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APPENDICES

APPENDIX A

Particle Sizing Method

There are various methods used in particle sizing. The most widely used techniques are discussed below:

A-1 Techniques for Measuring Natural Radioactive Aerosol

Sieves: Sieve is an old fashioned, but cheap and readily usable technique for large particles such as those found in mining and some food processing applications. It allows separation into some size bands if required. Using this technique it is not possible to measure sprays or emulsions and dry powders under 38mm.The longer the measurement times the smaller the answer, as particles orient themselves to fall through the sieve. A true weight distribution is not produced as the method relies on measuring the second smallest dimension of the particle. It is a low-resolution method.

Sedimentation: In Sedimentation, the speed of particles falling in a suspension liquid is determined with the help of light or X-ray beams. The particle size is then expressed in terms of the equivalent spheres which have the same settling speed in laminar flow conditions. Centrifuges are sometimes used to accelerate sedimentation process. This technique has a resolution that is many times that of Dynamic Light Scattering (DLS) and also has excellent reproducibility. But sedimentation technique has a limited range, with particular difficulties below 2 mm and above 50 mm. Moreover this method depends on the ambient temperatures that affect viscosity. Other disadvantages include slowness of measurement, which makes repeat measurements tedious. Irregularly shaped particles, such as disc-shaped, take even longer to settle due to their increased drag compared with spherical particles.

Electrozone Sensing: Electrozone sensing technique was originally developed for sizing blood cells. It is difficult to measure emulsions and impossible to measure sprays. This method is slow for materials of relatively wide particle size and it is not easy to measure particles below 2 mm. Optical and Electron Microscopy is an excellent technique that allows direct examination of the particles, and one that is relatively cheap. However, it is not suitable for measuring airborne particles. A quick look with a microscope applying this technique often gives a great deal of information that other methods are unable to give .The method inspires great confidence in the results. However, the number of particles measured applying this technique is usually small compared to other particle sizing methods, so representative sampling becomes critical. And also the sample preparation for electron microscopes is slow, expensive, and requires considerable technical expertise.

Microscopy: This is an excellent technique that allows direct examination of the particles in question, and one that is relatively cheap. However, it is not suitable as a precise measurement technique beyond the level of simple judgment. Also, as relatively few a particles are examined, there is a real danger of unrepresentative sampling and if weight distribution is measured results are magnified. Missing one 10 mm particle has the same effect as missing one thousand 1mm particles. Sample preparation for electron microscopy is laborious and slow, and for manual methods fewer particles are examined.

Laser Diffraction: Laser diffraction method utilizes a laser beam (a source of coherent, intense light of a fixed wavelength) that passes through a sample of particles, and light intensity data is collected at different (low) scattering angles away from the axis of the laser beam. The method relies on the fact that diffraction angle is inversely proportional to particle size. Laser diffraction method can measure dry powders and liquid suspensions and emulsions, as well as sprays and aerosols. It is very fast in data collection with proper sample preparations, high reproducibility and precision; it is relatively simple to use. But this method is based on the assumption of sphericity of the measured particles. The laser diffraction method is precise but for particles with low aspect ratio, the results are inherently non-accurate, regardless of the definition of a "true" particle size. The measurement depends on the optical parameters (refractive index, light absorption) used to calculate the scattering properties of the particles. Light absorption characteristics are often unknown and must be "estimated". In addition, no multiple, or subsequent, scattering is considered. Resolution is limited by the number of detectors. The larger the measured particles, the lower the resolution of laser diffraction systems are.

Dynamic Light Scattering (DLS): Dynamic Light Scattering (DLS) or Photon Correlation Spectroscopy (PCS) technique monitors the doppler shifts in reflected laser light created by the Brownian motion of submicron fine particles. A minimum amount of information about the sample is needed to run an analysis. But this measuring technique has extremely low resolution; The method does not really provide much "size distribution" data, only a mean size and estimate of standard deviation.

