出國報告（出國類別：開會）

參加 2011 年第 18 屆放射性核種度量及應用研討會(ICRM)

服務機關：行政院原子能委員會
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派赴國家：日本
出國期間：100 年 9 月 18 日至 100 年 9 月 23 日
報告日期：100 年 11 月 23 日

摘 要

本次奉派參加2011年9月18日至9月23日於日本茨城縣筑波市國際會議中心(Tsukuba International Congress Center)舉行之第18屆放射性核種度量及應用研討會(International Conference on Radionuclide Metrology and its Application, ICRM)。本屆ICRM會議第一次在亞洲召開，並由日本國家標準實驗室(National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology (NMIJ/AIST))舉辦，計有29個國家共104人與會。亞太地區計有台灣、中國大陸、南韓、日本、澳洲及紐西蘭等國派員參加，不過ICRM會議現場還是以歐美學者爲主。ICRM起源於1972年，由幾個國家的放射性核種度量專家在南斯拉夫的暑期學校召開第一次會議，目前每兩年召開一次會議，上一屆會議是於2009年由歐洲斯洛伐克之斯洛伐克度量學會實驗室所主辦。本屆會場外除有美國Canberra、美國ORTEC、日本SEIKO-EG&G、日本Hitachi Aloka、日本KAWAGUCHI、蘇俄RADEK等6家輻射偵檢儀器製造商展示儀器外，另有英國Metrologia及Physicsworld 2家期刊及日本同位素協會協助參展，現場並擺放期刊及產品DM，供與會人員參觀索取。本屆本會核研所共發表3篇論文海報，分別爲Proficiency test for clearance mixed-nuclide samples、The performance evaluating of a movable gamma-ray counting system for radwaste measurement、Evaluating praciticability of an method for routinely monitoring gross alpha and beta activities in Taiwan。未來應該鼓勵國內從事核種度量與分析的實驗室，積極參與ICRM兩年一次的會議，並積極參與國際間量測的比對，藉此與國際接軌以提升自己分析的能力及技術。

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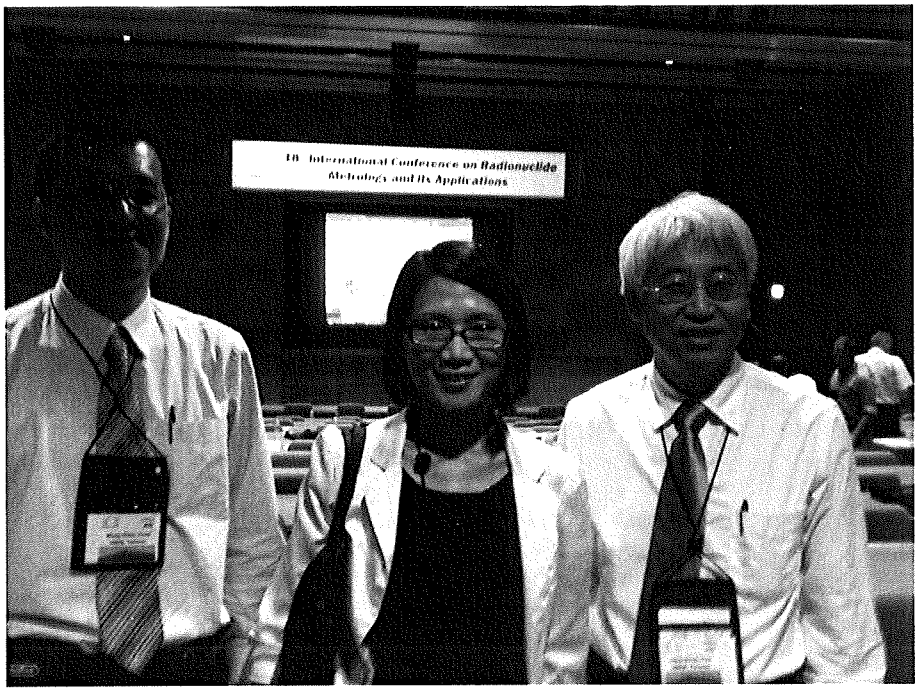
一、目的：

國際放射性核種度量委員會(International Committee for Radionuclide Metrology , ICRM)，其成員主要來自幾個國家的放射性核種度量國家實驗室(爲準會員)，另從事放射性核種量測與應用的研究人員(爲副會員) 所組成。它主旨是以資訊技術、應用程式的傳播及放射性核種劑量研究爲主的國際論壇，並爲基礎研究及工業應用問題提供了一系列的參考工具。

ICRM 會議幾乎是由全世界游離輻射國家標準實驗室共同參與，因此會議所討論的議題或結論，經常被國際度量衡局 (Bureau international des poids et measures, BIPM)游離輻射技術諮詢委員會(Consultative committee for ionizing radiation, CCRI)所採納，ICRM 設有主席、副主席、秘書及執行委員。

ICRM 會議目前每兩年舉辦一次，本會核研所在 2003 年於愛爾蘭都柏林大學舉辦的第 14 屆會議中，藉由日本及韓國代表之推薦與全體會員通過成爲 ICRM 會員。2011 年第 18 屆會議場地選擇在日本茨城縣筑波市國際會議中心 (Tsukuba International Congress Center) (圖 1)舉行。本次會議參加人數約爲 104 人，大部份是由世界各地的放射性核種度量專家參加，而台灣參加此會議除本人外，另由本會核研所國家游離輻射標準實驗室袁明程博士及葉俊賢先生參與(圖 2)。而透過兩年舉辦一次的 ICRM 會議可以了解各國核種度量實驗室最新的研究技術及成果，而會議中所提及量測方法、不準確的評估、活度量測結果的判斷，可作爲我國實驗室之間比對的參考。

(圖 1)筑波市國際會議中心(Tsukuba International Congress Center)



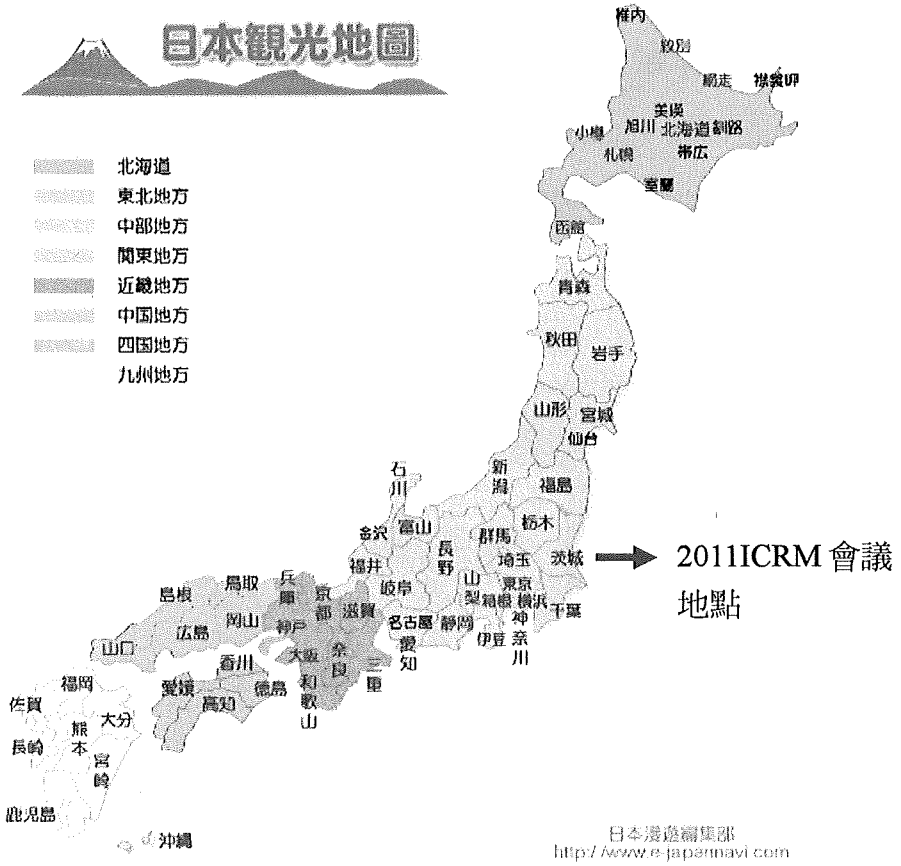
(圖 2)台灣與會同仁，左起袁明程(核研所)、本人(原能會)、葉俊良(核研所)

在大會舉行期間，此屆會議內容包括了口頭報告、ICRM各工作小組討論會議、論文張貼觀摩及現場相關分析儀器設備展示。其中口頭報告共分爲12個議題報告，分別如下：

1. 特別演講及國家度量概況(Special talk and aspects of international metrology)
2. 量測比對(Intercomparisons)
3. 核種衰變資料(Nuclear decay data)
4. 量測標準及參考物質(Measurement standards and reference materials)
5. 低背景量測技術(Low level measurement techniques)
6. 射源製備技術(Source preparation techniques)
7. 放射性核種度量技術(Radionuclide metrology techniques)
8. 加馬能譜(Gamma-ray spectrometry)
9. 液體閃爍量測技術(Liquid scintillation counting techniques)
10. 生命科學的放射性核種度量(Radionuclide metrology in life sciences)
11. 阿伐粒子及貝他粒子能譜(Alpha-particle and beta-particle spectrometry)
12. 特別演講(Special session: Invited talks about Fukushima NPP)

二、行程：

日　期	行　程	工 作 內 容
100/9/18	台北→日本茨城縣筑波市	去程
100/9/19~100/9/22	日本茨城縣筑波市	參加ICRM2011 會議
100/9/23	日本茨城縣筑波市→台北	回程



摘錄自日本漫遊編集部網站

三、會議紀要：

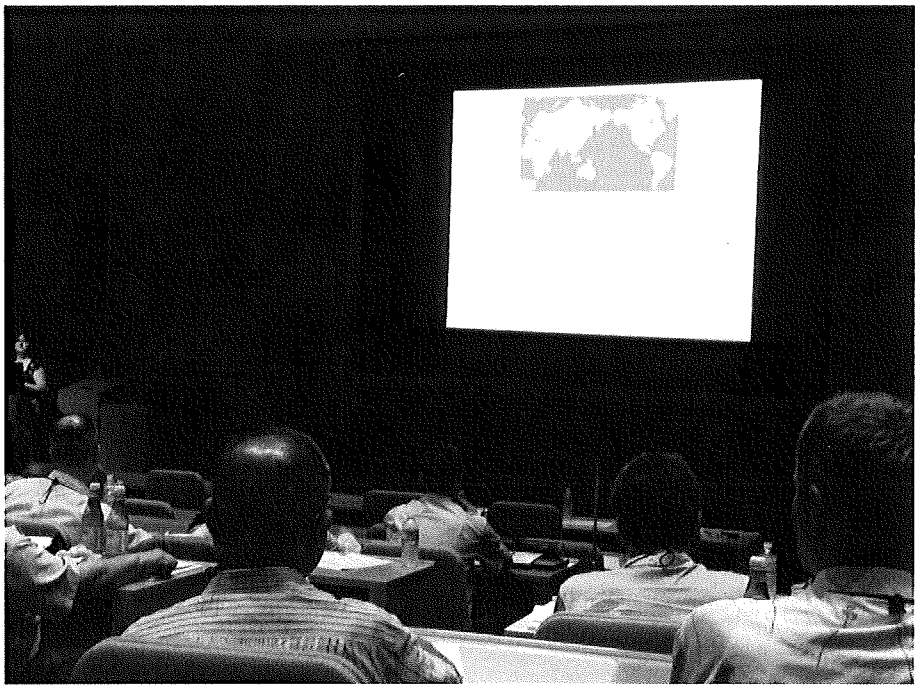
(一)國際放射性核種度量及應用研討會：

國際放射性核種度量及應用研討會(ICRM)於 2011 年 9 月 18 日至 9 月 23 日在日本茨城縣筑波市舉行，會議地點爲筑波市國際會議中心(Tsukuba International Congress Center)。

