2009 International Nuclear Atlantic Conference - INAC 2009
Rio de Janeiro,RJ, Brazil, September27 to October 2, 2009
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
***ISBN:* 978-85-99141-03-8**

WIPE SAMPLING FOR CHARACTERIZATION OF NON-COMPACTABLE RADIOACTIVE WASTE

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ABSTRACT

Wipe sampling is a method of monitoring radioactive surface contamination on working area and on radioactive, non-compactable wastes, constituted of large pieces of replaced parts of equipment in nuclear and radioactive installations. In this method, sampling is executed by rubbing a disc of filter paper on the contaminated surface in such a way as to collect entirely or partially the deposited material. The target radioisotopes are subsequently measured directly on the wipe or extracted by appropriate radio analytical methods and then qualitatively and quantitatively determined. The collection factor, or the efficiency with which the material is removed from the surface and deposited on the smear, is the main source of error in quantitative measurements. The determination of the collection efficiency is the object of this communication.

1. INTRODUCTION

The Radioactive Waste Management Laboratory (RWML) at the Nuclear and Energy Research Institute (NERI), in São Paulo, Brazil, develops methods for characterization of radioactive raw waste. One of these wastes is non-compactable, solid wastes originated in the maintenance and decommissioning of nuclear facilities. This waste is composed mainly of replaced parts contaminated on the surfaces with the radioisotopes handled in the facility where they originated. The aim of this work is to estimate the radioisotopic inventory of the wastes for regulatory and operational purposes.

The use of direct methods to measure surface contamination, for instance pancake probes with Geiger-Muller detectors or large area proportional counters, is limited in many cases because background radiation raises detection limits much above regulatory limits, because of too large inaccuracies in quantitatively detecting alpha-emitting contaminants, or other drawbacks. Indirect methods, with wipe samples are used instead. Alternative methods with passive radiation detectors like TLD dosimeters, track detectors, or electret-type ionization chambers [1] are being proposed but are too expensive or too time demanding to be of practical use in the routine of radioactive waste treatment.

The wipe sampling method is executed by rubbing a disc of filter paper on the contaminated surface in such a way as to collect entirely or partially the deposited material. The target radioisotopes are subsequently measured directly on the wipe or extracted by appropriate radio analytical methods and then qualitatively and quantitatively determined.

The wipe sampling is a method routinely used for surface contamination control in occupational radioprotection in the nuclear industry, in nuclear medicine and in radioisotope research laboratories mainly as a qualitative or semi-quantitative method.

Results of quantitative analyses of surface contamination based on wipe sampling are affected by large inaccuracies because of uncertainties on the fraction of the deposited material that is transferred to the wipe, what is called the transfer factor error.

Different values of the transfer factor are reported for routine use. The value used by the health physics personnel of NERI is 0.1. The International Atomic Energy Agency recommends the same value [2]. The International Standardization Organization (ISO) [3] lists three different values depending on the material of the wiped surface: for glass, stainless steel, and other smooth surfaces the recommended transfer factor is 0.5; for wood and masonry the factor is 0.05; for other surfaces 0.1 is adopted. The operation manuals of some manufacturers of radiation detection equipment recommend values between 0.1 and 1.0 depending on the material of the wipes, the material of the sampled surfaces, and on other variables [4, 5].

Experience of RWML staff shows that the transfer factor can be much lower when sampling surfaces that stood contaminated for long times, that is the case of some radioactively, surface contaminated wastes, like radioactive lightning rod metal scrap contaminated with ^{241}Am oxide.

In the present paper we present some results of the ongoing research to select a method of wipe sampling that yields accurate results to characterize radiologically contaminated non-compactable wastes.

2. METHODS AND RESULTS

Wipe sampling was according to ISO standard [ISO-7503] but also followed prescriptive rules for internal use of some organizations [6, 7, 8].

To produce contaminated surfaces to smear, flat 25x25 cm sheets of stainless steel, carbon steel, painted carbon steel, acrylic glass, and vinyl flooring tiles were purposely contaminated with americium oxide dust inside a glove-box used for dismantling radioactive lightning rods. Care was taken to spread the radioactive dust as smoothly as possible onto the surfaces and to remove large free-falling particles from test surfaces to avoid or minimize the risk of contamination around the laboratory bench.

A previously drawn grid on the surfaces allowed sampling in a preset area without the need of a template and served to mapping the readings of alpha emission from the surface using a pancake probe counter, aiming at correlating the readings with the pancake probe and the wipe samples.

Discs of filter paper with 50 mm diameter were smeared on 100 cm^2 surface area square fields, in each sampling run and then counted by 1 minute in an Eberline SAC-4 counter. Three or four samples were taken in succession from the same field in an attempt to empirically determine the collection efficiency of the wipe. The collection efficiency f_i in each sampling was then calculated by the formula:

$$f_i = \frac{C_i - C_{i+1}}{C_i}$$

where the C_i's are the counting rates of each subsequent wipe samples.

