Development of a field portable tritium instrument - applicable detector technologies

Richard Marsh¹
(rm1202@noc.soton.ac.uk)
National Oceanography Centre, Southampton, SO14 3ZH

I Croudace¹, P Warwick¹ and R Greenwood²

¹University of Southampton / GAU-Radioanalytical
²AWE Aldermaston
Talk Outline

- Why develop a field portable tritium monitor?
- Problems associated with portable tritium measurement

-Approaches - possible detector types
  - Gas filled detectors
  - Scintillation based detectors
  - Solid-state detectors

- Potential sample handling techniques

- Summary - where to go next?
Why develop a field portable tritium instrument?

- Numerous applications for $^3\text{H}$ ⇒ set to increase?
  fission, fusion, research etc, decommissioning…
- Currently, a high percentage of samples are sent to third party labs:
  Sampling ⇒ Extraction ⇒ Measurement ⇒ reporting
  - Costly
  - Not in-situ
  - Time consuming

Aim:
To develop a ‘field-portable’ instrument capable of extracting and measuring tritium in a wide range of materials with detection limits of 10’s of Bq g$^{-1}$.
Problems associated with tritium monitoring

- $^3$H is a very weak pure beta emitter - $E_{\text{max}} \sim 18.6$ keV, $E_{\text{ave}} \sim 6$ keV
  
  $\Rightarrow$ such low energies require specialised equipment / techniques

- **Ideally:**
  Sample should be in intimate contact or VERY close to the detector (or inside in the case of proportional counters) - maximum range of $^3$H beta’s in air $\sim 1$mm or $1\mu$m in solids / liquids (Wampler and Doyle, 1994).

  Should be viewed with a $4\pi$ geometry to maximise efficiency.

  Detector should be designed with maximum tritium counting efficiency in mind - i.e. thin / no entrance window, low noise…

  Background should be as low as reasonably possible, including compensation for other radionuclides e.g. Radon and other mixed $\alpha/\beta/\gamma$

**In addition a portable detector must be light, robust, reliable…**
### Approaches - possible detectors:

-Detectors for $^3$H measurement can be broadly broken down into three categories:

| Gas Filled detectors - ion chambers, GM, GFPC |
| Scintillation detectors - solid and liquid |
| Solid state detectors |

All have advantages and disadvantages that should be considered!
Gas filled detectors for $^3\text{H}$

- **Ionisation chambers** - current, charge integration or pulse mode
- Low activities can be detected, the LOD will be a function of:
  
  Chamber volume, electronics, count time, background
- Memory affects - contamination resistant chambers

(Worth et al. 2005) Low internal surface area, $400 \text{ cm}^3$, fine wire electrodes tested to $51 \text{ Bq cm}^{-3}$

(Colmenares, 1974) Contamination resistant, no elastomeric material, $1000 \text{ cm}^3$, LOD of $18.5 \text{ Bq cm}^{-3}$

Additional ion chambers developed for tritium include: Pearson et al. 1991, and Weesner and McManus, 1988
### Commercially available portable ionisation based tritium monitors

<table>
<thead>
<tr>
<th>Model</th>
<th>Configuration</th>
<th>γ compensation</th>
<th>LOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Technical Associates PTG-7</td>
<td>2000 cm³ chamber including filter and de-ioniser on input</td>
<td>Sealed 2000 cm³ reference chamber</td>
<td>0.037 Bq cm⁻³</td>
</tr>
<tr>
<td>Overhoff SP1400DD</td>
<td>1400 cm³ chamber including filtered input, HTO/DTO option</td>
<td>1400 cm³ reference chamber</td>
<td>0.037 Bq cm⁻³</td>
</tr>
<tr>
<td>Sartrex 309</td>
<td>250 cm³ chamber including filtered input</td>
<td>Sealed 250 cm³ reference chamber</td>
<td>0.037 Bq cm⁻³</td>
</tr>
<tr>
<td>Premium Analyse βIonix</td>
<td>100 or 1000 cm³ chamber, 5 kg weight</td>
<td>Sealed 100 / 1000 cm³ reference chamber</td>
<td>0.0025 Bq cm⁻³</td>
</tr>
<tr>
<td>Lab Impex Systems H35L-P</td>
<td>2000 cm³ chamber including filter and de-ioniser on input</td>
<td>Sealed 2000 cm³ reference chamber</td>
<td>0.033 Bq cm⁻³</td>
</tr>
<tr>
<td>Canberra TAMD-73</td>
<td>2400 cm³ chamber including filtered input.</td>
<td>Sealed 2400 cm³ reference chamber, radon compensation</td>
<td>0.037 Bq cm⁻³</td>
</tr>
</tbody>
</table>

Richard Marsh - development of a field portable tritium monitor
Gas filled detectors for $^3$H (2)

- Proportional counters - internal gain, but usually require a gas supply
- Energy information - energy discrimination

(Aoyama et al. 1987) Air flow counter, $\gamma/\beta$ compensation by two guard counters. LOD of 0.037 Bq cm$^{-3}$

(Surette and Dubeau 2005) DGEM and PCB collector pads, $\gamma/\beta$ compensation. LOD 0.074 Bq cm$^{-3}$

Commercially available detectors include Raytest RAGA, Protean instruments MPC, Berthold LB 110, and Tech. Assoc. PTS-26.

