WORKPLACE MONITORING TECHNIQUES FOR TRITIUM

Kirsty Higham

AWE plc., Aldermaston, Reading, Berkshire, RG7 4PR, UK.

ABSTRACT

Investigations have been carried out into various tritium monitoring techniques for low level surface contamination taking into account operational factors.

Comparisons were made of various smearing media and analysis techniques. As a result, changes have been made to the smearing materials used. Additionally, different analysis techniques have been shown to be preferable for different applications. These techniques can be optimised to balance an accurate result with an operationally acceptable timescale which can be adapted for different applications where varying accuracies and limits of detection may be required.

The effects of operational factors (physical nature of contamination, delays in counting smears, smearing techniques, counting environment, etc.) were also evaluated to identify potential sources of error. These are eliminated where possible and quantified where not.

The results of this work have enabled provision of an optimised range of monitoring services together with guidance on the accuracy and limitations of each technique.

INTRODUCTION

Tritium causes quite complex contamination problems due to its physical and chemical properties. It will displace ^{1}H atoms in most hydrogenous materials, is small enough to diffuse through almost any substance and can be absorbed to some depth in surfaces. Areas which are regularly cleaned can therefore become re-contaminated as tritium molecules diffuse back to the surface from absorbed contamination.

Tritium is a very low energy beta emitter (18.5 keV max.) with a range of ~ 0.36 mm in air and ~ 0.18 μm in aluminium. Monitoring with standard windowed probes is therefore impossible and monitoring must instead be carried out by a method in which the sample is in direct contact with the sensitive volume of the detector. Whilst monitors are available which measure surface contamination levels directly, based on a windowless gas proportional detector which is clamped onto the surface to be monitored, these have currently been discounted for routine use due to a number of practicable considerations. The most widely used technique is therefore smear sampling. A small circle of material is wiped across the surface to be monitored. The activity on the smear is then measured and this is related to a surface contamination level by knowledge of the area smeared and the pick up factor (fraction of the total surface activity removed by the smear). The activity on the smear must be again measured by a suitable technique; the most commonly used methods are liquid scintillation counting, where the smear is placed directly into liquid scintillant, or by windowless gas proportional counting.

The reliance on smear counting, which is an inherently less reproducible technique than direct monitoring with a probe and which has numerous potential sources of error (pick-up factor, area smeared, etc.), coupled with the technical difficulties in accurately measuring low activity emissions at low levels and the complex physical nature of tritium contamination combine to make tritium surface contamination monitoring a relatively uncertain process. This ongoing study has therefore been carried out to identify principle sources of error, to eliminate these where possible or else to quantify the error and ultimately to provide an optimised range of monitoring services together with guidance on the accuracy and limitations of each technique.

SELECTION OF MONITORING TECHNIQUE

At the start of this study two monitoring options were available - analysis of foil smears by gas proportional counting and analysis of glass fibre (GFA) smears by liquid scintillation. Both had their drawbacks; foil smears scratched surfaces and were considered to give poor results and GFA smears were fragile and so were limited to smooth surfaces. Additionally the two were not interchangeable so that foil smears could not be re-analysed by liquid scintillation if desired.

A large study was carried out to compare results from three smearing media (GFA, foil and the paper smears routinely used in monitoring other contaminants) and two available analysis techniques (gas proportional counting and liquid scintillation). As a result paper smears were chosen for all monitoring tasks for their combination of accuracy and robustness. Liquid scintillation was shown to give a more accurate result and lower limit of detection versus the quicker but less precise technique of gas proportional counting. Comparison of the two led to derivation of a self-absorption correction factor for gas proportional counting to bring the results in line with those from liquid scintillation analysis.

Currently therefore both gas proportional and liquid scintillation analysis are used, the choice being dependant on the required balance between speed and accuracy. Typically gas proportional counting will be used during direct coverage of operations, decontamination work, etc. whereas clearances, monitoring of transport containers, etc. will be analysed by liquid scintillation.

A further analysis technique has recently become available. Flame oxidisation fully combusts organic samples (e.g. smear papers) and collects the activity as tritiated water which is analysed by liquid scintillation. Whilst the technique is too costly and time consuming to be considered for routine monitoring purposes it could be used experimentally to accurately determine the efficiency of the other two techniques.

SMEAR PICK UP FACTOR

In any smear monitoring technique the pick up factor (proportion of activity on the surface transferred to the smear) is essential to interpretation of the results. This is always a difficult and variable quantity, being dependant on a number of factors including smearing media, nature of contamination and surface, and applied pressure. With tritium there are further considerations: are we trying to relate activity on a smear to the ‘loose’ fraction of activity, the total activity present on and in the surface, or for example the fraction of total activity able to be absorbed through the skin and so

contribute to occupational exposures (a value probably between the other two). The first of these is the easiest to derive a pickup factor for operationally, but even this is complicated by the fact that removed surface contamination can be fairly quickly replaced from the absorbed contamination. Attempts were made to derive an operational pickup factor by repeated resmearing of the same area (subsequent smear activities should reduce by a factor equivalent to the pickup factor) but the results were wholly inconclusive. In the absence of other information therefore the industry default of 10% is used but it is recognised that this will contribute the largest (and least quantifiable or avoidable) error on the results.