This formula is based on the assumption that in each wipe the collection efficiency is the same and the process of collecting the settled material from the surface remains unchanged. Therefore, care was taken to reproduce as much as possible the same conditions in each sampling run: the area smeared, the applied pressure, and the smearing movement, an S-stroke, edge to edge direction covering the entire sample field.

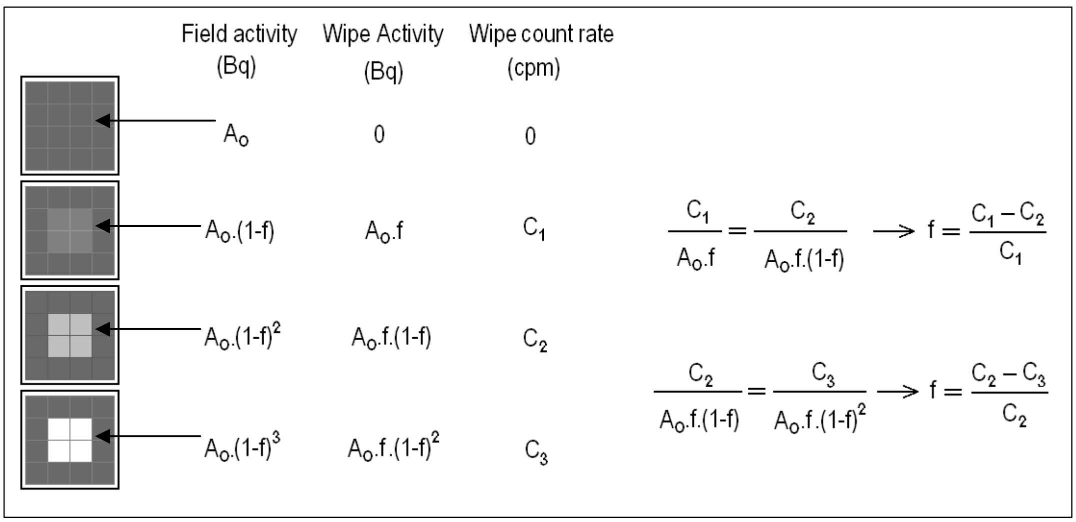

Figure 1. Derivation of the formula to estimate transfer factors.

However, this assumption cannot hold true every time because the nature of the contamination can change between successive runs, for instance as a consequence of changes in particle size distribution of the settled dust, differences in the nature and intensities of the forces of adhesion between remaining settled particles and the surface, etc.

Besides any microscopic phenomena that can disturb the sampling process, it was clear that small differences in the rubbed area in each run induced large errors in the results of the collection factor. A circular paper wipe and a square sampling field made these mistakes a frequent occurrence.

In an attempt to circumvent this difficulty two approaches were tried: a) frames to limit the rubbing movements during wiping; b) tape lift sampling.

The frame used in the sampling is an outer ring to limit the sampling field. The paper wipe is fastened to a holder as to precisely define the sample surface area. The diameter of the ring was calculated as to produce a one hundred square centimeters sampled area. Successive samples are taken from the same field by keeping the frame fixed in position during successive runs. Tape lift sampling uses an adhesive film disc applied onto the surface and peeled off immediately thereafter. The area sampled is equal to the area of the adhesive film. The size of the adhesive film must be equal to the paper wipe because the counting chamber limits the wipe diameter to 50 mm. A number of different adhesive film types and brands are being test in respect to their performance as candidate to further testing, although results from one type only are presented here.

Table 1 shows the results of sampling runs in 20 sampling fields, with both methods.

Table 1 – Transfer factors obtained in wipe sampling runs. [(1)]

	cpm	f	cpm	f	cpm	f	cpm	f
	FIELD 1[(2)]		FIELD 2		FIELD 3		FIELD 4	
Wipe 1	6538	0,80	6284	0,75	4916	0,75	3322	0,86
Wipe 2	1333	0,52	1559	0,67	1243	0,76	476	0,61
Wipe 3	636	0,40	518	0,04	293	0,49	184	0,63
Wipe 4	380	0,28	496	0,22	150	0,29	68	
Wipe 5	272		385	0,52	106			
Wipe 6			184					
	FIELD 5		FIELD 6		FIELD 7		FIELD 8	
Wipe 1	3843	0,88	4181	0,87	1627	0,58	2485	0,68
Wipe 2	480	0,48	534	0,43	691	0,68	804	0,50
Wipe 3	249	0,74	306	0,39	220	0,78	398	0,68
Wipe 4	64		188	0,18	49		129	
	FIELD 9		FIELD 10		FIELD 11		FIELD 12	
Wipe 1	1789	0,90	1636	0,95	2092	0,95	1154	0,95
Wipe 2	176	0,67	77	0,45	98	0,54	60	0,22
Wipe 3	58		42		45		47	
	FIELD 13		FIELD 14		FIELD 15		FIELD 16	
Wipe 1	450	0,81	358	0,52	374	0,67	287	0,67
Wipe 2	87	0,67	173	0,56	124	0,49	95	0,68
Wipe 3	29	-0,21	76	0,46	63	-0,02	30	0,43
Wipe 4	35		41		64		17	
	FIELD 17		FIELD 18		FIELD 19		FIELD 20	
Wipe 1	444	0,70	389	0,66	388	0,69	483	0,79
Wipe 2	132	0,60	132	0,66	120	0,31	101	0,43
Wipe 3	53	0,89	45	0,36	83	0,80	58	0,64
Wipe 4	6		29		17		21	