GM - Seimiya et al. (1965) - ultra-thin polycarbonate windowed GM capable of measuring tritium
Scintillation detectors for $^3$H

- Solid or liquid scintillators, latter may offer higher efficiency, but creates a waste stream $\Rightarrow$ solid scintillators often re-usable.

- **Solid scintillant** tritium monitors:

  **Flow through (liquid)** - Rathnakaran et al. (2000), Falter and Bauer (1992) - ‘sponge’ filled cell formed from perforated scintillating film. LOD 37 Bq ml$^{-1}$ / 5 minute count.

  Gaseous - Arosio et al. (2000) - a personal monitor using a scintillating filter (Cerium-activated Y Al Perovskite powder) or plastic disc. LOD of 0.8 Bq cm$^{-3}$.

Others include Colmenares et al. (1974) and Ellefson et al. (1995).
### Commercially available solid scintillant $^3$H monitors

<table>
<thead>
<tr>
<th>Model</th>
<th>Configuration</th>
<th>Background &amp; E</th>
<th>LOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Technical Associates SSS-33M8</td>
<td>Flow through cell with scintillating micro-crystals and 2 PMT's</td>
<td>-</td>
<td>3.7 Bq ml$^{-1}$</td>
</tr>
<tr>
<td>Raytest RAMONA</td>
<td>Flow through solid / liquid cell radio-HPLC detector, 2 PMTs</td>
<td>Up to 20%</td>
<td>4 Bq ml$^{-1}$</td>
</tr>
<tr>
<td>Berthold LB 509</td>
<td>Flow through cell radio-HPLC detector</td>
<td>8-10 cpm, 8 - 50%</td>
<td>-</td>
</tr>
<tr>
<td>Mound BSD275</td>
<td>60 cm$^3$ sample chamber and CaF$_2$[Eu] crystal</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>Mound BSD133</td>
<td>20 cm$^3$ sample chamber and CaF$_2$[Eu] crystal</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>
Scintillation detectors for $^{3}$H (2)

-Liquid scintillant tritium monitors - usually designed around lab based systems made ‘portable’!

**Hofstetter et al. (1998/9) - ‘FDTAS’ based on automated sample / scintillant mixing system and Packard 525TR LSC. LOD of 6.4*10^{-3} \text{ Bq ml}^{-1} (100 \text{ min}, 0.025 cps bkd).**

**Huntzinger et al. (1984) - real time monitor based on a cross flow filter to collect samples from a sample stream and Radiometric Instruments Flo-One LSC. LOD 9.8 Bq ml^{-1} (20 \text{ min}).**
**Commercically available portable LSC for $^3$H monitoring**

<table>
<thead>
<tr>
<th>Model</th>
<th>Configuration</th>
<th>Background / E</th>
<th>LOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hidex Triathler</td>
<td>Single vial manual LSC with single PMT and optional shielding</td>
<td>Tritium efficiencies of $&gt;$45%</td>
<td>$\sim 25$ Bq ml$^{-1}$</td>
</tr>
<tr>
<td>Raytest Malisa Star</td>
<td>Single vial manual LSC with shielding option</td>
<td>-</td>
<td>As low as 0.1 Bq ml$^{-1}$</td>
</tr>
<tr>
<td>Technical Associates SSS-12</td>
<td>Single vial manual LSC with single PMT and optional shielding</td>
<td>Energy discrimination for noise and background reduction</td>
<td>-</td>
</tr>
<tr>
<td>Technical Associates SSS-22P</td>
<td>Single vial manual LSC specifically for tritium, dual PMT, shielding</td>
<td>Tritium efficiencies of $&gt;$60%</td>
<td>-</td>
</tr>
<tr>
<td>Lumi-scint DOE</td>
<td>Drawer loading single vial LSC.</td>
<td>Threshold based noise reduction</td>
<td>$\sim 0.9$ Bq ml$^{-1}$</td>
</tr>
<tr>
<td>IN/US $\beta$-RAM</td>
<td>Flow through cell radio-HPLC detector</td>
<td>$&lt;3$ cpm, $&gt;$50%</td>
<td>-</td>
</tr>
</tbody>
</table>

Other counters not designed specifically for tritium may also be applicable such as the Ballard MicroCount.
Solid-state $^3$H monitors

- Usually based on PIN photo-diodes or Avalanche photodiodes (APD’s - offer the benefit of additional internal gain).
- McGann et al. (1988) - first to really investigate use of diodes for tritium detection - APD system with an LOD of 33 Bq cm$^{-2}$ (500 s count).

Wampler and Doyle (1994) - Hamamatsu PIN diodes used in portable and bench-top monitors. LOD of 10 Bq cm$^{-2}$.

Scott Willms et al. (2005) - utilised a large area APD from Radiation monitoring devices Inc. LOD of 0.17 Bq cm$^{-2}$ (96 hr count!).

Additional detectors include Surette et al. (2007) who have developed a PIN diode based dosimeter and Shah et al. (1990).
A large range of detectors have been developed for tritium monitoring. Examples of all three types may offer suitable detection limits, however:

- Both proportional counters and LSC systems require consumables, and in some cases create additional waste.
- Proportional counters that utilise air are both susceptible to variations in T, P and humidity and maybe fragile in construction.
- Ionisation chambers are prone to memory effects, particularly at low levels, and require large sample volumes.

As a result, the most suitable detector types to pursue maybe either solid scintillators or solid-state detectors as they are potentially robust, compact and reusable.
Acknowledgements

The author is grateful to AWE for an Outreach award to support this project.


References


