# OPTIMIZATION OF THE SMEAR SAMPLING METHOD FOR EVALUATION OF TRITIUM CONTAMINATION ON METALLIC SURFACES

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*Abstract*

The paper presents the results of the experimental tests for determination of performances of the methods used in evaluation of the surface contamination of metallic objects. The tests have been designed to allow the determination of the critical parameters considered for the radio-analytical process, such as: the source efficiency in 4π geometry Liquid Scintillation counting (LSC), the sampling factor for the smear test applied to different types of surfaces and the percent activity recovery when the sample distillation method is used to eliminate the matrix and other radionuclide interferences. During experimental activities, practical methodologies have been developed which may serve as basis for designing optimized test procedures based on the smear sampling method for evaluation of tritium contamination on metallic surfaces.

***Keywords:*** *Smear sampling method, Surface contamination, Tritium*

**Introduction**

The safe operation of nuclear installations depends essentially on the performance of the monitoring systems used. The personnel of these units must be informed about the quality of environmental factors and must be aware of the hazards in the installation so that in the event of anomalies protective measures can be applied or corrective action taken to limit the exposure of workers and the environment. Site monitoring for determination of air and surfaces contamination is important to prevent accidental intake of contaminated material by personnel. It is also important that when leaving the controlled area of the installation, any object or equipment to be monitored for radiological clearance [1].

Tritium contamination of surfaces is a common problem of nuclear and/or radiological installations whose operation involves handling of materials with high tritium content (eg HWR, Tritium removal facilities etc). Determination of tritium contamination of surfaces is a basic tool of contamination management, being an important component of the radiation protection program in such a facility [2].

Tritium in the form of Tritiated Hydrogen, HT, or Tritiated water, HTO, is easily fixed to the surfaces of metal objects, plastic or rubber, by adsorption phenomena. In the case of metal objects, tritium remains well fixed in the vicinity of the surface, as long as the material is not heated to high temperatures. At room temperature, the permeation of tritium in such materials is extremely slow.

Determining tritium contamination of surfaces is a complex issue as the available techniques most often provide discrepant results. This is largely due to the peculiarities of this type of contamination. Thus, since tritium is a low energy beta-emitting radionuclide from the point of view of radiation protection, only the unfixed component of contamination is significant. Total contamination, which includes both the fixed and the non-fixed component, is, in most cases, a quantity that cannot be determined directly, as it requires the destruction of the sample. Unfixed contamination can be estimated by smear sampling, but the range of uncertainty is quite wide due to the high variability of the sampling factor. The results of the direct measurement represent a fraction of the total contamination, being an image of the tritium activity in the immediate vicinity of the surface.

According to ISO 7503-1 [3], loose contamination is defined as that part of the contamination that can be removed or transferred under normal working conditions. The normal working conditions consist, as shown in the said standard, in the normal, non-accidental contact of the human body (protected or not protected by clothing) with the surfaces and parts of equipment handled directly by a worker.

The United States Nuclear Regulatory Commission (NRC) defines [4] loose contamination as: "*radioactivity that can be transferred from a surface to a paper smear by rubbing under moderate pressure*." The US Department of Energy also defines [5] loose contamination as: "*radioactive material that can be removed from a surface by non-destructive methods such as occasional contact, wiping, brushing or washing*".

Smear sampling is used to determine loose contamination, while total contamination can be determined by direct measurement with special monitoring equipment. When the contamination determined by direct measurement is below the investigation limit for loose contamination, the separate determination of this component of the contamination is not justified. However, even if the determination of loose contamination may seem irrelevant, there may be circumstances in which this may be required.

Thus, when direct determinations cannot be made due to the low sensitivity of the equipment to the contaminant being targeted, as is the case with low-energy beta-emitting radionuclides, the only way to check for contamination is by smear testing. On the other hand, there is a significant risk of not detecting contamination, where the determinations are based exclusively on the indirect method, especially when direct measurements can be made, a risk due to the high inaccuracy of the indirect method and the high limit of detection.

The use of smears to determine surface contamination seems to have originated during the *Manhattan Project* in World War II, and was then used to monitor workspaces for the presence of plutonium contamination. At that time, the lack of portable equipment led to the need to improvise.

A smear sample is obtained by rubbing the surface to be characterized with a dry or moistened piece of material (paper or fabric). In general, a single smear is not enough to collect the entire loose contamination from the tested surface.