ANALYSIS ERRORS - GAS PROPORTIONAL COUNTING

A number of factors have been observed operationally or reported in literature which may affect the uniformity of results. These have been investigated where possible and either disproved, quantified or eliminated as appropriate. Significant points include:

Ambient conditions, particularly temperature and humidity, can have a significant effect on the efficiency of the detector. In areas with no controlled environment, instrument efficiency checks are carried out repeatedly throughout the day to quantify this; operating limits have been calculated to give acceptable counting statistics. Similarly moisture on the smears can have a significant effect on the results; damp smears must be analysed by liquid scintillation rather than gas proportional analysis.

Tritium loss during counting is a reported problem but trials involving repeat counting of smears have shown no such effect. The length of time between taking and analysing smears (simulated by taking and counting a smear immediately and then recounting after a time) had a measurable but inconsistent effect on the result and so merely increases the error on the result as it cannot be modelled and accounted for.

It was noted on occasion that repositioning and recounting a smear could give a different result, leading to the suspicion that the sensitivity of the counting chamber was not uniform. The design of the chamber, with unidirectional wiring, made this plausible. This is significant as the activity on a smear is unlikely to be uniformly distributed so the results will be affected dependant on whether the bulk of the activity lies in the more sensitive areas of the detector. Studies have failed to reproduce this effect with any consistency and so the consequence is difficult to quantify. The effect can be controlled by inserting the smears in a uniform direction.

ANALYSIS ERRORS - LIQUID SCINTILLATION COUNTING

The liquid scintillation detectors are far more accurate than the gas proportional counters, both in terms of influence by external factors and in pure counting statistics.

A major potential source of error is opacity of the sample as scintillation counters work on the production and detection of light photons in proportion to activity. The liquid scintillation counters used have a built in function which assesses and compensates for this opacity by use of an external source standard. More opaque samples do however give worse counting statistics (as less ‘events’ (photons) per Bq are measured).

A major factor in interpreting results is the time taken for activity to leach from the smear into the scintillant. Effective detection requires the tritium to be in intimate contact with the scintillant; whilst it remains on the smear self-absorption occurs. Repeated counting of smears show that the detectable activity increases over several days, to up to twice the initial measured activity. There is a wide spread of patterns (see Figure 1) with some smears showing a large increase is measured activity with time and some very little; but the majority show a similar pattern.

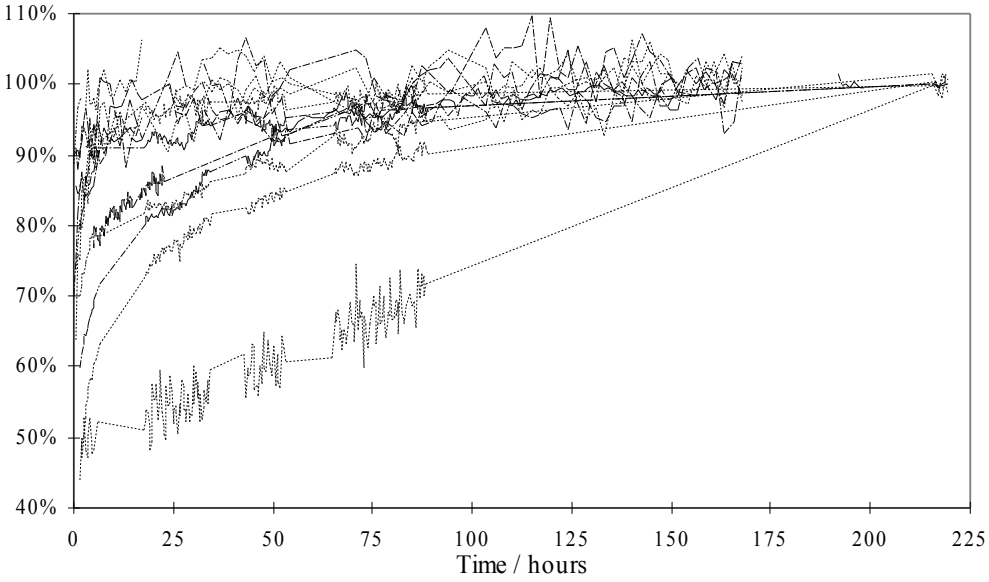

Figure 1 - Relative increase in measured activity of smears over time

To aid early interpretation of the results a representative 'correction factor' is therefore applied to predict the final activity. The correction factors are conservative (i.e. will tend to overpredict activity) and become more accurate the longer the smear has had to soak. If an early count of a smear gives a result slightly higher than the desired value the customer may opt to recount the smear a few hours later rather than spend potentially unnecessary time decontaminating.

CONCLUSIONS

A number of factors which could influence workplace tritium contamination monitoring were investigated. The outcome was a change to more appropriate monitoring techniques and an improved understanding of the sources and magnitude of counting errors. Changes have been made to monitoring regimes to minimise these errors where possible; otherwise their effect has been quantified.

A range of monitoring techniques, including gas proportional smear counting and progressive interpretation of liquid scintillation analysis, can now be offered together with guidance on the speed, accuracy, limit of detection and interpretation of each method. This allows selection of the appropriate technique to optimise accuracy within available timescales and improves customer understanding and confidence in the results.