Time of Flight Mass Spectrometry (TOFMS): Time of Flight Mass Spectrometry (TOFMS) technique works with dry powders. It has broad total measurement range, -0.2 to 700 μm. The technique also has fast analysis time, normally about 1 minute. Although by this technique, liquid suspensions of particles may be difficult or impossible to measure. Particles <0.2 μm can't be measured; measurements below 0.5 μm will likely be low resolution. High resolution analysis is not possible due to physical limitations of the method.

Focused-Beam Reflectance Measurement (FBRM): Focused-Beam Reflectance Measurement (FBRM) is a particle size determination technique based on a laser beam focusing in the vicinity of a probe sapphire window. The beam follows a circular path at speeds of up to 6 m s-1. When it intersects with the edge of a particle passing by a window surface, an optical collector records a backscatter signal. The time interval of the signal multiplied by the beam speed represents a chord length between two points on the edge of a particle. The chord length distribution (CLD) can be recalculated to represent either a number or volume weighted particle size distribution. And this method provides on-line and in situ information about the chord length distribution (CLD) of a population of particles in dispersion. But a measured CLD does not directly represent a particle size distribution (PSD). Conversion of CLD to PSD is not straightforward and requires sophisticated mathematical software that is not easy to validate.

Laser Obscuration (Time-of-Transition): Laser Obscuration (Time-of- Transition), the Ankersmid systems are based on well-founded principles of the Laser Obscuration, or Time of Transition (TOT) theory. A sample containing moving or stationary particles is scanned with a focused He-Ne laser beam, rotating with a constant frequency by a wedge prism. Since the angular velocity is known, the size of each individual particle can be calculated from the duration and form of the beam obscuration signal. This has got a very fast data collection with proper sample preparations, very high reproducibility. The TOT method refers directly to particle size rather than to any secondary properties. This eliminates inconsistencies due to refractive index, density and viscosity variations, Brownian motion, thermal convection and other physical phenomena. But measurement minimum of 0.5 μm. Need to change lenses (once) to cover the whole distribution from 0.5 to 3600 μm.

Inertial Impactors: Inertial impactors are devices widely used for the sampling and size selective collection of aerosol particles. Their principal of operation is simple: an aerosol stream passes through a nozzle and impinges upon a collection plate. Particles in the aerosol stream having large enough inertia will impact upon the collection plate while the other particles will follow the airflow out of the impaction region. There are two classes of inertial impactors: single stage impactors and multiple stage impactors (also called cascade impactors). The cascade impactors are commonly used to determine the aerosol size distribution. They have sharp cut-point characteristics, low internal particle holdup, and are easy to operate. Particles can be collected on substrates for microscopic analysis or additional measurements of mass or composition. They are designed for applications where wide size-range coverage and fast response are required.

APPENDIX B

Impactors in Particle Size Distribution

Impactors are simple devices, consisting of air flowing around a body or an impaction plate. Particles with sufficient inertia will slip across the air streamlines and impact on the impaction surface. Particles with less inertia will not slip across the streamlines sufficiently to strike the surface and will instead follow the airflow from the impaction area. In the impactor air is drawn through a series of orifices of decreasing size; the air flow is normal to collecting surfaces on which aerosols are collected by inertial impaction. The particles, separated stepwise by their momentum differences into a number of size ranges, are collected simultaneously.

B-1 Constructional Details

Typically, cascade impactors are consists of a number of impactor stages connected in series. Each stage of a cascade impactor is made up of a number of nozzles and an impaction plate arranged perpendicular to the flow of sampled air stream as shown in Figure B-1.