In order to express the ease with which the radioactive material is removed by erasure from the contaminated surface, the sampling factor $F$ is defined as the fraction between the activity taken on the smear and the total unfixed activity present on the contaminated surface before the test.

$$F = \frac{A_1}{A_T}$$

where: $A_1$ is the activity taken on the initial smear, $A_T$ is the total non-fixed activity obtained by summing the activities taken by several successive smear tests, $A_T = \sum_i A_i$

The sampling factor is determined experimentally for each set of measurement conditions, using successive sampling methods until the total removal of the unfixed activity. If it is not possible to experimentally determine the sampling factor, it is recommended to use a conservative value of 0.1. Depending on the known information on characteristics of the surface to be characterized, it is possible that the use of the generic sampling factor of 0.1 introduces a very high uncertainty on the measurement result, which may not be reported with the result.

Total efficiency in measuring the surface contamination is considered as the result of the product of two factors: the efficiency of the measuring instrument ($\varepsilon_i$) and the efficiency of the source ($\varepsilon_s$). The efficiency of the measuring instrument is defined as the fraction between the response of the measuring instrument and the emission rate of a source in a specified detection configuration.

Source efficiency is defined as the ratio of the source emission rate to its activity. This quantity reflects the effects of phenomena that occur at the source and can affect the detection of radiation emitted by it, such as self-absorption and backscattering. In the case of detection techniques in 4π geometry (LSC) the efficiency of the source is an essential parameter.

From the above it is observed that the main parameter that affects the interpretation of the results of the determination of loose contamination is the sampling factor. Although universally accepted, the fact that only a fraction of the contamination is taken at a first smear pass is not, however, often realized in practice. Some of the parameters that control the value of this parameter are: the chemical form of the contaminant and how it interacts and is retained by the surface of the contaminated material. They mainly affect the fixed component of the contamination, but the chemical form of the contamination can affect the decontamination factor by the way it depends on the sampling process (wet or dry).

The sampling factor depends on the extent to which the contaminant has an affinity for the contaminated surface or the sampling material. Tests presented in the literature show that, in the case of tritium contamination of surfaces, the effectiveness of the smear sampling method depends on the solubility of tritium compounds relative to the solvent used to wet the smear. Also, the experimental data show that among the factors affecting the measurement results of the transferable activity are: the chemical composition of the contaminant and solvent, the state of aggregation, the sampling, the particle size of the contaminant and the extent to which it adheres to the contaminated surface. The characteristics of the contaminated surface (composition, roughness, etc.) are also factors that may influence sampling.

Experimental tests presented in the literature [6] show that in order to obtain a correct estimate of the entire unfixed activity, up to ten successive smear passes are required, even in the case of well-finished metal surfaces. The variability of the smear test results, even in the case of well-controlled experiments, is an order of magnitude above the level of statistical uncertainty, due to the uncertainty of the sampling factor.

The ISO 11929-7 standard [7], in the calculation example for the detection limit when measuring contamination by the smear test, proposes the use of a rectangular distribution with an average of 0.34 and a range of 0.6 - 0.62 for the sampling factor. There are authors which propose to model the fraction of loose contamination by using an asymmetric triangular distribution in the range 0-1 with a maximum of 0.1. The sampling factor can be determined experimentally by using the method of ISO 7503, consisting of repeated wiping tests until complete removal of the contamination.

**Experimental tests**

The tests were performed using a tritiated water solution having a tritium activity concentration of 2.54 × 106 dpm/g at October, 5, 2000). The solution concentration was checked by LSC, applying the external standard method. At the same time, validity of the calibration curve used was determined, calculating corresponding correction factors.

To determine the source efficiency, three types of sampling materials and two types of liquid scintillators were selected, aiming to determine the recovery fraction when determining the activity. Thus, smears, with a diameter of 4 cm, made of filter paper, textile and cellulose nitrate were used for testing, being placed in polyethylene scintillation vials. A controlled amount of tritiated water solution was dosed on each smear. Different liquid scintillators (UltimaGold XR and Filter Count, respectively) were added to the scintillation vials. The activity of each vial was determined by using a TriCarb4910TR LSC analyser. The source efficiency was calculated as the ratio between the measured activity and the calculated activity.

