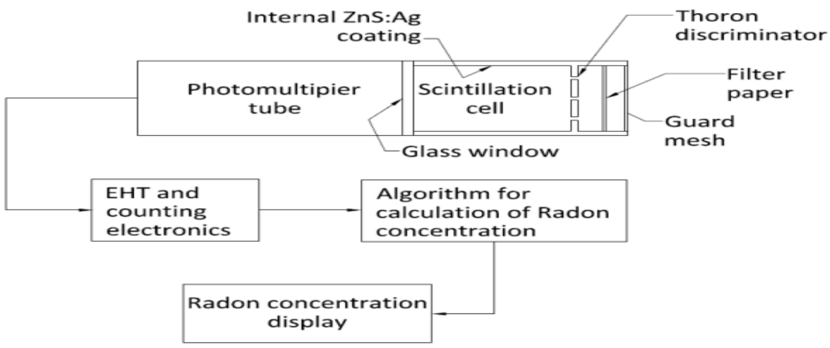


Figure 2.8: Schematic diagram of Smart Rnduo (a portable monitor) (Sapra et al., 2016)

2.7.1 Protocol for Measurement of Radon in Water

Smart RnDuo, a scintillation based detector, developed by BARC, Mumbai, India, has been used to measure the activity concentration of radon in water. Radon measurement in collected water samples was carried out in-situ, keeping the delay between sampling and counting of radon to minimum so as to avoid loss of radon due to radioactive decay. The measurement of radon in water sample was carried out using SMART RnDuo, radon monitor (AQTEK System, India) and following measurement protocol prescribed by BARC Radon Handbook (Sapra et al., 2016). The SMART RnDuo is ZnS:Ag scintillation cell based continuous automatic radon monitor. The scintillation cell based technology makes the detector immune to the effect of humidity on measurement, a very important parameter for measuring radon in water, and gives it a higher sensitivity as compared to other commercial radon monitors. The measurement setup consisted of a water bubbler fitted to sample bottle and attached to SMART RnDuo monitor in a sealed closed loop arrangement. Gas was collected from the water sample into a scintillation cell (150 cc) for radon measurement using a diffusion method. The gas is passed via a "progeny filter" and a "thoron discriminator" during diffusive sampling, which removes radon/thoron progenies and thoron. The radon measurements in RnDuo are based on the continuous counting of alpha particles generated from radon and its decay products formed inside a cell volume by the PMT and the associated counting electronics. The collected alpha counts are processed by a microprocessor unit using a devised algorithm to display the radon concentration. The schematic diagram of the same is shown in Figure 2.9.

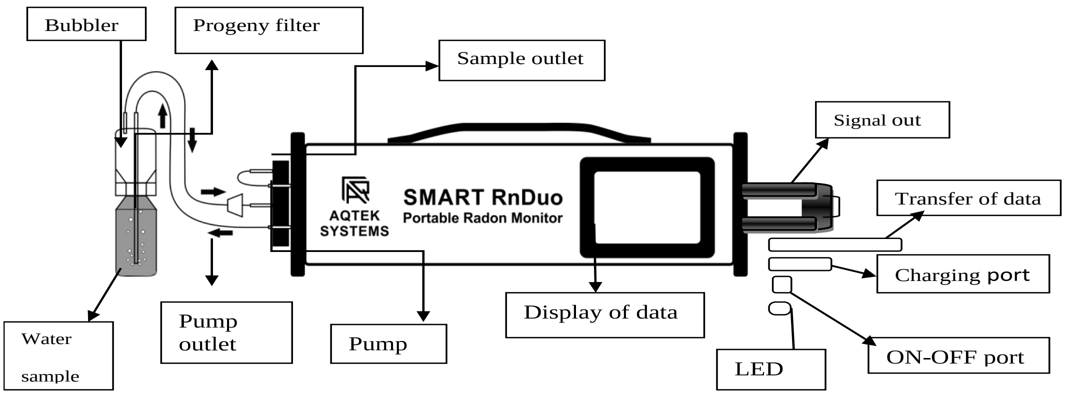


Figure 2.9: Set-up for measurement of radon in water sample (Sapra et al., 2016)

After connecting the setup, the internal pump of the monitor was turned on for 5 min so that the bubbling of air through water sample removed the dissolved radon and transferred it to the detection volume. The pump was then turned off and a further delay of 5 min was provided so that the thoron gas ($t_{1/2}$ = 55.6s), if present in the volume, decayed out completely. The radon in the air was then counted continuously in 15 min measurement cycle. The average of three successive measurements was noted as C_{air} (Bqm^{-3}).

2.7.2 Protocol for Measurement of Radon Mass Exhalation Rates in Soil

Smart RnDuo has been used for the quantification of radon mass exhalation rates in soil samples. The radon mass exhalation rate is defined as the activity concentration of radon per unit time from per unit mass of soil matrix and is a simple approach to calculate radon flux density. For the estimation of radon mass exhalation, Smart RnDuo is set to diffusion mode. As illustrated in Figure 2.10, each soil sample was placed in an accumulation chamber that was connected to a detector. The gas was collected from the sample into a 153-cc scintillation chamber, where it was passed through a "progeny filter" and "thoron discriminator" to remove radon/thoron progeny and thoron. The thoron discriminator's "diffusion time delay" prevents the thoron from entering the detector. The alpha particles released by accumulated radon and its progeny inside the detector were used to quantify radon activity. The material was continually analysed for 12 hours, with each cycle lasting 60 minutes, as per the protocol. The detector's build-up data was recovered and used to calculate the sample's radon mass exhalation rate using conventional equations.