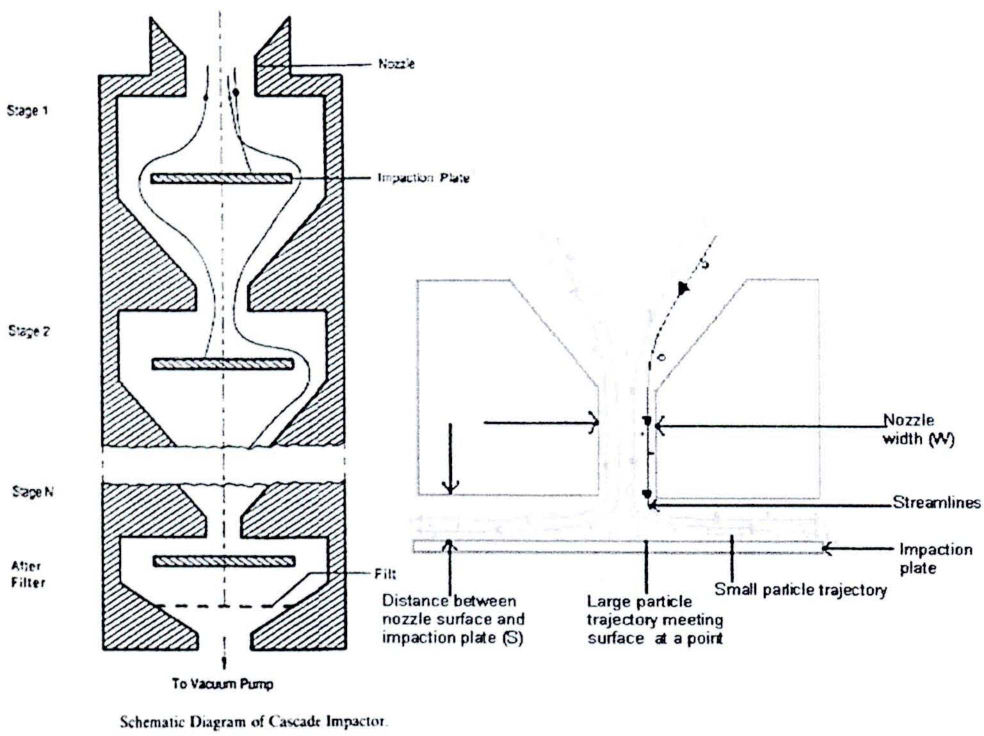

Figure B-1 Schematic projection of one single stage within impactor, showing divergent streamlines in flow approaching collection surface.

Nozzle plate of each stage of the impactor comprises of circular shaped nozzles of diameter W and the flat circular impaction plate is placed at a fixed distance S from the nozzle plate. In each stage an aerosol stream passes through the nozzle and impinges upon the plate. Particles in the aerosol stream having a large enough inertia will impact upon the plate, and smaller particles will pass as an aerosol onto the next stage. The magnitude of this differing inertia reflects the resistance to a change in direction of the laminar flow streamlines [45, 46]. As the incoming flow passes through the nozzle plate, the impaction plate deflects the flow to form an abrupt 90° bend in the streamlines.

By designing each successive stage with higher aerosol velocities in the nozzle, smaller diameter particles will be collected at each stage. Particles too small to be collected in the last stage are generally collected on an after-filter. The particulate mass deposited on each stage is determined using well-established techniques such as gravimetric or atomic absorption spectrometry (AAS) or by any other radiation detector. For associated radioactivity, the deposits are analyzed for alpha or beta–gamma activity.

B-2 Principle of Impactor

The theory of impactors has been well developed by Marple and colleagues over several years, based on a two-dimensional solution of the Navier-Stokes equations defining the gas flow field in the absence of particles, and then using Newton's equation of motion to model the passage of different sized particles through various stage geometries [62, 63, 74].

Good understanding of the design basis has been achieved on the subject of impactors since they were first introduced by May [75]. Computer-based analysis for prediction of fluid flow and particle trajectories has enabled the development of design guidelines. These have helped in enabling the impactors to operate according to specific requirements with sharp cut-off characteristics.