在大會舉行期間，有關會議口頭報告(Oral Presentation)、 ICRM 各工作小組討論會議(Working Group Meeting)及論文張貼 (Poster Session)內容等，分別說明如下：

1. 大會開幕式的特別演講(Special talk)：

首先，由今年主辦單位 National Metrology Institute of Japan(NMIJ)的局長 Yukinobu Miki 致開場白，感謝各位與會人員從世界各地來到日本開會。而開幕式中特別演講題目是由美國國家標準技術局(National Institute of Standards and Technology, NIST)的 Lisa Karam 博士爲大家介紹目前 ICRM 中各實驗室的消息(圖 3)。



(圖 3)開幕式的特別演講

2011 年的 ICRM 研討會主要以實驗室在建立及致力於放射性核種度量能力方式呈現，而放射性核種度量的能力決定了健康照護、環境監測及核能發展與興新的結合。另有希臘、立陶宛、印尼及挪威等四個國家實驗室同意參加，所以每個實驗室有關他們實驗的能力及標準也在會議中被討論。

2. 口頭報告：

這屆 ICRM 口頭報告共分爲 12 個議題，每個議題報告完畢後接著是相關議題的論文張貼簡述，讓與會人員在聽完這些議題報告後，在休息時間可以到論文展示的會場針對個人有興趣的題目，更進一步作了解。每個議題也有不少篇的口頭報告，每個報告人連同發問時間共有 20 分鐘的時間，大會只願意發給參加人員簡易摘要版講義，不允許將報告者的詳細簡報檔提供給參加人員。在會議期間共有 12 個議題，其中口頭報告共有 41 篇。

- 9 月 19 日上午報告

  Aspects of international metrology，計2篇

  Intercomparisons-I，計3篇

- 9 月 19 日下午報告

  Nuclear decay data-I，計4篇

  Nuclear decay data-II，計2篇

- 9 月 20 日上午報告

  Measurement standards reference materials，計3篇

  Low level measurement techniques，計2篇

- 9 月 20 日下午報告

  Source preparation techniques，計2篇

  Radionuclide metrology techniques-I，計3篇

  Radionuclide metrology techniques-II，計3篇

- 9 月 21 日上午報告

  Intercomparisons-II， 計1篇

  Gamma-ray spectrometry，計4篇

- 9 月 21 日下午報告

  Liquid scintillation counting techniques-I，計3篇

  Liquid scintillation counting techniques-II，計3篇

- 9 月 22 日上午報告

  Radionuclide metrology in life sciences，計 1 篇

  Alpha-particle and beta-particle spectrometry，計 2 篇

- 9 月 23 日下午報告

  Special session: invited talk about Fukushima NPP，計 3 篇

重點摘錄如下：

1） 量測比對 (Intercomparisons) 編號 100 的口頭報告，是在探討土壤中天然及人爲放射性核種的決定在歐盟之間比較的結果 (Determination of natural and anthropogenic radio-nuclides in soil-results of an EU comparison)。這個比較清楚的論證出在歐盟一些量測實驗室，需要改善有關決定鍶(Sr)及鈽(Pu)的分析過程，另外同位素鈾(U)也要額外

注意。然而，在銫-137、鉀-40、鉛及鉍同位素的加馬能譜決定中，大部分實驗室可以操控很好。

2） 核種衰變資料 (Nuclear decay data)編號 102 的口頭報告，是在探討 U-230 半衰期的量測(Measurement of the U-230 half-life)。 對於 U-230 半衰期的決定是藉著各式各樣的技術測量 U-230 的衰變曲線，包括小立體角的阿伐粒子計數、液體閃爍計數、4π 無窗 CsI 層疊偵檢器、比例計數器、HPGe 偵檢器的加馬射線能譜儀及離子植入的矽偵檢器。研究發現其 U-230 之衰變期超過 80 天，是目前所知 U-230 半衰期的 4 倍，有可能是受長半衰期子核種 Pb-210 的影響，而在過去子核種其實是被忽略的。

3） 量測標準及參考物質(Measurement standards and reference materials)編號 83 口頭報告，主要是探討氣體 Rn-222 標準化量測技術的發展 (Development of the primary measurement standard for gaseous radon-222 activity)。這一篇研究主要由南韓國國家標準實驗室(Korea research institute of standard and science, KRISS)所發表之成果，這個發展的系統是建立在Rn-222氣體分子在冷點(cold point)時，藉固體角方式計數阿伐粒子以決定活度。計數效率的決定在於光圈半徑(a)、Rn-222 的半徑(b)、射源及光圈距離的偏軸(e)、冷點與光圈間距離(z)，其中 a 及 z 主要使用座標量測機器去量測。KRISS 實驗室的系統當 z=82.6(1)mm、a=4.004(2)mm、b=3.9(4)mm，e＜0.5mm 所得的幾何因子(G)爲 5.854×10^{-4}，活度爲 42kBq 的 Rn-222 技術統計上的不確定性爲 0.336%，而系統度不確定性爲 0.263%，因此 Rn-222 量測得總不確定爲 0.427%。

4） 低背景量測技術(Low level measurement techniques)編號 123 口頭報告，主要探討廣島核爆後受到中子活化的鋼鐵樣品其 Co-60 的分布(Distribution of Co-60 in steel samples from Hiroshima)。HADES 實驗室使用地表下 225 公尺的兩個巨大鋼鐵樣品去量測，這樣品是分別由 Yokogawa 橋及 Aioi 橋而來。其中 Aioi 橋的樣品來自於距離原子彈爆炸處 671 公尺處，使用 0.5 公分的解析度去測量分析。而 Yokogawa 橋則距離爆炸處有 1423 公尺，因爲活度較小所以用 1 公分的解析度做分析。實驗結果顯示 Aioi 橋的樣品在中間 5.2 公分厚處的 Co-60 活度低於表面活度的 40%，而 Yokogawa 橋在中間 4.1 公分厚的活度較小，並低於 Aioi 橋的活度。

5） 射源製備技術(Source preparation techniques)編號 47 口頭報告，在探討量測 Ho-166m 半衰期的新方法((A new measurement of the half-life of Ho-166m)。在 1965 年時，所知道 Ho-166m 的半衰期爲 1200 年(其較大的相對不確性爲 15%)。當使用游離腔去校正量測 Ho-166m，另使用多收集式誘導偶和電漿質譜儀去量測 Ho-166m 原子的數目。而目前最新的 Ho-166m 的半衰期量測結果爲 1132.6±3.9 年(k=1)，她和 1965 年所決定的值相容的，但是現在所知的半衰期較之前少 5.6%，而且準確度多 43 倍，所以改善 Ho-166m 的半衰期不確定性，並盡可能當作參考游離腔的參考射源。

6） 放射性核種度量技術(Radionuclide metrology techniques)編號 96 口頭報告，在探討參考射源寬區域中光子激發的特性(Characterization of photon-emitting wide area reference

sources)。對於寬區域射源的光激發需求，並用濾片去移除不想要的 x 光及電子的貢獻。這些射源被使用去校正表面污染的偵檢器，對於均勻 class1 的參考射源，最近 ISO8769 修訂需要超過 5%。除此，有相當的限制在量測使用偵檢器前面之面罩設備。偵測系統的主要部分是鉛屏蔽裡的 NaI 偵檢器，並由電腦去控制校正射源的位置，另有一個 5 公分鉛的面罩板位在偵檢器前面，以決定低能量射源的穩定性。這個圓柱狀偵檢器的直徑爲 13 公分、厚度爲 10 公分且窗口是由 50 微米的鋁箔組成，對於低能量可以得到較合理的偵測效率。這個方法除應用於 Co-60、Cs-137、Co-57 及 Am-241 的核種外，也可以延伸至 I-129 及 Pu-238 光子射源的特性。

7） 加馬能譜(Gamma-ray spectrometry)編號 140 號口頭報告，探討康普吞抑制分光儀中一致合併性的修正(The coincidence-summing correction of the Compton-suppression spectrometer)。高純度純鍺偵檢器加馬能譜儀(High purity germanium (HPGE) gamma spectrometer)通常是使用於測量放射性核種，其中低能量加馬能譜的波峰通常是位在連續康普吞的高能量加馬射線中。當環境的樣品被量測時，爲了要累積足夠波峰的計數，所以收集的時間要花較久。然而，有一些弱的波峰無法看的到，所以較困難去定性與定量樣品中的放射性核種。幾個單一加馬核種使用於校正 HPGe 偵檢器的峰效率，如 HPGe 偵檢器及 NaI(Tl)偵檢器在 60 至 1115keV 的全部效率。而將能量延伸從 60 至 2800keV 時，蒙地卡羅的模擬軟體 MCNP4B 被使用。更進一步，可藉由 Co-60、Ba-137 及 Cs-134 核種活度的量測被

認證而延伸它的正當性。這個結果最後指出Compton-suppression spectrometer(CSS)的修正是根據HPGe活度的值。

8） 液體閃爍量測技術(Liquid scintillation counting techniques)編號 19 口頭報告，在探討一些商業液體閃爍體混合液中膠質粒子的決定(Determination of micelle size in some commercial liquid scintillation cocktail)。在典型涉及商業液體閃爍體混合的實驗中，放射性核種存在於可反轉的膠質粒子內。當一電子從放射性核種離開在經過水溶液物質因而失去能量，但這些能量不會沉積於閃爍體的物質中，所以不會導致閃爍光，就是所謂的膠質效應，而這對於鄂惹電子(Auger electron)激發核子的效率會大大降低。這一篇研究在於描述水及酸性的內容物會導致每個混合液中膠質粒子較穩定，並已在觀察流體粒子直徑中報告出來。它們也討論在計算膠質粒子效應的變動，及對於不正確膠體粒子大小所造成的誤差。例如，對於 Fe-55 在膠體粒子中用 8nm 取代 4nm 的直徑將會導致計數效率低於 0.4%至 0.6%。