(1) Sampling methods: swiping with filter paper: fields 1 to 6; tape lift sampling: fields 7 to 20; (2) Sampled surfaces: vinyl flooring tiles: fields 1, 2, 7, 8, and 13 to 20; painted carbon steel: fields 3, 4, 9, and 10; stainless steel: fields 5, 6, 11, and 12. For more details, see text.

Wipe alpha counts are expressed in counts per minute and the corresponding efficiencies are presented as absolute numbers. Sampling method and surfaces sampled in each field are indicated in the footnote of table 1. Efficiency results show a rather larger variation then expected. The reason is mainly systematic errors incurred in defining exactly the sampling field between successive samples. Small deviations from the perimeters defined in previous smears induced large errors in the collected activity. Consequently, these results can give no support to the conclusion that different sampling methods or different surfaces sampled can influence the transfer factors. A more rigorous statistical analysis of results on these aspects will be done after experimental conditions became more reliably controlled.

Negative values of collection efficiencies could lead to the negation of the hypothesis of constant transfer factors. However, before refusing definitively the proposed formula as not valid to determine wipe sampling transfer factor under operational conditions, experimental procedures will be careful reexamined.

The priority in continuing this research is to deal with difficulties in sampling the same area of successive wipes and to establish methods of sampling that can issue reliable results.

3. DISCUSSION AND CONCLUSIONS

The present study is relevant to waste characterization and aims at determining non-fixed surface contamination in materials and objects collected as radioactive waste. However, in a later phase, the methods under development could apply to the control of surface contamination in occupational radioprotection routine. To accomplish this later phase, the cooperation of researchers in the radioprotection field will be required.

The purpose of the waste characterization undertaken in the present study is twofold: first, segregate materials and objects formerly classified as radioactive waste that nevertheless can be released for unrestricted use; second, to inventory radionuclides present in the materials that will undergo management as radioactive waste and to determine their concentrations.

Under the head of the first purpose above, the objectives of waste characterization and of occupational radioprotection are the same: to control dissemination of contamination avoiding or minimizing exposure of individuals – individuals of the public in the case of declassified wastes releases, and workers in the case of occupational radioprotection.

Materials and objects that are classified as radioactive waste because of the presence of non-fixed surface contamination is a relatively common occurrence in waste management. Examples of this class of waste are replaced parts of equipment from radiopharmacy facilities, radioactive lightning rod scrap metal, and replaced or disused parts of equipment from mining and milling operations with naturally occurring radioactive materials.

Transfer factors in routine wipe sampling for waste characterization are seldom known. Operators estimate the contaminant activity assigning a value to the transfer factors, but with little assurance either that the adopted value is close to the true value, or that it is conservative, unless it is sufficiently low. If it is possible to demonstrate that transfer factors can be empirically determined in real conditions, with successive samples of the same area, transfer factor guessing is no longer necessary. It is conceivable that, under certain conditions, successive samples of the same area yield constant transfer factors.

The methods of the present research work are based on the premise that sampling conditions can be controlled, as to allow the demonstration or not of the constant transfer factor hypothesis. If valid, the method can be used in more reliable procedures in assessing surface contamination of scrap metal and other wastes.

Transfer factors can be determined by carefully measuring the activity concentration on the sampled surface before and after wiping and by measuring the activity present in the wipe. In a later phase of this research work, this will be done as a confirmatory test and as a means of estimating uncertainties in the determination of transfer factors. In the present step, the focus is on controlling sampling process as to standardize procedures and to minimize systematic errors, such as to displacing contamination to other parts of the surface, to sampling in different areas of successive wipes, to changing the degree of fixation of contaminants onto the surface, etc.

Results of collection efficiency obtained thus far show much larger variation then previously expected. Source of error is mainly systematic errors caused by the difficulties in covering the same area smeared in each successive sampling trial. Techniques to overcome this problem have been already devised and are being put into practice.

ACKNOWLEDGMENTS

We acknowledge the collaboration of Paulo Senise in performing a number of sampling runs. We also acknowledge with thanks the comments of the anonymous reviewer of this paper.

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