The activity recovery factor for the simple distillation step of the samples with added water was determined by performing three tests, as follows:

- *Distillation test - 1*: A paper smear was placed in a round-bottomed flask on the surface of which it was gravimetrically measured a predetermined amount of tritiated water solution (< 0.1 g), then 20 ml of distilled water was added. The flask was connected to a distillation installation. The obtained distillate was weighed and mixed with a predetermined amount of UltimaGold AB scintillator, after which its tritium activity was determined by LSC.
- *Distillation test - 2*: It proceeded similarly to test 1, but the amount of distilled water added was 50 ml.
- *Distillation test - 3:* The procedure was similar to test 2, but the filter paper smear was moistened with distilled water before dosing the tritiated water solution, with the aim to determine the sampling method (wet / dry) effect on activity recovery factor.

As shown above, in the case of tritium contamination, the sampling factor strongly depends on the nature of the contaminant, its chemical form and its solubility in the solvent used in sampling. It is obvious that a test to determine a generally valid sampling factor, in the case of tritium contamination of metal surfaces, is not feasible, especially since the simulation of a persistent tritium contamination is very difficult. However, in order to gets an idea of the performance of the smear sampling method, in the case of tritium contaminations in the form of tritiated water, two tests have been performed and are presented in the following.

As support, two metal plates were used, one made of stainless steel, with a well-finished surface and the second one was made of aluminium with a very rough surface. The two supports represent, in the authors' view, the extremes of the types of metal surfaces (susceptible to contamination in a radiological installation). First support was made of a chemically inert material and has a high degree of finishing (easily mechanically decontaminable). The second one was made of a more chemically reactive material and has a large pore surface due to high roughness. Controlled amounts of tritiated water solution were deposited on the two metal surfaces, thus simulating a tritium contamination. The surfaces were wiped with filter paper smears moistened with distilled water, and the sampled activity was measured by LSC. The sampling factor was determined as the ratio between the activity taken at the first pass and the total activity submitted.

**Results and discussions**

Table 1 presents the results obtained by measuring the activity concentration of the solution using Ultima Gold XR and Filter Count scintillators, scintillators used in subsequent tests.

Table 1. Results for validation of the calibration curve for three types of liquid scintillators

| **Parameter** | **Ultima Gold XR** | **Ultima Gold AB** | **Filter Count** |
|---|---|---|---|
| Solution mass [g] | 0.0197 | 0.0451 | 0.0219 |
| Calculated activity [Bq] | 378 | 865 | 420 |
| Count rate [cpm] | 5480 | 15233 | 11296 |
| Efficiency | 0.261 | 0.326 | 0.441 |
| Measured activity [Bq] | 350 | 779 | 427 |
| Activity recovery [%] | 93 | 90 | 102 |

As can be seen, in the case of LSC, by using Ultima Gold XR and Ultima Gold AB scintillators, the results obtained underestimated the concentration by 7% and 10%, probably due to colour extinction as a consequence of the degradation of the scintillators used. Consequently, the efficiency correction factors were calculated to compensate for this effect, which were used in subsequent measurements performed with these scintillators.

To determine source efficiency, the tests were performed using combinations of scintillator and absorbent materials (smears), and the results are presented in Table 2.

The activity recovery rate represents the source efficiency, being given by the ratio between the activity determined by LSC (using the calculated efficiency for homogeneous liquid source) and the activity calculated based on the dosing of the tritiated water solution.

Table 2. The results of the tests for determination of the source efficiency

| **Parameter** | **Paper filter** | | **Fabric** | | **Cellulose nitrate membrane** |
|---|---|---|---|---|---|
| | **UG XR** | **FC** | **UG XR** | **FC** | **UG XR** |
| Solution mass [g] | 0.0227 | 0.0201 | 0.0181 | 0.0270 | 0.0269 |
| Calculated activity [Bq] | 435 | 385 | 347 | 518 | 516 |
| Count rate [cpm] | 4636 | 8104 | 2284 | 10005 | 6747 |
| Efficiency | 0.241 | 0.422 | 0.239 | 0.416 | 0.245 |
| Measured activity [Bq] | 346 | 320 | 172 | 401 | 496 |
| Activity recovery [%] | 79 | 83 | 50 | 77 | 96 |

As it can be seen, the source efficiency (measured immediately after preparation) when using the Filter Count scintillator is systematically higher than those with the Ultima Gold XR scintillator, which may be due to the lower wettability of the Ultima Gold XR scintillator as related to the sampling material. In the case of cellulose nitrate smears, source effectiveness is practically unitary, because both scintillators have the ability to completely dissolve this type of material.