9） 生命科學的放射性核種度量(Radionuclide metrology in life sciences)編號 132 口頭報告，在討論核醫製造廠商在放射性活度量測的品質保證過程(The NIST radioactivity measurement assurance program for the radiopharmaceutical industry) 。核子醫學中用於生產時活度的量測及沒有植入放射核種的使用追溯性是重要的，美國的國家標準技術局是在維持放射性藥物中活度量測的標準下建立，依次藉著比較與國際參考系統(International reference system, SIR)以

維持追溯的國際標準。96%的參與者超過 3000 種比較結果其對於 NIST 的差異是小於 10%，其中單一的放射性核種比較是在 164 至 379 種之間，如 Ga-67、Y-90、Tc-99m、Mo-99、In-111、I-125、I-131 及 Tl-201 這些的放射性核種，比較結果是在 NIST 的 10%內，也就是 89%至 96%之間，這些結果已經被參與者接受，其中參與最大的差異在於衰變的誤差及量的級數不一致性。

10） 阿伐粒子及貝他粒子能譜(Alpha-particle and beta-particle spectrometry)編號 93 口頭報告，在論述使用低溫度的偵檢器衰變能量光譜的發展(Development of decay energy spectroscopy using low temperature detector)。當使用低溫偵檢器發展高解析度去量測能量及阿伐粒子衰變活度，這些是包覆在 4π 金屬的接受器裡。目前的報告當中，對於各樣核種如 Am-241、Cm-244、Pu-238、Pu-239 及 Pu-240 總衰變能量幾種實驗 Q 光譜的量測被認證。在衰變的能量 Q 光譜中，當每一個加馬釋放衰變時，會在它們的 Q 值貢獻特性波峰，而可量測大於 6keV 的 FWHM 高解析度的 Q 光譜，這也清楚證明且被討論應用於 Pu 同位素和傳統的量測技術，其中這種絕對活度量測的能力也在使用 4π 量測方式中被討論。

11） 特別演講(Special session: Invited talks about Fukushima NPP)主要針對此次日本 311 地震後所發生福島核子意外事故當作主題，第一篇(編號 S-01)是由法國報告其國家資料中心(NDC)針對日本核子事故作一分析(Analysis of the Fukushima accident by the French national data center)。事故發生後 CEA-DAM 透過 NDC 自動反應過程，提供整天所

偵測後分析的空氣粒子及惰性氣體，去幫助了解事故後可能產生的分裂產物。並利用中尺度與全球尺度規模同向的大氣傳遞模式，預測雲團的外釋以評估日本與全球人類之輻射曝露。

第二篇(編號 S-02)是由日本放射性同位素協會主要演講有關日本福島核電廠危機中依 ICRP2007 建議做出緊急輻射防護反應(Radiological protection based on ICRP2007 recommendations-emergency response in Fukushima NPP crisis)，ICRP2007 建議區分三種曝露狀況，即計畫、緊急及既存曝露，這是包含所有可能的狀況。輻射防護是建議應用每個曝露狀況，也將曝露分爲三大類及職業、公眾及醫療曝露。ICRP 也應用三個基本防護原則，即正當化、最適化及劑量限度對於不同的曝露狀況，其中劑量限度不不適於醫療曝露。當緊急曝露或既存曝露狀況被認證時，一個參考的標準當作公眾曝露的個人劑量約束值。此次日本福島核電廠所引起的高污染空浮造成城鎮及村落污染，稱做輻射緊急計測之公眾量測，其須考慮公眾的反應。ICRP 對於緊急情況的建議是建立在突然的情況下當有最糟的事情發生時，並於幾天內被控制，所以未來 ICRP 有可能建議將緊急狀況由幾天延長至幾個月。

第三篇(編號 S-03)是探討從日本福島第一電廠核子事故中的經驗回饋(An aspect of learning lessons from Fukushima daiichi accident)初步檢討因缺乏氫爆概念、輻射監測系統不足、應變中心處理能力不夠且原子爐冷卻水供應不足造成降熱過慢而導致事故發生。故應發展在電力被破壞下仍具有監測能力的輻射量測系統，其可用於當電路

系統中斷原子爐已損壞時的高劑量率複雜作業環境。研究模擬精確分析氫產生的總量及氫氣突然燃燒與爆炸的特性，以判斷原子爐結構及設備是否損壞。藉由電腦模擬計算資料可以強化輕水式反應爐安全，以提供國際間電廠間參考。

3. ICRM 各工作小組討論會議(Working Group Meeting)

1） 低背景量測技術小組(LLMT-WG)：同意低背景的應用，不只限於輻射防護亦應用於洋流或大氣擴散模式驗證。下次 LLMT 會議於 2012 年於南韓國家標準實驗室(KRISS)主辦。

2） 放射性核種度量技術小組(RMT-WG)：CCRI-S7 比對結果，各國國家標準實驗室的不確定度分析仍缺乏共識，故實驗室分析結果差異頗大。

3） 液體閃爍量測技術小組(LSC-WG)：FPGA 技術用於 TDCR 及 CIEMAT 測量技術，另下次 LSC 會議於 2012 年在德國 PTB 實驗室舉辦。

4） 生命科學小組(LS-WG)：收集及改善醫用核種活度計檔位設定參數，歐盟 Metrology for molecular radio therapy 計畫徵求參與者，重點於活度測量、吸收劑量及影像品質。

5） 阿伐粒子及貝他粒子能譜小組(α、β-WG)：重新評估 Actinid 衰變數據，Cf-252、Pu-240、Pu-242 及 U-238 等 22 種核種的衰變數據不完整或較舊，應重新評估。

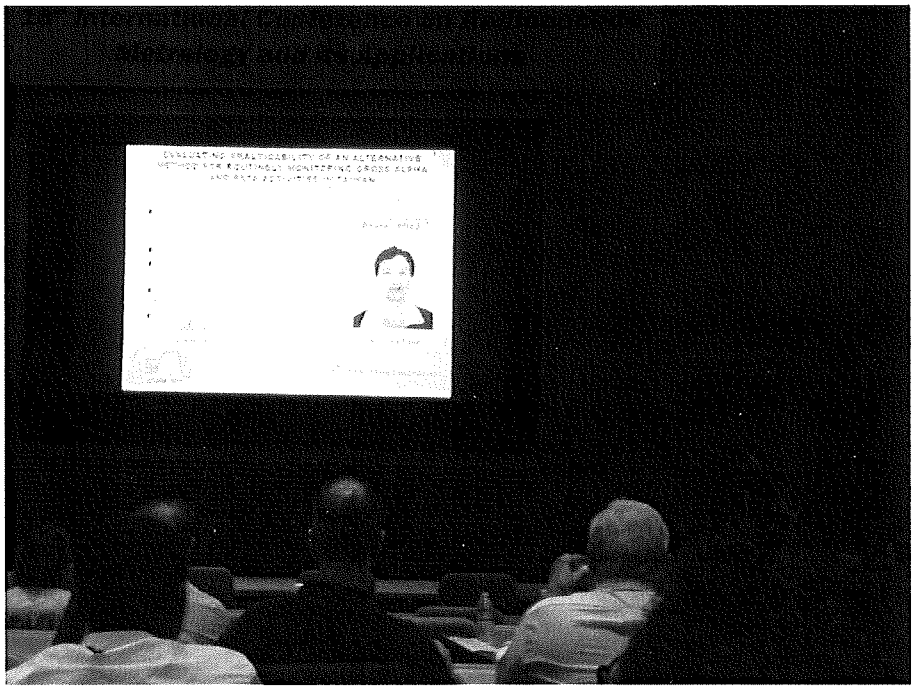
4. 論文張貼方式（**Poster Session**）

本次年會壁報論文，共有 12 個主題共有 68 篇，其主題如下所示：

- QA，計5篇
- Intercomparisons，計1篇
- Nuclear decay data，計7篇
- Measurement standards and reference materials，計6篇
- Low level measurement techniques，計4篇
- Source preparation techniques，計1篇
- Radionuclide metrology techniques，計16篇
- Gamma-ray spectrometry，計13篇
- Liquid scintillation counting techniques，計5篇
- Radionuclide metrology in life sciences，計6篇
- Alpha-particle and beta-particle spectrometry，計4篇

(二)展示場海報張貼

每當一個口頭報告主題結束，主持人就會介紹相關領域的論文壁報展示，其中有包括 3 件來自核研所的作品，一起同行的袁明程及葉俊賢先生的論文著作在會議中間也有被展示出來(圖 4)，報告的題目爲 "Evaluating practicability of alternative method for routinely monitoring gross alpha and beta activities in Taiwan"。



(圖 4)會議進行中展示袁明程先生的論文海報

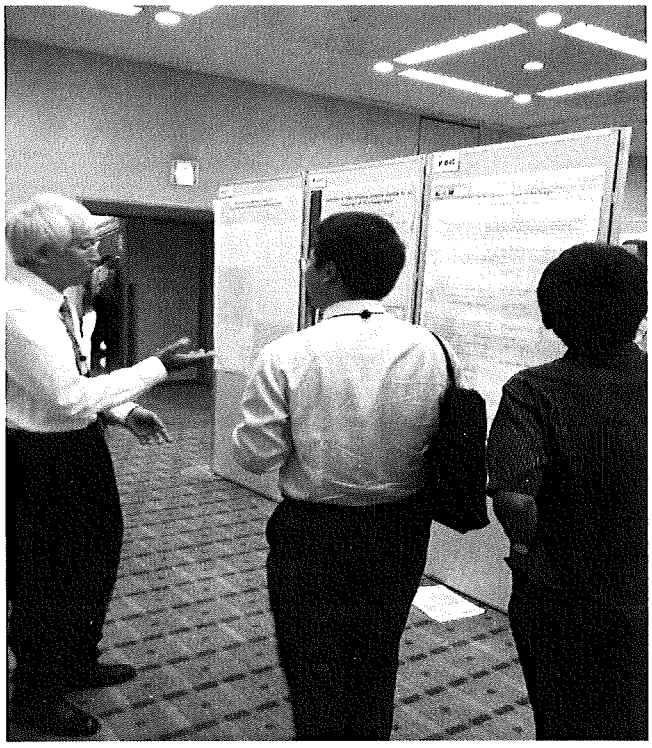
與會人員也於會議中休息時刻到海報展示處參觀張貼海報，了解各國家研究人員最近的所做的研究，而論文作者也在會場解答所有參觀人的疑問。核研所發表3篇論文海報，分別爲Proficiency test for clearance mixed-nuclide samples (編號P-046)、The performance evaluating of a movable gamma-ray counting system for radwaste measurement (編號P-011)、Evaluating praciticability of an method for routinely monitoring gross alpha and beta activities in Taiwan (編號P-043)。