An impactor is composed of a nozzle plate bearing N number of nozzles, each of diameterW , an impaction plate and a stage wall. Among these, the nozzle plate plays the most important role in determining the particle collection efficiency. Impactors that have a 'sharp cut-off' collection efficiency curve approach the ideal step-function in which all particles greater than a certain aerodynamic size are collected and all particles less than that size pass through. This size is called the cut-off diameter of the stage. The response of an impactor is characterized by the dimensionless parameter, the Stokes number S_{tk}, defined as:

$$S_{tk} = \frac{\rho_p V_0 C D_p^2 18\mu}{W/2} \qquad \text{(B-1)}$$

where, ρ_p is the density of particles, V_0 the flow velocity through the nozzle, D_p the diameter of particles, μ the dynamic viscosity of the fluid medium, W the nozzle diameter and

The Cunningham slip correction factor C, given by the following expression:

$$C = 1 + \frac{0.163}{D_p P} + \left[\frac{0.054}{D_p P} \times e^{(-6.66 D_p P)}\right] \qquad \text{(B-2)}$$

where, P is the absolute atmospheric pressure (atm) upstream of the nozzle and Dp is specified in units of mm. The quantity of interest while designing an impactor is the S_{tk50}, which is the Stokes number that gives 50% collection efficiency. This is equivalent to assuming that the mass of particles larger than the cut-off size that get through equals that of the particles below the cut-off size that are collected.

The standard value for S_{tk50} calculated for a circular jet impactor is 0.25, while that for a rectangular jet is 0.59. As it is more convenient to make circular nozzles with precision, it was decided to develop the impactor with circular jets and hence all further design parameters were estimated using the value of 0.25 for S_{tk50}.

The particle collection efficiency (E) of an ideal impactor stage, expressed as a percentage, will increase in a step-wise manner between limits of zero to 100% at a critical value of S_{tk}. In practice, for a well designed stage, E is a monotonic sigmoidal function of S_{tk50} or d_p that increases steeply from $E \sim 0\%$ to >95%, typically reaching its maximum steepness when E is 50% that is represented in Figure B-2, corresponding to the stage effective cut-off diameter, also termed its d_{50} value.

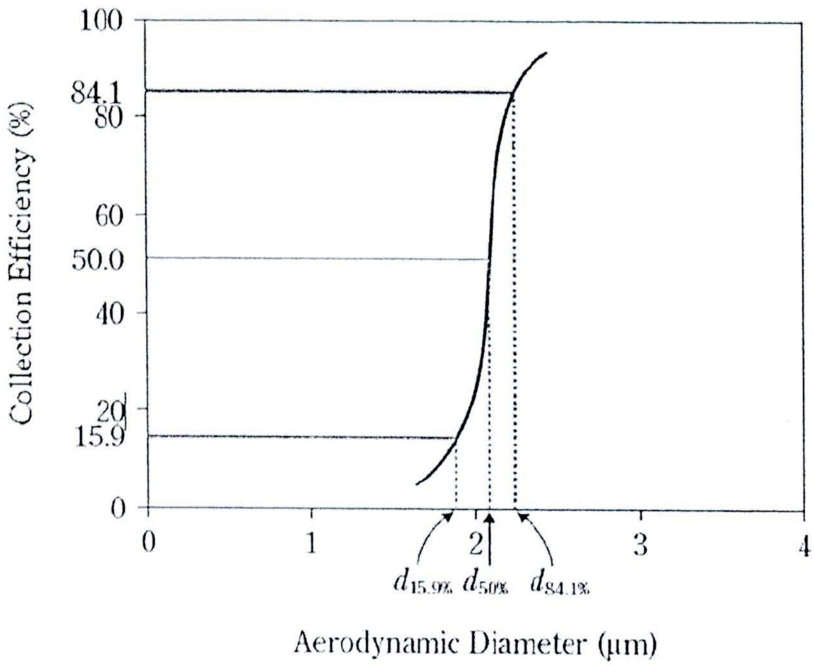

Figure B-2 Idealized collection efficiency curve for a single round-nozzle impactor stage.

