A STUDY OF THE DECONTAMINABILITY OF SURFACE MATERIALS FOR USE IN NUCLEAR INSTALLATIONS

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Introduction

The use of surface materials allowing the easy removal of contaminations, which may occur during routine operation or during accidents is an important part of the efforts directed to the safety of nuclear installations. However, requirements and methods of test influencing the selection of such surface materials have always been subjects of controversial developments and discussions at national and international level. The Federal Institute for Materials Testing (BAM) has, at both levels, contributed to an improvement of the situation by an own research project and by participating in the troublesome work of relevant standardization. This work was furthered by the Federal Ministry of the Interior.

Terms and Definitions

Some of the more important terms to be used in the following need a careful definition, since they are often handled in an ambiguous way.

In practice <u>contamination</u> of a surface is simply defined as the presence of a radioactive material soiling the surface. According to this <u>decontamination</u> is the removal or reduction of the soiling radioactive material. Both definitions do not take into account the existence or strength of an interaction between surface material and radioactive material. As soon as the decontaminability of surface materials is considered, such interactions become the main point of interest and the above definitions are insufficient, if used in the context of a test method. All these methods are based on the following definition: <u>Decontaminability</u> is the ability of a surface material (being in contact with radioactive material)

to resist fixation

<u>and</u> (should a fixation have occured)

to allow easy reduction of the fixed radioactive material as a result of cleaning efforts.

From this definition the two components of decontaminability <u>Contamination Resistance</u> and <u>Ease of Reduction</u> are clearly visible. There have been repeated attempts to measure both components seperately, e.g. in the French standard method, but neither an adequate experimental method nor the necessity for doing so could be demonstrated. Since decontaminability is a specific quality of a surface material, care has to be taken, that the cleaning efforts mentioned above do not comprise single steps which tend to reduce fixed radioactive material in an unspecific manner by use of abrasive or corrosive agents.

The numerical description of decontaminability has mainly been done in terms of the decontamination factor DF, obtained after more or less well defined contamination and decontamination procedures.

Related terms of similar structure, e.g. residual activity fraction or percentage as well as decontamination percentage have also been used. The decontamination factor is defined as

$$DF = \frac{\text{initial activity}}{\text{residual activity}} = \frac{\text{initial pulse rate}}{\text{residual pulse rate}}$$

The initial activity as it is defined in practice and by most of the test methods is the activity brought into contact with the surface during contamination

Problems

A careful consideration of the term "initial activity" immediately leads to difficulties. In most cases only a small activity is finally fixed to the surface by various physical and chemical processes. The bulk of the activity remains in a state free of interactions with the surface and will not contribute to the residual activity. Concentrations being kept constant, the fixed activity is a constant for a specific surface material. An increase of the initial activity will then not result in an increase of the fixed activity or the residual activity. In the calculations, however, an increased initial activity leads to an increased DF-value. The contribution of the fixed activity, reflecting by quantity and behavior the specific qualities of the surface material, to the DF-value is in this way arbitrarily masked and diluted. The test method is loosing its discriminating power and will produce high DF-values for the whole range of surface materials. In this connexion it is unimportant, whether the contaminating material on the surface is a liquid or a salt crust, obtained by evaporation of a liquid. Expressed in terms of crystalline monolayers of the Co- and Cs-compounds used for contamination, three of the test methods offer between 1.6 and 14 monolayers for fixation. The highest value for a fixation measured in the author's laboratory was around 0.3 monolayers, but a material like this would never be considered for use in nuclear installations. Typical fixation values for surface materials of excellent decontaminability are two orders of magnitude lower.

In a test method the size of the initial activity, representing a quantity of the contaminating element (active plus inactive), is normally derived from other experimental parameters. For example, a certain minimum quantity of the contaminant solution is needed to cover a constant and sufficiently large area of the test specimen. Furthermore, care has to be taken, that sufficiently high residual count rates are obtained in case of surface materials of excellent decontaminability. The initial activity, therefore, is not a free variable which may be adjusted in accordance to the aspects discussed above. It would be more appropriate to consider, whether there is a real need for terms like decontamination factor and initial activity in the assessmant of decontaminability.