Figure 2.10: Setup for radon mass exhalation rates measurement (diffusion mode) in soil samples

2.7.3 Protocol for Measurement of Thoron Surface Exhalation Rates in Soil

Smart RnDuo has been used for the measurement of thoron surface exhalation rates in soil samples. Because thoron has a short diffusion length in the environment, it is not evenly distributed in the accumulator. As a result, the detector was employed in "flow mode" with an inbuilt pump with a flow capacity of 0.7 $Lmin^{-1}$ to estimate thoron surface exhalation rate in soil samples (Figure 2.11). The air containing thoron gas (in the chamber) is circulated in a closed loop into the lucas cell by an inbuilt pump, which first goes through a "progeny filter." The alpha particles released by accumulating thoron and its progeny inside the detector were used to measure thoron activity. The equilibrium value of thoron concentration (C_t) in the accumulation chamber was measured over the course of one hour, with four 15-minute cycles. In a 15-minute thoron mode cycle, the sampling pump is maintained on for the first 5 minutes, giving a measure of thoron and background, followed by a 5-minute delay to ensure thoron decay, and finally 5 minutes of counting delivers the measure of background counts for that cycle. The thoron surface exhalation rates were calculated using the measured value of C_t.

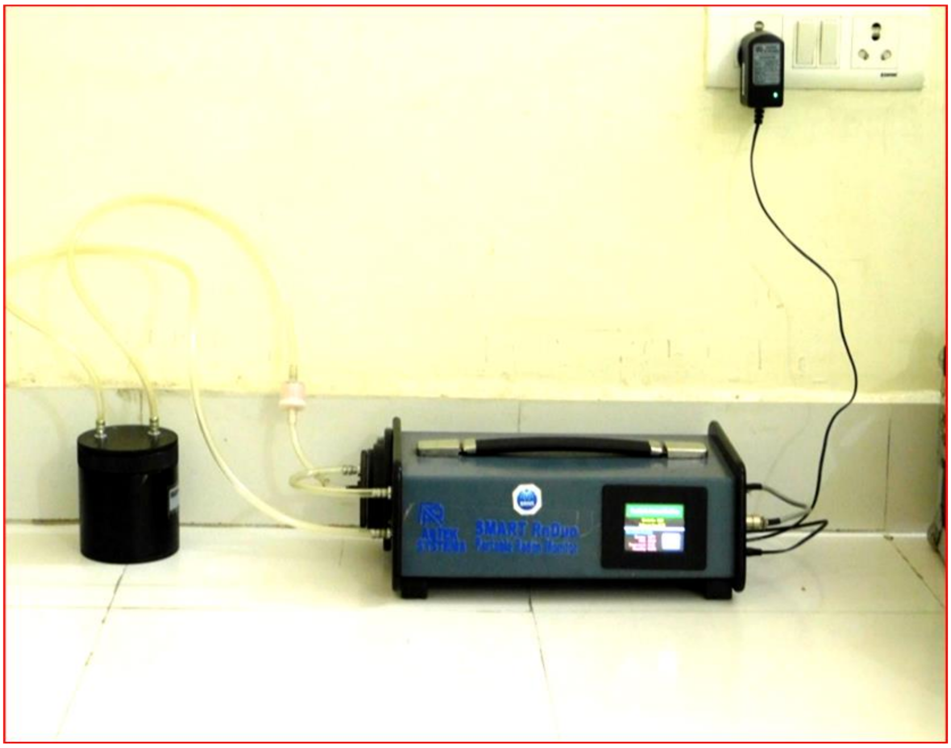


Figure 2.11: Setup for thoron surface exhalation rates measurement (flow mode) in soil samples

2.8 GAMMA SURVEY METER

Preliminary survey of the studied area was done using gamma survey meter (Polimaster PM-1405, Republic of Belarus) before the deployment of dosimeters as per the protocol of research project (Figure 2.12). The radon/thoron level in an environment depends upon several factors such as geology of the location, environmental conditions, type of houses, type of building materials used in the house. Range of gamma radiation levels across the villages is required to categorize the villages falling into various gamma zones. The gamma dose is mainly due to gamma rays emitted from daughter products of Radon (214Pb, 21-Bi, &210Pb) and Thoron (212Pb, 212Bi &208Pb) present in the soil matrix.

Therefore the survey has been done at a one-meter height from the earth's surface as it is not affected by airborne decay products. The Survey meter incorporates large energy compensated Geiger Muller tube for precise measurement of the ambient equivalent dose rate of the gamma radiation in the range from background level to 100 $mSvh^{-1}$ (10 Rh^{-1}). The Polimaster (PM1405) has a gamma energy response from 0.05 to 3 MeV and can be used for dose rate measurement vary from 0.01 $\mu Sv\ h^{-1}$ to 130 $mSvh^{-1}$ suggesting suitability for environmental gamma survey. It has a calibration accuracy of ± (20 +1/H) % where H is the dose rate in $\mu Sv\ h^{-1}$.

Figure 2.12: Gamma Survey Meter