It is possible to take into account the shape of the actual collection efficiency curve of the stage in the analysis of impactor data [76], but this refinement is rarely done for measurements of inhaler performance. Instead, the assumption is made that the mass of particles larger than d_{50} that penetrate the stage, is exactly compensated by the mass associated with particles finer than this size, that are collected [77]. Thus the d_{50} value is a constant for a given stage at a fixed flow rate. Marple et al. [78] and more recently Rader et al. [79], with an improved theory taking into account the effect of ultra-Stokesian drag, have identified that $\sqrt{S_{tk}}$ at E_{50} ($\sqrt{S_{tk_{50}}}$) should be close to 0.49 for well-designed round-nozzle impactors as shown in Figure B-2, where differences in particle inertia dominate the size separation process. The $\sqrt{S_{tk_{50}}}$ remains close to this value when the flow Reynolds number is within the range from 500 to 3000 [62].

The sharpness of cut of a given stage is defined in terms of the geometric standard deviation (GSD) of the collection efficiency curve, derived from the expression:

$$GSD_{stage} = \left(\frac{d_{84.1}}{d_{15.9}} \right)^{\frac{1}{2}} \tag{B-3}$$

where, $d_{84.1}$ d and $d_{15.9}$ d are the sizes corresponding to the 84.1 and 15.9 percentiles of the curve by analogy with the log-normal distribution function expressed in cumulative form, to which the shape of the collection efficiency curve approximates as is seen in Figure B-2. GSD for a well functioning stage should ideally be<1.2 (the GSD for an ideal size fractionators would be 1.0) [78].

However, in practice, values in many designs of commercial impactor can exceed this limit, especially with stages that classify particles larger than 5 μm aerodynamic diameters, where gravitational sedimentation significantly contributes to the size separation processes [79]. The influence of gravity becomes especially evident at low flow rates with impactors such as the NGI that are designed to operate over a range of flow low rates [78].

For multi-orifice impaction stages, Fang et al. [80] also identified the importance of the cross-flow parameter (X_c), as a parameter that affects stage *GSD*. X_c is defined as:

$$X_c = \frac{nW}{4D_c} \quad \text{(B-4)}$$

where, D_c is the diameter of a cluster of nozzles on the stage and n is the total number of nozzles on a stage. If the value of this dimensionless parameter is greater than 1.2, the potential exists for spent air leaving the orifices closer to the centre of the nozzle plate to interfere with the flow exiting the outer orifices, thereby preventing their flow from reaching the collection surface.

B-3 Chronicle of Impactor

There have been numerous impactor designed, built and described in various reports and in the open literature. Many impactors have been built for special studies, but only a few of the impactors have been widely distributed. Of the impactors that have been widely used, there are many writings in the literature describing variations and modifications made to these more "standard" impactors.

Of all the impactors, Anderson Cascade impactors have gained its enormous popularity. In 1956 the Anderson impactor was developed at Dugway Proving Ground for the specific purpose of determining the size distribution of viable airborne particles [81].

The impactor was consisted of six stages having 400 nozzles and impaction plates which were Petri dishes containing agar culture medium. The original prototype design used four stages consisted of 340 nozzles and impaction plate. After a sampling period, the Petri dish impaction plates were incubated and the viable deposits counted. May [75], did a redesign of the Anderson viable impactor after doing an extensive calibration and evaluation of stage deposit. In 1966 the Anderson viable impactor was modified for respiratory health hazard assessment of any airborne particles by substituting stainless steel impaction plates for the Petri dishes. The number and size of the nozzles per stage remained unchanged from the original viable sampler and this version was known as the nonviable Anderson impactor.

An MKI version of the impactor was introduced in about 1970 with the addition of two upper stages to make an eight-stage impactor with a larger cut size than the original. Later, the top two stages were modified and a pre-impactor was added in the MKII version following the work of Mc Farland et al. [82].