編號46海報是在討論對於解除管制混合性核種中精確的測試(Proficiency test for clearance mixed-nuclide samples)(圖 5)，分爲盒裝及桶裝兩種包裝型式的樣品，裡

面包含了 Co-60 及 Cs-137 混合的溶液被參與的實驗室量測，最後結果和參考值作比較。其中 7 個參與者使用塑膠的閃爍計數系統，另 2 個參與者使用 HPGe 能譜分析儀系統去參與這個研究，共得到 40 種量測結果。這個評估結果顯示所有的參與者皆通過這個精確度的測試，即 En≦1，差異性在-25%至 50%之間。在這個精確的測試當中，參與者不能強烈被要求去評估 Cs 及 Co 的比值。大多數的參與者只能使用國家標準實驗室(National Radiation Standard Laboratory, NRSL)所提供比值，但仍有一些參與者試著去評估他們，並且得到 k=2 的可信度。對於我們實驗室間在未來的量測狀況下，不論是合作或技術的改變都有一致性的觀念。這一篇海報也獲得羅馬尼亞及中國大陸與會人員興趣，當休息時就前往海報會場詢問兩位核研所的同仁(圖 6、7)。

(圖 5)核研所張貼編號 46 之「對於解除管制混合性核種精確的測試」海報

(圖 6) 羅馬尼亞與會人員和袁明程及葉俊賢先生討論海報內容



(圖 7) 大陸與會人員和葉俊賢先生討論海報內容

編號 118 海報是在探討土壤不同深度中量測修正常數的決定(Correction coefficients determination for soil depth profiles measurement)(圖 8)，這個研究的目的是當取來的土樣在現場直接用加馬偵檢儀器(Gamma surveyor)去量測不同核種時，要如何用適當的係數去修正結果。其中 Gamma surveyor 是用來量測表面及鑽洞的設備，內裝主要爲 3×3 吋 NaI(Tl)晶體的多頻道加馬射線能譜儀。除使用 Gamma surveyor 來量測不同深度的放射性核種，另外將土樣收集後使用實驗室的 36%相對效率的 HPGe 加馬能譜分析儀做比較，共取表面及每 10 公分深度爲間格的 8 個土樣分析，並將分析結果做修正。實驗後發現結果還算滿意，但因爲不同深度中土壤的不均勻性是一個問題，故額外的修正係數仍須考量。

Correction coefficient determination for soil profiles measurements

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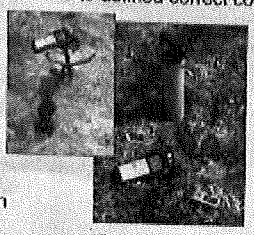
Introduction

situ gamma spectrometry measurements and the depth distribution of radionuclides in soil are still discussed and investigated problem. The Czec public is situated in the area with high content of natural radionuclides in rock and soil and knowledge of gamma-ray sources distribution fo liation monitoring, geophysical mapping or geological studies is necessary. The occurrence of radionuclides and also radon concentrations depe tribution are important information and both require depth profile measurements. 5 depth profiles (in different geological conditions) to the depth o cm (10 cm step of sampling) were measured and compared with laboratory gamma spectrometry sample measurements.

terials and methods

e of the in-situ gamma ray spectrometry objectives is the determination of the radioactive source deposition in the soil expressed in units of ime activity. For measurements using man-made calibration standards, the depth distribution of naturally occurring radiation sources in the soil is umed to be uniform. Measurements in a „bore-hole" have better counting efficiency and provide more information about subsoil comparing to 2π asurements and allow for shorter measurement times. On the other hand the excavation of the bore-hole may be difficult depending on the lock. The main task of the presented work was to defined correct coefficients to adjust results obtained by a Gamma Surveyor device that we use in-situ depth profile measurements.



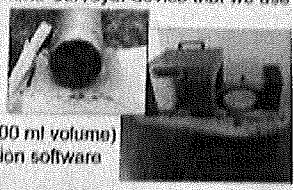
situ measurements

mma Surveyor
3" x 3" NaI(Tl) detector
512-channel gamma-ray spectrometer
surface and bore-hole measurements
offers spectral measurements with
determination of K (%), U (ppm eU), Th
(ppm eTh) concentrations.

Laboratory measurements

Semiconducting HPGe detector
- relative efficiency 36%
- 1.65keV energy resolution at 1.33MeV

Marinelly baker measurement geometry (600 ml volume)
MAESTRO Multi Channel Analyzer Emulation software
Genie 2000 Gamma Analysis Software
Stationary 10cm thick lead shielding



Measurement result notes

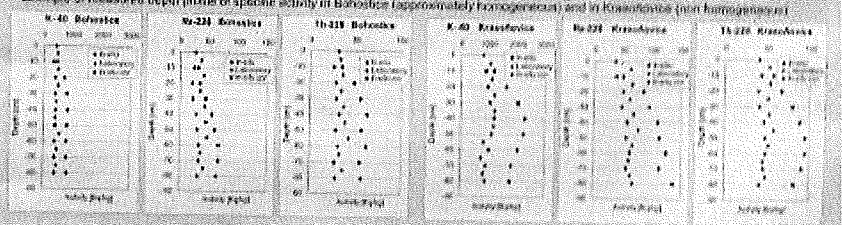
Comparison of in-situ and laboratory results. Bohostice depth profile can be consider as homogenous in soil composition and density, while Krasoňovice area is non homogenous. Corrections requirements are necessary mainly for deeper measurements. Use of corrections achieve best results for K-40.

pth profiles geological description

correct interpretation of results obtained it is
essary to take under consideration the
logical situation of the site.

Depth profile	Geological bedrock
Bohostice	Leucocratic biotitic orthogneiss
Dubenec	Argillaceous and calcareous sandstone, locally gritty, and conglomerate
Krasoňovice	Double-mica migmatite up to orthogneiss
Nový Dvůr	Biotitic and muscovite-biotitic gneissose granite
Na Kubánině	Biotite and muscovite-biotitic gneissose paragneiss, leucocratic migmatite

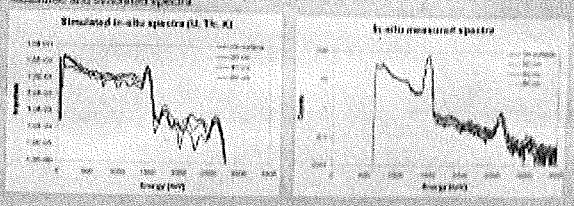
Example of measured depth profile of specific activity in Bohostice (approximately homogeneous) and in Krasoňovice (non homogeneous)



nte Carlo simulation

arate calculations for three sources were performed:
nium and thorium chain (twenty most abundant intensities of
mma rays) and potassium.
nogenous distribution of radionuclides in soil was assumed.
mma ray spectra calculated for the source dimensions 112 cm in
meter.

Measured and simulated spectra



rection coefficients

e types of methodology were compared.
orrection calculation based on measurements. Comparison of depth profile and laboratory
easurement: Calculated – measurement
orrection calculated from Monte Carlo simulation: Calculated - MCNP
rrection based on information available in reference literature (Matolín M., Minty B. IAEA-CN
-65, UNAM 2009): Used – Matolín 2009
ections for different soil density, which influence the radiation attenuation in surrounding
enals were not included in calculations.

Depth [cm]	Calculated - Measurement	Depth [cm]	Calculated MCNP	Used Matolín 2009
0-10	1	0	1	1
10-20	0.89	10	0.85	0.753
20-30	0.68	20	0.72	0.59
30-40	0.62	30	0.71	0.515
40-50	0.59	40	0.64	0.47
50-60	0.54	50		0.3
60-70	0.53	60	0.61	0.3
70-80	0.54	70	0.61	0.3

Conclusions

he depth distribution of radionuclides is still not thoroughly understood. The main task of this
work was to compare calculated geometrical correction factor for in-situ measurements with correction factors available in reference literature.
The results are considered satisfactory, but some additional calculations have to be performed, because influence of material density and the
not-homogenous depth profile were not included. Both of these parameters are important to achieve most precise results.

Acknowledgement

This work was supported by the grant of the CTU SGS 10/212/OHK4/2T/14 and of the Czech Science Foundation GAČR 202/09/H086.

(圖 8)編號 118 之「土壤不同深度中量測修正常數的決定」海報

而今年最佳論文海報的得獎者仍爲法國題目爲"Simulation of Cherenkov photons emitted in photomultiplier windows induced by Compton diffusion using MonteCarlo code GEANT4"(圖 9)。

(圖 9)得獎者及得獎海報

四、心得與建議：

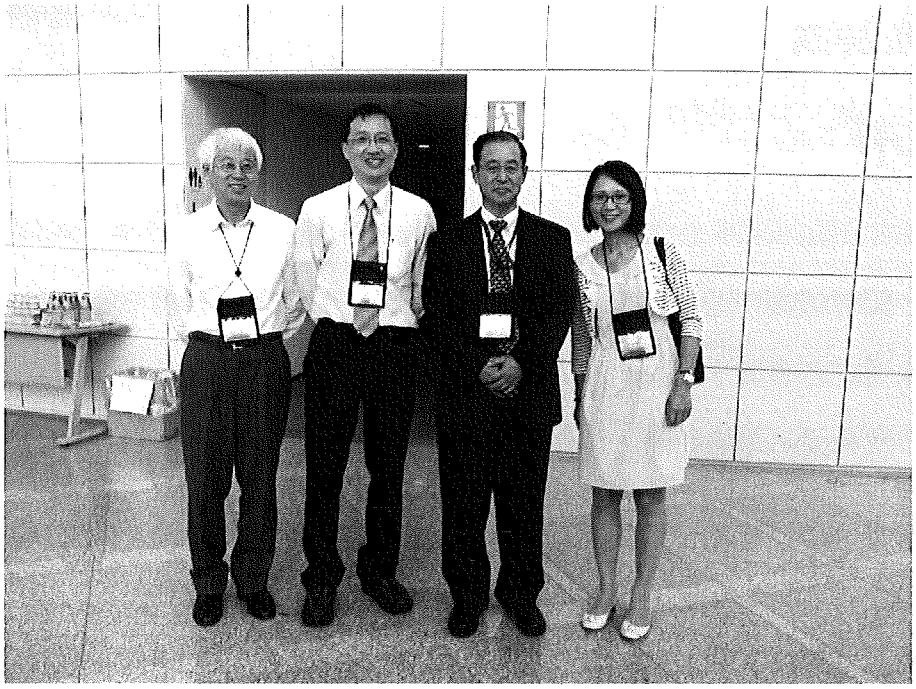
(一) 心得：

1.目前國內全國認證基金會舉辦放射性實驗室核種量測之間的比對，參與實驗室包括核研所、輻射偵測中心、清華大學、成功大學及台電放射試驗室等，藉由能力試驗比對，來增進各實驗室核種分析的準確度。此次會議中所提及量測方法、不準確的評估、活度量測結果的判斷，可作爲各實驗室之間比對的參考。