The tests of distillation of the smear samples, by adding water and entraining vapours in a stream of nitrogen allowed the determination of the activity recovery factor by applying this process of elimination of interferences. Table 3 shows the results obtained in the three distillation tests considered for this work.

The measured activity and the recovery ratio of the activity were calculated taking into account the correction fraction due to sampling.

As one can see, in distillation test 3 (wet sampling simulation) the result of the smear activity is quasi-quantitative, while for distillation tests 1 and 2 the results indicate a slight underestimation of the activity, probably due to an "immobilization" of the contaminant in the smear matrix.

To determine the sampling factor, dry and wet sampling tests were performed using two types of support for simulating contamination: stainless steel with a well-finished surface and aluminium with a rough surface. For each of the supports, tritium contaminations were simulated by dosing tritiated water on the surface. In the case of wet sampling tests, two successive smear samples were taken, which were measured separately. The test results are presented in Table 4.

The sampling factor was calculated as the activity recovery ratio corrected for source efficiency.

As one can see, both tests showed that the best sampling ratio is obtained in the case of dry sampling, because the nature of the contamination (the presence of liquid on the contaminated surface) favours its fixation on the smear, by absorption in its porous medium.

Table 3. Results of the tests by using distillation of samples with added water

| **Parameter** | **Distillation test #1** | **Distillation test #2** | | **Distillation test #3** |
|---|---|---|---|---|
| | | **Et. 1** | **Et. 2** | **Et. 1** |
| Solution mass [g] | 0.0568 | 0.0605 | 0.0605 | 0.0532 |
| Calculated activity [Bq] | 1088.8 | 1159.7 | 1159.7 | 1019.8 |
| Added water quantity [g] | 19.3 | 50.6 | 50.6 | 50.6 |
| Collected water quantity [g] | 7.8 | 8.5 | 8.6 | 8.2 |
| Mass fraction | 0.40 | 0.17 | 0.17 | 0.16 |
| Count rate [cpm] | 5680 | 2812 | 3012 | 2657 |
| Efficiency | 0.290 | 0.293 | 0.299 | 0.309 |
| Measured activity [Bq] | 899 | 1056 | 1092 | 982 |
| Activity recovery [%] | 83 | 91 | 94 | 96 |

Table 4. Results of the tests for determination of the sampling factor

| **Parameter** | **Stainless steel support** | | | **Aluminium support** | | |
|---|---|---|---|---|---|---|
| | **Dry** | **Wet 1** | **Wet 2** | **Dry** | **Wet 1** | **Wet 2** |
| Solution mass [g] | 0.0836 | 0.0604 | | 0.0504 | 0.0566 | |
| Calculated activity [Bq] | 1600 | 1156 | | 965 | 1083 | |
| Count rate [cpm] | 30436 | 19004 | 1122 | 15646 | 16356 | 2335 |
| Efficiency | 0.415 | 0.407 | 0.402 | 0.413 | 0.389 | 0.385 |
| Measured activity [Bq] | 1222 | 778 | 47 | 632 | 700 | 101 |
| Activity recovery | 76% | 67% | 4% | 65% | 65% | 9% |
| Sampling factor | 92% | 81% | 5% | 79% | 78% | 11% |

**Conclusions**

The analysis of the data in the literature led to the conclusion that the only way to assess the tritium contamination of surfaces is to take smears and count LSC. Although manufacturers of radiation measuring equipment have been identified that offer portable devices to determine total tritium contamination of surfaces, data from other authors show that due to the low energy of tritium radiation, self-shielding can play a key role in the process, leading to a strong underestimation of the measured contamination.

Consequently, practical methodologies for sampling have been established in order to estimate the total contamination and the non-fixed contamination of metal surfaces. Also, to eliminate interferences in the measurement process, the proposed radioanalytical methods include steps of distilling the tritium present in the form of tritiated water.

The experimental tests performed allowed the determination of some critical parameters for the considered radioanalytical process, such as the efficiency of the source for LSC metering in 4π geometry, the sampling factor for the smear test applied to some types of metal surfaces and the efficiency of recovery activity by distillation with the addition of water and entrainment of vapours in a stream of nitrogen.

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