High Volume Cascade Impactors are commercially available to measure the size distribution and respirable mass fraction of airborne particles in outdoor and indoor environments. Compared to multiple circular jet designs, it has eight times the sensitivity, the sharpest cut-off, the lowest internal losses, and is easiest to use. They are designed to minimize particle reintrainment (or blow-off), to minimize inter-stage losses, and to maximize sharpness of cut-off.

MOUDI™[1] –micro-orifice, uniform-deposit impactor is a precision cascade impactors that provide high sampling flow rate, sharp cut-point, and low-loss characteristics. In the rotating version, the nozzle plates are rotated relative to the impaction plates to obtain near uniform particle deposit on the collecting substrate over the 25 mm impaction area. The aerosol particles are size-fractionated by the MOUDI™ into four equal geometrical increments per decade of particle size, making data interpretation simple and easy.

From 1970 forward, the design of impactors became rather routine work, and numerous impactors of all shapes and sizes for all manner of purposes now a day are designed and built.

B-4 The Low Pressure Andersen Cascade Impactor

Cascade impactors that are used in normal pressure suffer from the drawback of minimum size selection of 0.4 μm in diameter. The low pressure cascade impactors are thus in-use for the reason to have a choice of minimum size range from 0.02 to 0.06 μm in diameter along with the operating ability at the reduced pressure of a few tens of kilo-Pascals (kPa) [83]. Measurement of activity size distribution of radon decay products using several kinds of low pressure cascade impactors (e.g. Andersen, Berner, Davis, MOUDI etc) were reported in the literatures [4, 5, 6, 83]. Although several types of impactors have been designed and operated over the past few decades, the most widely used commercially available instrument is the Andersen impactor from Andersen Graseby Inc, USA. This is considered a standard instrument and has been widely tested and used in aerosol studies.

APPENDIX C

Radiation Detectors

1. Charcoal Canisters - Charcoal Canisters (CC) are passive devices requiring no power to function. The passive nature of the activated charcoal allows continual adsorption and desorption of radon. During the measurement period, the adsorbed radon undergoes radioactive decay. Therefore, the technique does not uniformly integrate radon concentrations during the exposure period.

2. Scintillation Counter- The scintillation counter is a solid state radiation detector which uses a scintillation crystal (phosphor) to detect radiation and produce light pulses. Scintillation counters are constructed by coupling a suitable scintillation phosphor to a light sensitive photomultiplier tube. This device is portable and easy to handle. There are three classes of solid state scintillation phosphors: organic crystals, inorganic crystals, and plastic phosphors. Inorganic crystals include lithium iodide (LiI), sodium iodide (NaI), cesium iodide (CsI), and zinc sulfide (ZnS). Inorganic crystals exhibit high efficiency for detection of gamma rays and are capable of handling high count rates. Organic scintillation phosphors include naphthalene, stilbene, and anthracene. This type of crystal is frequently used in the detection of beta particles. Plastic phosphors are made by adding scintillation chemicals to a plastic matrix. The plastic has high hydrogen content; therefore, it is useful for fast neutron detectors.

3. Solid State Nuclear Track Detector or SSNTD- is a sample of a solid material (photographic emulsion, crystal, glass or plastic) exposed to nuclear radiation, chemically etched, and examined microscopically. The basis of solid state nuclear track detection is that charged particles damage the detector within nanometers along the track in such a way that the track can be etched many times faster than the undamaged material. The size and shape of these tracks yield information about the mass, charge, energy and direction of motion of the particles. The main advantages over other radiation detectors are the detailed information available on individual particles, the persistence of the tracks allowing measurements to be made over long periods of time, and the simple, cheap and robust construction of the detector.