Another principal problem arises from the need for cleaning a test specimen prior to testing. It has been demonstrated by use of autoradiography, that dust particles and other types of dirt picked up by the test specimens during transport will spoil the test result, if not removed by an effective cleaning process. This has to comprise the use of cleaning mixtures for degreasing as well as wiping and rinsing steps. It has been argued, that such efforts are not only far away from practice, but furthermore tend to change surface properties due to abrasion, chemical attack or pickup of solvents by the surface material. Such aspects are indeed of importance and should be carefully considered. The fact, that in practice surfaces are normally not cleaned with similar care cannot be a reason to accept dirt of unspecific composition and quantity as a component of a specific surface material to be tested. Abrasion can be avoided, otherwise wiping with soft cellulose tissues would decrease the decontaminability, which is not the case. Chemical attack by organic solvent mixtures used for degreasing is a possibility that would only be excluded, if the cleaning agents could be carefully adjusted to the chemical resistance of the material to be tested. This, however, is a difficult task and in a standard the choice of the cleaning agent should not be left to the judgement of the testing center. The use of detergent solutions for degreasing has been recommended, but there is experimental evidence, that detergent residues on the surface will also influence decontaminability results.

The choice of the decontamination agent for the test is a third complex of different approaches. In the French standard method the contaminated area of the test specimen is rinsed with 1.7 litres of water and no reduction of the fixed activity is expected as a result of this treatment. In Germany water is a favourite decontamination agent for a number of reasons and our own experiments indicate, that water is able to remove up to 90 % of the fixed activity, depending on the type of surface material. Other decontamination agents may be used instead of or in addition to water, but their effect is, as a rule, proportional to the effect of water and in some cases smaller than to be expected. This has been demonstrated by the systematic evaluation of the results obtained with the German standard method. This method applies water, an acid detergent solution and 1 m hydrochloric acid in three successive decontamination steps. - A German round robin experiment concerning the effect of three commercially available decontamination agents on four different contaminated surface materials resulted in a mean value of 4 for the factor by which the products were better than water. Extreme values for the factor were 0.6 and 16.

Proposed test method

Starting in 1980 Working Group 10 of ISO/TC85/SC2 first considered the current national standards for the test and assessment of decontaminability in search of a possible ISO standard method. The British and the German standard were then selected for comparison in international round robin experiments. The results for both methods were unsatisfactory due to their low interlab reproducability. The German method proved to be only slightly better than the British one. Therefore, it was decided to develop a compromise method from the best elements of both national methods. This compromise method was tested and improved in three round robin experiments with nine participating institutions from four countries. Finally a sufficient interlab reproducability was achieved and the main sources of error were identified.

In December 1983 a first draft proposal on the basis of the compromise method was completed and circulated among SC2-members to collect comments.

In the following a short listing of the essential features of the proposed method is given together with a description of the advantages to be expected while applying the method:

1. Test specimen dimensions of 50 mm x 50 mm allow transport in slide storage containers without contact of the surface areas to be tested.
2. Separate application of ^{60}Co- and ^{137}Cs-contaminant solutions allows use of different types of detectors with solutions from a common source and increases the value of the information obtained.
3. Composition, production and storage of the decontaminant solutions are carefully described in order to minimize the contributions from this most important source of errors.
4. A four-step wiping and rinsing procedure on the basis of a petrol ether-isopropanol mixture and demineralized water was included to reduce the impact of dirt on the measurement results.
5. Demineralized water is applied as the sole decontamination agent for the sake of simplicity and the ease of waste management. Being the most widely used decontamination agent water is likely to be a good standard material for this purpose.
6. Contamination is done by help of a special holder for test specimen, allowing for the first time the contamination of a constant area by a constant volume of contaminant solution.
7. Five test specimens are decontaminated simultaneously in a cage-stirrer apparatus under identical conditions. Waste volumes and the time spent for a test are considerably reduced; the influence of differences between operators is brought to a minimum.
8. The terms decontamination factor and initial activity are no longer used. The measurement results are expressed in terms of Standardized Mean Residual Pulse rates (SMRP) for the single radionuclides and and of the Final Residual Pulse rate (FRP) for the combination of both radionuclides.
9. The assessment of the decontaminability or ease of decontamination is done on the basis of the FRP-values by means of a classification table which has been empirically compiled.

In the course of the round robin experiments some highly sensitive experimental parameters have been identified: pH-values and carrier concentrations of the contaminant solutions, effectiveness of the cleaning procedure, chemical resistance of a silicone rubber ring being in contact with the test specimen and the contaminant solution and the way of handling the apparatus used for contamination.

Variations in some other parameters were of minor importance: temperatur and volume of the contamination and decontamination agents, the length of time spent for contamination and decontamination, the stirrer speed of the cage-stirrer apparatus, the type of detector and the counting geometry used.

In a discussion of sources of errors it should be noted that inhomogeneities in a group of test specimen often occur to such an extent that experimental errors contribute only slightly to the deviations obtained.