2.透過 ICRM 會議可以了解國際間各核種分析實驗室所研究的動向，並經由各分組的工作會報了解每一分組目前正進行的工作，及分享彼此最近的成果，並了解目前各組實驗室方向及新的研究成果。

3.雖然此次會議內容幾乎都以核種度量分析技術及應用爲主，但主辦單位在會議結束前特別針對日本福島核子事故舉辦三場特別演講。目前日本相關單位正研究模擬去正確分析氫產生的總量及氫氣突然燃燒與爆炸的特性，以判斷原子爐結構及設備是否損壞。而藉由電腦模擬計算資料可以強化輕水式反應爐安全，以提供國際間電廠間參考。

4.感謝這次日本 NMIJ 承辦單位努力，承辦單位籌辦期間恰巧發生 311 福島核子事故，而舉辦地點茨城縣又離福島縣距離不遠，爲了讓參與會議的各國人士了解當地輻射劑量率，NMIJ 特別將一些輻射相關資訊放在報名的網站上，要讓所有參加的人員可以安心地前往開會，所以特別在會議結束前和承辦秘書合影(圖 10a.10b)。



(圖 10a)台灣與會人員與日本主辦單位 NMIJ 科學秘書 Yoshio Hino 合影

(圖 10b)台灣與會人員與日本主辦單位 NMIJ 會議秘書 Akira Yunoki 合影

(二) 建議：

1.國內核種度量與分析實驗室應積極參與 ICRM 會議，透過論文的發表產生互動，可進而了解國際間各國家實驗室研究的新趨勢，也能提升國內量測技術的能力。並於會議期間收集最新國際核種活度度量的發展資訊，可作爲實驗室未來發展或互相合作的參考。

2.國內各實驗室應該積極參與國際間量測天然及人工核種的比對，藉此與國際接軌並提升自己的實力。這除了是對於自己量測技術的肯定，也能確保自己與國際間分析結果的一致性。

3.此次日本核子事故之應變處置作爲，將緊急狀況由幾天延長至幾個月，ICRP 可能會在未來改變的原先建議，本會應該注意 ICRP 在此議題上之最新發展，當作國內核子事故緊急應變的參考。

4.目前日本正研究模擬精確分析氫產生的總量及氫氣突然燃燒與爆炸的特性，以判斷原子爐結構及設備是否損壞。藉由電腦模擬計算資料可以強化輕水式反應爐安全，以提供國際間電廠間參考，相關之成果也可作爲我國參考。

5.目前國內「核能電廠現有安全防護體制全面體檢方案」中，本會已要求台電公司檢討喪失所有廠區交流電源之處置措施，及強化氫氣排放與避免爆炸因應措施。亦於 100 年第 37 次核子設施輻射防護管制會議中要求台電公司檢討核能電廠廠區輻射偵檢器之備用電源(電源應維持 72 小時以上)，決議中已請台電公司就擴充不斷電系統容量及增設電源車以及移動式發電機規劃時程，應請台電公司確實落實。

五、附錄
(一)現場展示海報

P-043

Evaluating practicability of an LSC method for routine monitoring gross alpha and beta activities in water samples in Taiwan

Chi-Feng Lin, Jeng-Jong Wang*, Ju-Chuan Huang, Chin-Hsien Yeh, Ming-Chen Yuan, Bor-Jing Chang

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Abstract

A procedure using liquid scintillation counting for the monitoring of gross alpha and beta activities in environmental water was implemented to improve the conventional procedure using GFPC adopted in Taiwan. The new procedure was acquired through calibration and validation, and then was applied to the monitoring of surface water in Taiwan. This procedure can improve 2~4 times of detection efficiencies and takes only 70~80 % of analysis time with reliable accuracy. With these features, the newly developed procedure is favorable during an emergency situations.

Introduction

The routine method for monitoring gross alpha and beta activities in Taiwan relies on gas-flow proportional counting (GFPC). However, several studies have suggested using the liquid scintillation counting (LSC) method in place of GFPC due to its advantages of higher detection efficiencies, simpler operation and faster analyzing time.

Method

Calibration of the Liquid Scintillation Counter → Validation of the Counting Methods and the Test Procedures → Determination of Gross alpha/beta Activity in Real Samples

Results

Table 1. Results of validation tests (k = 1)

Counting method	Reference value (Bq/L) alpha	beta	Measure value (Bq/L) alpha	beta	Deviation (%) alpha	beta
LSC	0.12 ± 0.01	-	0.12 ± 0.02	< MDA	0	-
	0.30 ± 0.01	-	0.27 ± 0.02	< MDA	-10.0	-
	0.61 ± 0.01	-	0.63 ± 0.02	< MDA	+3.3	-
	-	0.21 ± 0.01	< MDA	0.23 ± 0.02	-	+9.5
	-	1.07 ± 0.02	< MDA	1.12 ± 0.04	-	+4.7
	0.12 ± 0.01	0.21 ± 0.01	0.12 ± 0.02	0.21 ± 0.02	0	0
	0.61 ± 0.01	1.07 ± 0.02	0.56 ± 0.04	1.18 ± 0.06	-8.2	+10.3
GFPC	0.12 ± 0.01	-	0.10 ± 0.02	< MDA	-16.7	-
	0.30 ± 0.01	-	0.27 ± 0.04	< MDA	-10.0	-
	0.61 ± 0.01	-	0.53 ± 0.06	< MDA	-13.1	-
	-	0.21 ± 0.01	< MDA	0.23 ± 0.02	-	+9.5
	-	1.07 ± 0.02	< MDA	0.95 ± 0.06	-	-11.2
	0.12 ± 0.01	0.22 ± 0.01	0.11 ± 0.02	0.18 ± 0.02	-8.3	+18.2
	0.61 ± 0.01	1.07 ± 0.02	0.54 ± 0.06	1.09 ± 0.06	-11.5	+1.9

Table 2. Comparison of the performance between LSC and GFPC (SQP(E) = 760)

	LSC alpha	LSC beta	GFPC alpha	GFPC beta
Bias	-0.03	0.06	-0.12	-0.04
Precision	0.07	0.08	0.14	0.13
Efficiency (%)	99.61	94.24	19.62	44.13
Background (cpm)	0.47	9.75	0.04	1.88
MDA (mBq/L)	19.12	67.37	29.76	60.55
Overall analysis time (hr)	3.5		4.5	

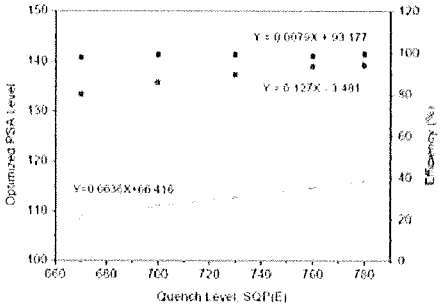


Fig. 1 Quench curves (■ optimized PSA level, ■ detection efficiency for alpha, ● detection efficiency for beta)

Table 3. Results of environmental monitoring utilizing LSC method (k = 1)

Source	Site	SMP date (2011)	Radioactivity gross alpha (10⁻³ Bq/L)	gross beta (10⁻³ Bq/L)
Keelung	a	May 3	< MDA	98.72 ± 28.63
	b	May 9	34.26 ± 7.79	332.49 ± 26.27
	c	May 18	< MDA	92.72 ± 28.67
	d	May 17	< MDA	103.40 ± 28.64
Hsinchu		Apr. 25	< MDA	144.12 ± 28.52
Han	a	Apr. 26	< MDA	< MDA
	b	May 3	< MDA	123.09 ± 28.11
	c	May 9	< MDA	< MDA
	d	May 18	< MDA	76.63 ± 29.96
	e	May 24	< MDA	111.10 ± 29.43
	f	Jun. 1	< MDA	141.46 ± 27.14
Maboli		May 2	< MDA	178.85 ± 26.65
Taichung		May 9	< MDA	116.31 ± 27.01
Nantou		May 16	< MDA	103.53 ± 27.85
Changhua		Jun. 1	< MDA	153.25 ± 26.46
Yunlin		Jun. 2	< MDA	109.05 ± 27.20
Hualien	a	Apr. 26	< MDA	163.03 ± 26.55
	b	May 3	< MDA	335.23 ± 26.36
	c	May 9	< MDA	198.24 ± 25.90
	d	May 18	< MDA	96.88 ± 28.54
	e	May 24	< MDA	< MDA
	f	Jun. 1	< MDA	< MDA
Taitung		May 24	< MDA	125.54 ± 28.67

Conclusion

The LSC procedure has been implemented for routine monitoring of radioactivity in water samples in Taiwan. The procedure reduces sample processing time for 1~2 hours with acceptable accuracy, in comparison to the conventional GFPC method. In addition, the procedure has been applied to the environmental monitoring of water in Taiwan, with results showing compliance with the regulation limits on the radioactivity of water quality.

Proficiency Test for Clearance Mixed-Nuclide Samples

Chin-Hsien Yeh *, Ming-Chen Yuan , Bor-Jing Chang

Abstract

In 2010, the National Radiation Standard Laboratory held a proficiency test for measurement and analysis of clearance mixed-nuclide samples. Two types of samples, box-shape and drum-shape, containing ^{60}Co and ^{137}Cs mixed solutions were measured by the participating laboratories and their results were then compared with the reference values. Seven participants used plastic scintillator counting systems and two participants used HPGe spectrometer systems to join in this study, obtaining 40 measurement results. The evaluation results showed that all the participants passed the requirements of this proficiency test, $E_n \leq 1$, and the discrepancy (B_i) was between -25 % and 50 %.

Methods

When the nuclide composition of the sample is known and a reference nuclide is selected, the relationship between the activity of the reference nuclide and the net counting rate is written:

$$A_s = \frac{N}{\varepsilon_s\left(1 + \sum_i (E_i \times R_i)\right)}$$

A_s = the activity of the selected nuclide,
N = net counting rate,
ε_s = the counting efficiency of the selected nuclide,
E_i = detection efficiency ratio of the i-th radionuclide against the selected nuclide.
R_i = activity ratio of the i-th radionuclide against the selected nuclide.

So, the activity of the i-th radionuclide, Ai, could be express as

$$Ai = Ri \times As,$$

Table 1. Specifications of box-type and drum-type mixed-nuclide samples

Sample Type	Box-type				Drum-type			
Sample I.D	MB1		MB2		MD1		MD2	
Total activity in the sample (Bq)	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co	^{137}Cs	^{60}Co
	1998	2356	2643	4044	12037	11037	10152	20319

Table 2. Typical uncertainty budget of the participant's counting result.