Alpha Track Detectors - An alpha track detector (ATD) consists of a small piece of plastic or film enclosed in a container with a filter-covered opening. Radon diffuses through the filter into the container and alpha particles emitted by the radon and its decay products strike the detector and produce submicroscopic damage tracks. At the end of the measurement period, the detectors are returned to a laboratory and placed in a caustic solution that accentuates the damage tracks so they can be counted using a microscope or an automated counting system. The number of tracks per unit area is correlated to the radon concentration in air, using a conversion factor derived from data generated at a calibration facility.

CR-39, or Allyl di-glycol polycarbonate - is a piece of plastic material, which can register alpha particles that hit it. This alpha radiation, which comes from radon and its progeny, does microscopic damage to the surface of the plastics. After chemical etching the damaged area is enlarged and seen as tracks. The tracks can be counted and related to the radon concentration in the air in which the detector was exposed. CR-39, has exceptionally few background tracks and it is protected from electrostatic influence and optimally sensitive (may be used over 14 days and up to 1 year).

4. Liquid Scintillation Counters (LSC) - are traditional laboratory instrument with two opposing Photo multiplier Tubes (PMT) that view a vial that contains a sample and liquid scintillator fluid, or cocktail. When the sample emits a radiation (often a low energy beta) the cocktail itself, being the detector, causes a pulse of light. If both PMTs detect the light in coincidence, the count is tallied. This type of detector is very expensive and not portable. The benefit of this type of detector is that it can detect many different types of isotopes. Most modern LSC units have multiple sample capability and automatic data acquisition, reduction, and storage.

5. Proportional Counter- is a common laboratory instrument with sample counting tray and chamber and argon/methane flow through counting gas. Most units employ a very thin ($\mu g\ cm^{-2}$) window, while some are windowless. Shielding and identical guard chambers are used to reduce background and, in conjunction with electronic discrimination, these instruments can distinguish between alpha and beta radiation and achieve low MDAs. Similar to the LSC units, these proportional counters have multiple sample capability and automatic data acquisition, reduction, and storage.

6. Multi Channel Analyzer- Multi channel Analyzer System with a sodium iodide crystal and photo multiplier tube (PMT) is a solid-state germanium detector or a silicon-type detector can provide a powerful and useful capability for counting liquid or solid matrix samples or other prepared extracted radioactive samples. Most systems are used for gamma counting, while some silicon detectors are used for alpha radiation. These MCA systems can also be utilized with well-shielded detectors to count internally deposited radioactive material in organs or tissue for bioassay measurements. In all cases, the MCA provides the capability to bin and tally counts by energy and thus identify the emitter. Again, most systems have automatic data acquisition, reduction, and storage capability.

Biography

Ms. Chutima Kranrod was born on April 1, 1976 at Samut parkan province. She graduated from Department of Applied Radiation and Isotopes, Faculty of Science, Kasetsart University in 1996 with a Bachelor of Science degree. Later, joined Department of Nuclear Technology, Faculty of Engineering, Chulalongkorn University and received Master of Science degree in Nuclear Technology in 2002. Then, continued in the same department as an assistant researcher under radionuclide analyses project for 3 years. In 2004, she was awarded a fellowship by Ministry of Education, Science and Technology (MEXT), Government of Japan to be trained in radon measurement techniques at National Institute of Radiological Sciences (NIRS), Japan. In 2006, she joined for a doctoral degree in nuclear technology at Department of Nuclear Technology, Chulalongkorn University. In 2007, she was awarded for the second time MEXT scholarship to undertake higher researches to facilitate her Ph.D. dissertation in NIRS for 11 months. In 2009, Prof. Dr. Shinji Tokonami invited her to continue her doctoral researches at NIRS for two more years. She presented her research papers at the international conference on Radioecology and Environmental Radioactivity at Bergen, Norway in June 2008 and in July 2009 presented another paper at 5th international Symposium on Radiation Safety and Detection Technology (ISORD-5) in Kitakyushu, Japan. These studies have resulted to three papers and have been submitted to Journal of Applied Radiation, Radioprotection and Isotopes and Radiation Protection Dosimetry.