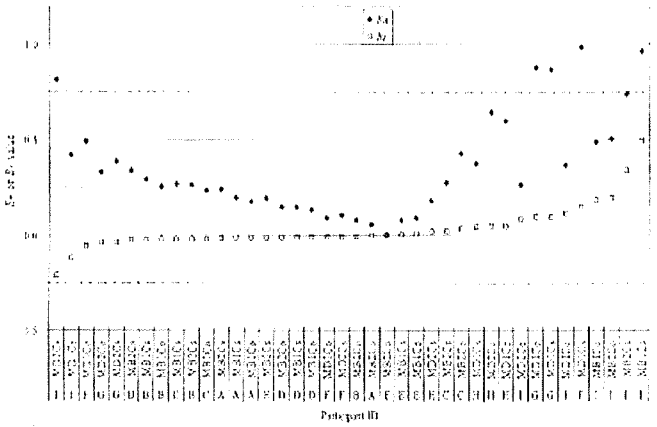
Source of uncertainty	Relative standard uncertainty (%, k=1)	
	Plastic Scintillator	HPGe
Net counting rate or peak area (dead time, background variation included)	2.1	5.1
Counting efficiency (Standard source variation included)	2.0	2.0
Weighing	0.5	0.5
Long term stability	2.0	2.0
Time base	0.1	0.01
Combined standard uncertainty	3.6	5.9
Expanded uncertainty (k=2)	7.2	12

Results

Most participants had measurement uncertainties between 6.5 % and 12 %; only participant "I" showed their measurement uncertainty much larger than others (probably because participant "I" used one portable HPGe detector to measure the samples under shield-less conditions). Low counting efficiencies and high background variations caused the greatest measurement uncertainty.

All of the measurement results could meet the criteria of this testing, i.e., $En \leq 1$ and the Bi between -0.25 and 0.5, and most of the Bi values of participants were within the range of -0.2 to 0.2, i.e., the discrepancy between the reference value and the results of participants is less than 20 %.

Fig . The E_n value and B_i value of this proficiency test.



Conclusion

In this proficiency test, participants were not strongly requested to evaluate the ratio of Cs:CO. Most participants used the ratio value provided by NRSL., but still there were some participants (H and I) who tried to evaluate them and obtain acceptable result in $k=2$ confidence level. That gave us some confidence for the cooperation and technical exchanges between laboratories in the real-life measurement situation in the future.

In the future proficiency test plans, the complexity of the sample and the strictness of criteria will be increased step by step. At that time, more nuclides will be added in solid, inhomogenous or heterogenous samples and the participants would be asked to report the activity of each nuclide totally from their own evaluation.

The examination of source distribution in a large sample by Monte Carlo simulation

No. #088

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Introduction

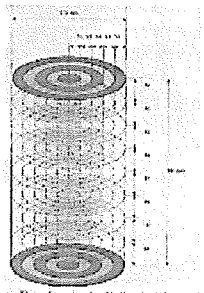
A particular problem in gamma ray spectrometry of large volume samples is represented by the variability of source distribution. In this work realistic Monte Carlo simulations were carried out for several distributions of activity in a waste drum with the purpose to observe the dependence of the efficiency on the source distribution and to test whether the efficiency can be correlated with the shape of the spectra. The simulations were carried out for the gamma rays of ^{60}Co and for the main photons emitted by ^{152}Eu. As quantitative measures of the shape of the spectra, the ratio of the count rates in two peaks of the same nuclide as well as the peak to Compton ratio were considered.

The methodology

- The volume of the drum was divided into a number *n* of smaller volumes that were each independently submitted to Monte Carlo simulations.
- The volume elements were defined by applying two divisions of the volume of the drum.

The first stage

- A random emission point was sampled in one of the intersections of the S1,..S8 and V1,..V5 domains selected for the current computation, i.e. a uniform distribution was assumed in one of the 40 domains. For each location of the source the simulations were carried out for the gamma rays of ^{60}Co and for the main photons emitted by ^{152}Eu. For each energy 1 2^{19} photons were traced in total for the 40 volume elements.



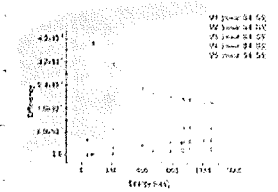
The volume invoked in the simulation

The efficiency strongly depends on the position of the source.

In the case of the outermost tube an important fraction from the emission points are close to the detector and to the boundary of the drum.

For these emission points, the detection solid angle is high and the attenuation in the drum is low.

At the other extreme for emission points located within V1 the attenuation is high (increasing with decreasing energy) and the solid angle is small.



The peak efficiency curves for the inner cylinder and the four tubes (mean S4, S5)

The second

- One set of simulations included point sources placed on the detector axis at several distances from the detector, such that the photon path length in the drum was 0.12, 5.12, 10.12, 15.12, 20.12 and 25.12 cm. Another case was for a point source located off axis of the detector, on a line inclined with 30° with respect to the axis, the position of the source was chosen such that the path length through the drum was also 25.12 cm, but the distance to the detector was bigger. In each case the simulations were carried out for the photons emitted by ^{60}Co (1173.24 and 1332.51 keV) and for two photons emitted by ^{152}Eu (778.90 and 1408.01 keV). In this case about 2^{9} photons were simulated for each energy, for each position of the point source.

- The Monte Carlo simulations were carried out using GEANT 3.21 code.

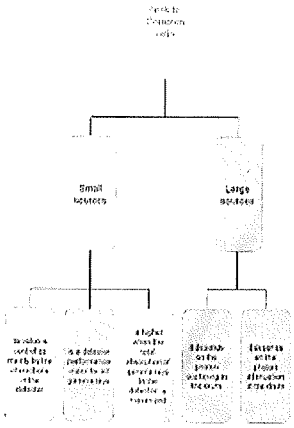
??? The shape of the spectra can provide information on the source location or directly on the appropriate efficiency corresponding to the source distribution ???

- The detected spectrum registered by the detector depends on all the interactions suffered by the photons prior to detection, dependent in turn on the photon path length and interaction coefficients in the materials. As quantitative measures of the shape of the simulated spectra, the ratio of the count rates in two peaks of the same nuclide as well as the peak to Compton ratio were considered in this paper.

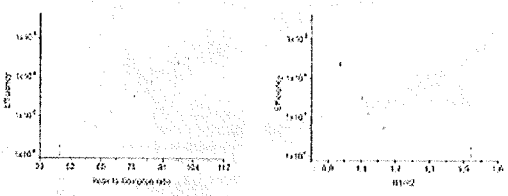
Results and discussions

Interested in the change of the peak-to-Compton ratio due to higher scattering and higher absorption in the drum when the source is located far inside the drum.

The case of point sources

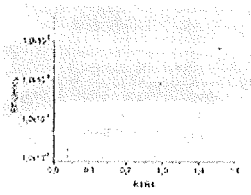


- Located on the axis of the detector at different depths inside the drum.
- The efficiency can be directly correlated with the position of the source.
- The shape parameters of the spectra (the ratio of the count rate in two peaks of the same nuclide and the peak-to-Compton ratio) are well correlated with the position.
- To see if the detection efficiency can be correlated with shape parameters of the spectra, the efficiency for the ^{60}Co photon with energy 1332 keV is represented as a function of the peak-to-Compton ratio and as a function of the ratio R1/R2 independent of the position of the source, the peak-to-Compton ratio was computed using the mean number of counts from the Compton plateau in the energy range from 1040 to 1096 keV and R1 and R2 are the counts from the 1332 and 1173 keV peaks.
- The ratio R1/R2 is less convenient as a parameter for efficiency estimation, because the efficiency changes strongly for a relatively reduced change of this ratio.
- On the other side, from the experimental point of view, the evaluation of the peak-to-Compton ratio related to the nuclide of interest requires a correct description of the Compton contribution of that nuclide to the spectrum, i.e. at least a good background subtraction.



1332.50 keV peak efficiency vs. Peak-to-Compton ratio (left) and ratio of R1 and R2 (right) for point sources

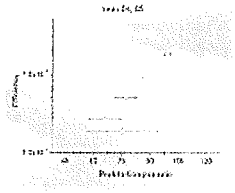
- The situation is better in the case of the efficiency for the 1408 keV peak of ^{152}Eu, in this case the ratio R3/R4 of the counts in the 1408 (R3) and 778 keV (R4) peaks, corresponding to bigger differences in the energies, has a larger range of variation.



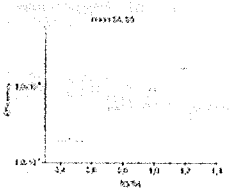
1408.01 keV peak efficiency vs. ratio of R3 and R4 for point sources

The case of volume sources

- The dependence of the efficiency on a position coordinate representative for the location of the domain is less obvious. On the other hand, what matters is not necessarily the source position, but rather the appropriate detection efficiency. The correlation is affected by bigger uncertainties. Due to the symmetry with respect to detector and drum, the mean values for S4 and S5 were displayed. The correlation pattern for the case of volume sources is similar with the correlation pattern for the case of point sources.



1332.50 keV peak efficiency vs. Peak-to-Compton ratio (left) and ratio of R1 and R2 (right) for volume sources



1408.01 keV peak efficiency vs. ratio of R3 and R4 for volume sources

Conclusions

The measuring strategy used in the assay of radioactive waste today is, in some aspects, not optimal, and the aim of this study has therefore been to investigate the effects of using alternative approaches, like Monte Carlo simulations. Simulations were carried out independently for several distributions of the sources in the volume of a waste drum. The peak-to-Compton ratio and the ratio of the count rates in the two peaks of ^{60}Co and respectively of ^{152}Eu were studied as providing information on the source distribution and in fact, on the peak efficiency appropriate for the specific source distribution. In favorable conditions these shape parameters can be used for reducing the uncertainty of activity evaluation resulting from unknown source distribution.

Acknowledgments

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Poster 082

ICRM 2011

A New Approach in Gamma-Ray Scanning of Rotating Drums Containing Radioactive Waste

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IFIN-HH

1. Introduction

The paper presents a new approach in the integral gamma scanning of rotating waste drums with relatively homogeneous matrix and heterogeneous activity distribution. It is based on solving a first-kind Fredholm integral equation using a small number of measurements. Numerical experiments show that the approach can give accurate measurement results especially for multi-gamma emitters. The approach was applied to the activity measurement of a certified linear ^{152}Eu source placed at different radial positions into a 220-liter drum filled with cement. [illegible]

2. Computational model of the peak efficiency for uncollimated HPGe detectors

[illegible]

Figure 1

3. Description of the new approach

The new approach in the integral gamma scanning of rotating waste drums is based on solving the first-kind Fredholm integral equation (1) by using a small number of measurements. The rotating waste drum is measured for different positions of the detector defined by d(k) and $z_d(l)$, where k=1, 2,...,k_{max} and l=1, 2,...,l_{max}. In this way, the peak count rates $R_{p,i}$ (i) corresponding to the ith position of the detector (i=(l-1)k_{max}+k) are obtained. Here we consider that the maximum number of measurements is twenty and accurate measurement results are used. To obtain a computer approximation of the activity, integral equation (1) is discretized by dividing the drum volume into a system of voxels of equal volumes composed of cylindrical rings and central cylinders. By means of the discretization, integral equation (1) is converted into the following system of algebraic equations: [illegible]

[illegible]

where [illegible]. Because α(j) takes values in a narrow interval around unity [illegible]. In this way, it was proved that the new approach can also be applied to the measurement of waste drums with relatively homogeneous matrix because the additional error due to the matrix heterogeneity is small. To compute A_t the system of algebraic equations (7) is firstly normalized in such a way that the Euclidean norms of the matrix and the right-hand side are equal to unity. As a result, we obtain the following system of equations Ax=b (8) [illegible] are Euclidean norms. To solve the ill-conditioned system of equations (8), the Tikhonov regularization method with non-negativity constraints was chosen because it is fast and simple. [illegible] where α (regularization parameter) is a real positive number. The minimization problem is equivalent to the equation $(A^TA+\alpha I)x=A^Tb$ (9), where I is the identity matrix and A^T is the transpose of A. To obtain a nonnegative solution for Equation (9), the nonnegative least squares algorithm introduced by Lawson and Hanson (1987) was applied. The non-negativity constraint is physically realistic because the components, A(j), of the activity distribution can not be negative.

The choice of the regularization parameter values is important. Several methods are available for choosing α, such as the L-curve method (Hansen, 1998) but they need a considerable amount of computation. We found from tests that our approach can provide satisfactory results for α=2.50E-07. Selecting a fixed value for α is the simplest method for choosing the regularization parameter but it is not optimal. The approach described above was implemented in MATLAB and several codes were developed for computing A_t in different numerical experiments.

4. Numerical experiments

[illegible]

Application of the approach to waste drums with homogeneous matrix using synthetic data

[illegible]

Application of the approach to waste drums with heterogeneous matrix using synthetic data

[illegible]

[illegible tables]

5. Experimental results

The performance of the new approach was also tested experimentally by measuring a 220 liter calibration drum filled with Portland cement and containing a linear 152Eu source. The calibration drum is provided with 3 hollow vertical tubes placed at different radial positions. The linear 152Eu reference source of length 62 cm was inserted into vertical tubes and the drum rotated during the measurements. The activity of the source was equal to 5.22MBq (1.06.2010) with a relative standard uncertainty of 2.5%. The density of the cement was determined by weighing and a value of 2.1 g/cm³ was obtained. An ISOCART gamma spectrometric system was used in the measurement of the calibration drum by integral gamma scanning. The system contains a SMART-1 HPGe detector type GEM 25P4, which is connected to a digiDART multichannel analyzer controlled by a laptop computer. ISOTOPIC 3.4 software was used for data acquisition and spectrum analysis. In all measurements the average dead time loss was smaller than 20% and counting times were chosen to achieve counting statistics better than 1%. Table 5 shows the values of the peak counting rate corresponding to two gamma lines of 152Eu determined for different detector and source positions. In the same table are also shown the measurement results obtained using both the new approach and the classical method. To compute the total activity A_t, the integral equation (1) was discretized using N=7x7=49 voxels and the values for the components of the vector $R_{p,i}$ were taken from Table 5. One can see from Table 5 that the new approach gives better results than the classical method at the expense of performing six measurements. Thus, the new approach gives errors of measurement smaller than 10% while the classical method has errors of up to 82 %.

6. Conclusion

[illegible]

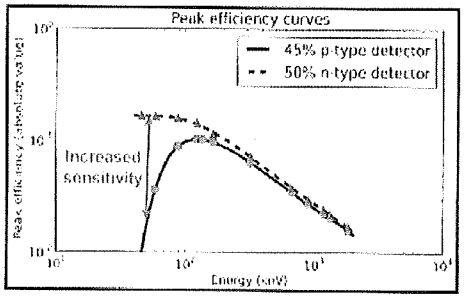
Technical note (P-107)

Simplified methods for coincidence summing corrections in HPGe efficiency calibration

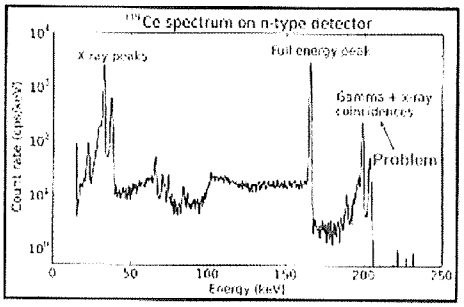
Alexander Mauring[1,*] and Jon Drefvelin[1]

Reasons for the project

Low-energy sensitivity of n-type HPGe detectors pose challenges with regards to true coincidence summing when calibrating for detector efficiency.



Corrections are needed when establishing peak and total efficiency curves. Commonly encountered "coincidence calibration nuclides": ^{57}Co, ^{60}Co, ^{88}Y and ^{139}Ce

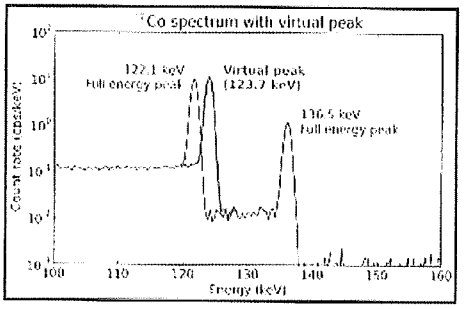


Simple peak addition is performed for the gamma-x-ray summing of ^{88}Y and ^{139}Ce:

Corrected peak efficiency:

Uncertainty:

Methods



A virtual peak is introduced to calculate the peak efficiency of ^{57}Co, as well as the total efficiency of ^{57}Co and ^{60}Co.

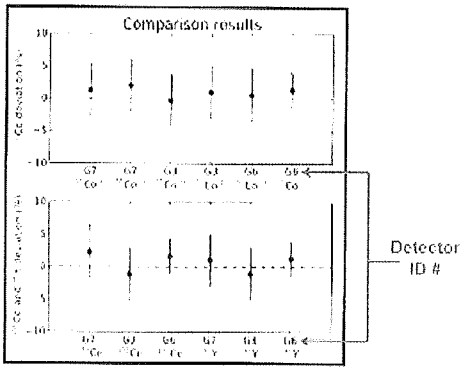
Virtual peak energy: $E_{v} = \frac{E_{57} \cdot p_{57} + E_{60} \cdot p_{60}}{p_{57} + p_{60}}$

Virtual peak efficiency: $\varepsilon_{v} = \frac{N'_{57} + N'_{60}}{A \cdot (p_{57} + p_{60})}$

Uncertainty:

Results

Correction factors vary between 0.93 and 1.17. Validation of the methods through comparative measurements of two independent reference solutions shows excellent agreement between nominal and calculated values.



Conclusion

The suggested corrections are simple, practical and successful in providing a solid foundation for subsequent activity calculations!

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ACTIVITY STANDARDISATION OF ^{45}Ca AND ^{204}Tl USING THE NEW TDCR SYSTEM AT CMI

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1. Introduction

The triple to double coincidence ratio (TDCR) method of liquid scintillation counting (Prochazka et al., 1988) was developed specifically for pure β- and EC emitters. As shown in reports of international comparisons during the last 10 years, the TDCR systems significantly contribute to the standardisation of beta and EC nuclides.

The new TDCR system was recently developed at CMI to replace or supplement previously used methods. A description of the mechanical set-up of the system and its electronics is provided in this paper, as well as results of the first measurements.

2. Mechanical set-up

The system consists of 3 steel tubes containing photomultiplier tubes. As shown in Figure 1, the tubes are fixed in place at angles of 120° to each other. In the centre of the system is a specially made optical chamber. The chamber is cylindrical, with three bores for photomultiplier tubes with diameters of 51 mm. The fourth hole at the bottom of the chamber is used to insert the sample. Interior surfaces are painted with reflective paint ($BaSO_4$, reflectance >90%). The vial (Perkin Elmer 20mL) is inserted from below with a piston; its distance from the front of the photomultiplier tube is minimised (ca 4mm). Insertion of sources is performed using a stepper motor, which allows for changes in efficiency by adjusting vertical positioning of the source in 0.5mm steps. Sources are changed automatically by the servo motor driven carousel.

The system is stored in a light-tight box with special entry ports, and the inner surfaces are painted matte black.

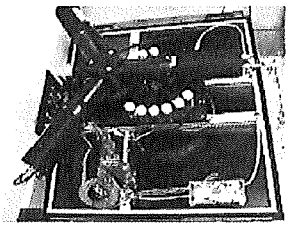

Fig. 1 The detail view of the TDCR system

3. Electronics

The photomultipliers (HAMAMATSU R8850, matched, similar quantum efficiencies, low dark current) are placed in a steel case with a rigid voltage divider and preamplifier developed at BQM. High positive voltage is applied to the anode to reduce noise. A computer-controlled transistor is used to adjust the focusing voltage along a range of 400-650V. The preamplifier located in the probe processes fast signals from the photomultiplier anode, the negative output pulses from the preamplifier are then fed to the HUB module, which provides separation of pulses for the MAC3 module (Bouchard and Cassette, 2000). The HUB module contains a delay circuit intended for setting of the analyzer in order to set discrimination of individual channels. Shaping amplifier handles the negative preamplifier output pulses in two different ways, for MAC3 and for the multi-channel analyzer. For MAC3 module, the pulses are only amplified x4, and the fast leading edge is maintained. For the pulse analyzer, the pulses are shaped suitably for MCA. This division to fast and slow branches allows a direct connection of MCA without an amplifier. MAC3 module processes the digital signal, and provides data necessary for activity calculation by the TDCR method.

Output signals from the logic module are connected to two NI 6602 counters, which are controlled by a computer via USB. The control program allows us to monitor the current state of the counter, and to make measurements with preset parameters (time, efficiency variation and multiplicity of measurements). Data can be saved in an ASCII file for further processing.

The computing program for activity calculation is based on TDCRD.2p application (Broda et al., 2000) and written in Visual Basic. The table of input parameters for efficiency calculation is continuously updated.

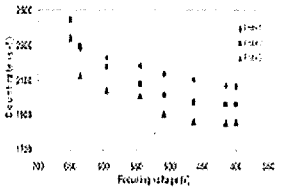

Fig. 2 Dependence of count rate on photomultiplier voltage. The behaviour of different probes varies.

4. Verification of the apparatus

Features and measurement capabilities were verified by measuring a set of ^{3}H sources. The sources (10mL volume in 20mL vial) are prepared from a weighed aliquot of active solution (cca 150mg), 0.9mL of distilled water, and a Ultima-Gold scintillator.

Three different method of efficiency variation were tested - defocusing from 400 to 650V in 30V steps, vertical positioning of the vial with in 2mm steps, and painting coloured bands on the vial.

Defocusing is a repeatable, very simple to use method. As shown in Fig. 2, the behaviour of individual probes varies, but influence of this effect on the resulting activity was not observed. The range of efficiency corresponding to the maximum attainable defocusing range was 0.44 - 0.58.

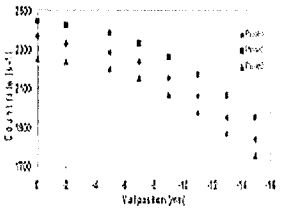

Fig. 3 Dependence of count rate on vertical position of the vial

Vertical positioning is very sensitive to the precise manufacturing of mechanical parts. Repeatability was tested by a measurement of 10 cycles using a set of 5 vials in 8 positions. No difference of measured values higher than 0.08‰ was observed. The range of efficiencies was 0.4 - 0.58, but it can be wider towards lower efficiency, which is useful for nuclides with higher β energies.

The coloured bands method needs no modifications of measuring device, but is time consuming and not repeatable.

Decrease in the measured count rate for all photomultiplier tubes depending on the position of the vial and number of coloured bands painted on the vial is shown in Figure 3 and 4 respectively.

The values of activity resulting from all measurements are in very good agreement and independent on method used for efficiency variation. The choice of method will therefore depend more on practical aspects of measurement of individual nuclides.

The specific activity of ^{3}H solution was determined with a combined uncertainty of 0.7% and was in good agreement (0.4‰) with values measured using a Tricarb 3100 TR calibrated with a commercial set of sources. The next step in validation of TDCR device was participation in BIPM comparison 2009 (CCRI(II)-K2.H-3).

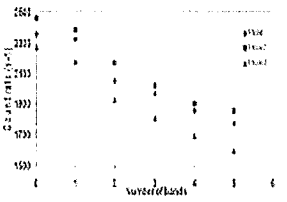

Fig. 4 Dependence of count rate on number of coloured bands painted on vial

5. ^{204}Tl and ^{45}Ca activity standardisation

The method still used at CMI for activity standardisation of ^{204}Tl and ^{45}Ca is the efficiency tracer method with ^{60}Co as the tracer. This method is relatively complex and time consuming, and includes significant intrinsic sources of uncertainty. Methods that use liquid scintillation counting are very promising for such types of nuclides, due to their simpler source preparation and measurement procedures. For this reason, CMI decided to test the feasibility of its newly built TDCR system as a replacement for the efficiency tracer method, perform activity standardisation of both nuclides using both methods and compare the results. The procedures, results and uncertainties are described in the following text.

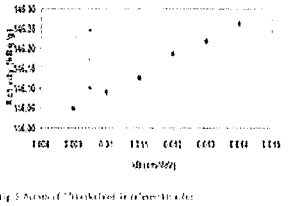

Fig. 5 Results of ^{204}Tl standardisation

5.1. Methods used

The TDCR measurements were performed to confirm the assumption that it is possible to replace the efficiency tracer method by TDCR method. Therefore it was necessary to verify the accuracy of input parameters and statistical models, as well as stability of samples.

The calibrated Tricarb 3100 TR device was used for control measurements.

The efficiency tracer method is based on the measurement of mixed sources of pure beta nuclide and tracer nuclide. It is essential that the tracer nuclide be easy measurable by the coincidence method, that both nuclides have similar spectra and that they are mixed homogeneously. The counting efficiency of pure β nuclide (ε_β) can be written as a function of β-inefficiency of tracer ($1-\varepsilon_{T\beta}$) (Campion et al., 1966):

$$\varepsilon_\beta = kf(1-\varepsilon_{T\beta}) \qquad (1)$$

The function can be approximated by a linear or quadratic function with sufficient accuracy.

The relationship between ε_β and $(1-\varepsilon_{T\beta})$ can be obtained by measuring mixed sources with different efficiencies. If the tracer activity is known from a separate measurement, it is possible to determine the count rate in β-channel attributed to pure beta nuclide, and activity can be calculated.

^{60}Co was used as the tracer for both nuclides, and the ratio of beta nuclide and tracer activities was approximately equal to one.

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5.2. ^{204}Tl activity standardisation

^{204}Tl decays by β⁻ unique first forbidden transition to the ground state of ^{204}Pb (97.08%) and by electron capture to the ground state of ^{204}Hg (2.92%). The maximum beta energy is 763.7keV, energies of K X-rays are in a range of 72.8 – 87.0keV and energies of Auger electrons are in a range of 5.1 – 14.8keV (Be et al.).

The original solution was diluted in two ways, one for the efficiency tracer method (approx. activity 220kBq/g, carrier solution 15mg/L TlCl + 0.1 M HCl) and one for LSC measurements (approx. activity 150kBq/g, carrier solution 35mg/L TlCl + 0.1 M HCl).

10 sources for LSC measurements were prepared using: 5 plastic and 5 glass 20mL vials. Aliquot (weight around 25mg) was added to 10mL of scintillator (Ultima-Gold) in each vial. The vials were measured repeatedly - immediately after closing, then after 24 and 48 hours. No significant decrease in calculated activity was observed.

TDCR method:

The efficiency was changed by defocusing with 8 counting points, and by vertical positioning (8 counting points 0-15mm). The maximum achieved efficiency was 0.978, the minimum was 0.96 for defocusing and 0.65 for vertical positioning. The activity calculations were made for values of kB in a range from 0.009 to 0.014. As shown on Fig. 5, the spread of results for different kB value was about 0.1%. For the final result, the value 0.012 was used because of the minimal dependence of resulting activity on the TDCR value. The influence of EC branch was accounted for, and the found ratio of EC and β branch efficiency was 0.021.

Efficiency tracer method:

The mixed solution was prepared using a ^{60}Co solution (20mg/L $CoCl_2$ + 3g/L HCl, approx. specific activity 200kBq/g), preliminarily standardised using the coincidence counting method, and a ^{45}Ca solution, with mixing ratio of 1:0.9 (^{60}Co solution : ^{45}Ca solution). The sources were prepared by depositing 15-25mg aliquots of mix solution onto conducting foil (gold coated VYNS foils ~ 40μg.g^{-1}) treated with Ludox and insalin. The beta - efficiency was varied using wet extrapolation method (Sochorova et al., 2008). As found previously, for sources with small amount of material (i.e. low carrier content (~20mg/L) and sufficiently enough specific activity (>200kBq/g)) no issue with inhomogeneity was observed. The quadratic efficiency function (1) for ε_β calculation was examined for ε_{11} in the interval (0.9 – 0.98), so both the branches β and EC are included.

The resulting activities and uncertainty budget for methods are shown in Table 1. Because two different solutions were measured, the resulting activity was recalculated using dilution factor. To avoid any gross errors, a control measurement using a calibrated Triearb 3100 TR device was performed, with a result of (146.0±1.5) kBq/g.

	TDCR	efficiency tracer
activity concentration (kBq/g)	146.11	146.45
uncertainty budget (%)		
counting statistics	0.1	0.1
dilution	0.05	0.05
weighing	0.01	0.01
background	0.05	0.1
dead time	0.01	0.03
source stability	0.1	
source homogeneity		0.2
tracer		0.2
input parameters and statistical model, quenching	0.4	
extrapolation of efficiency curve		0.7
half life	0.01	0.01
combined uncertainty	0.45	0.77
combined uncertainty (kBq)	0.62	1.1

Table 1. The results and uncertainty budget of ^{204}Tl activity measurements

5.3. ^{45}Ca activity standardisation

^{45}Ca decays by β⁻ unique single allowed transition to the ground state of ^{45}Sc (99.998%) (Lagoutine et al., 1986), the 0.0217 branch leading to the excited state of ^{45}Sc is negligible and ^{45}Ca can be considered a pure β-emitter with a maximum beta energy of 256keV.

10 sources for LSC measurements (in 5 plastic and 5 glass 20mL vials) were prepared from radioactive solution containing 20mg/L $CaCl_2$ + 3g/L HCl. Scintillator (Ultima-Gold) was added to 2 drops of solution (total weight around 25mg) and 0.5mL of carrier to obtain total amount of 10mL in each vial. The vials were measured repeatedly - immediately after closing, then after 24 and 48 hours. A slight decrease (0.15%) in count rates in all channels after 24 hours was observed, but no significant decrease in calculated activity.

TDCR method:

8 counting points with different efficiencies were measured. The efficiency was varied by defocusing (400-650V) and by vertical positioning (0-12mm). The maximum achieved efficiency was 0.952; minimum was 0.935 for defocusing and 0.69 for vertical positioning. After assessment of activity dependence on TDCR values for different kB, the kB value of 0.011 was used.

Efficiency tracer method:

The ^{60}Co solution (20mg/L $CoCl_2$ + 3g/L HCl) was diluted to achieve a specific activity of solution of approximately 135kBq/g, matching the preliminary determined activity of ^{45}Ca solution. The specific activity was determined using the coincidence method. The Co and Ca solutions were mixed with a 1:1 mass ratio.

The sources were prepared by depositing 25-40mg aliquots of mixed solution onto conducting foil (gold coated VYNS foils ~ 40μg.g^{-1}) treated with Ludox and insalin. The beta efficiency was varied using wet extrapolation method. Counting efficiency in β channel for ^{45}Ca was determined from efficiency function (1) for ε_{11} in a range of 0.8-0.95.

The resulting activities of tracer and TDCR method are shown in Table 2, the result of control measurement using Tricarb was (133.5±0.9)kBq/g.

	TDCR	efficiency tracer
activity concentration (kBq/g)	133.8	134.1
uncertainty budget (%)		
counting statistics	0.1	0.1
dilution	0.05	0.05
weighing	0.01	0.01
background	0.05	0.1
dead time	0.01	0.03
source stability	0.1	
source homogeneity		0.2
tracer		0.2
input parameters and statistical model, quenching	0.35	
extrapolation of efficiency curve		0.5
half life	0.01	0.01
combined uncertainty	0.40	0.59
combined uncertainty (kBq)	0.39	0.79

Table 2. The results and uncertainty budget of ^{45}Ca activity measurements

6. Conclusions

A new TDCR system was constructed at CMI and its features were verified by the ^{3}H solution measurement. Although comparison of efficiency variation methods showed good agreement in resulting activity for all methods, positioning of vial provides a wider range of efficiencies than defocusing method, is repeatable and can be operated automatically.

Two nuclides ^{204}Tl and ^{45}Ca were measured using TDCR method in order to prove its feasibility as a replacement of the efficiency tracer method. The procedures for samples preparation were verified and the stability of sources was tested. No sorption on the walls or diffusion was observed in either glass or plastic vials. Activities were calculated by the same code, which is based on the TDCR-2p code. The results were in good agreement. The uncertainty of TDCR method was almost twice as low as the uncertainty of the tracer method.

The TDCR system at CMI can be successfully used for standardisation of pure β nuclides with lower β energies because of its high detection efficiency (about 95% for ^{3}H and ^{45}Ca) and simple calculation model. Future efforts will focus on nuclides with complex decay schemes, and related extension of the software.