**RADIOLOGICAL CONTAMINATION
PENETRATION DEPTH IN FERNALD
TRANSITE PANELS**

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BY

**William R. Russ
John D. Valentine
Wei Chung**

**University of Cincinnati
Department of Mechanical, Industrial,
and Nuclear Engineering**

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Radiological Contamination Penetration Depth in Fernald Transit Panels

DRAFT

William R. Russ, John D. Valentine, and Wei Chung
University of Cincinnati
Department of Mechanical, Industrial, and Nuclear Engineering
Cincinnati, Ohio 45221-0072

ABSTRACT

To characterize the penetration depth of radiological contamination through the thickness of transit (an asbestos-cement building material) from the Department of Energy (DOE) Fernald site, both destructive and non-destructive analysis techniques were used. The destructive techniques were based on progressively removing layers of material and subsequent direct analysis of successive surfaces. These laminar analyses included quantitative measurements using a Geiger-Mueller (G-M) detector and qualitative measurements based on autoradiography and ultraviolet photography. G-M detector measurements during layer removal provided quantitative distributions consistent with diffusion theory and have served to validate a novel non-destructive technique. The ultraviolet analysis provided qualitative information with the advantage of instantaneous results that may be useful for screening samples. The autoradiographic analysis also provided qualitative results for comparison and image analysis. Both quantitative and qualitative results from this study indicated that the contamination did penetrate into the volume of the transit. However, this penetration depth was observed to be strongly dependent on the manner in which the transit was exposed to the contamination. Consequently, it is likely that significantly different penetration depths will be observed for different processes, buildings, and sites.

INTRODUCTION

A typical decontamination and decommissioning concern for both DOE facilities and nuclear power industry facilities is accurately determining the depth to which radiological contamination has penetrated building materials [1,2]. The optimal means of decontaminating or disposing of such materials will often be dependent on this penetration depth. The building material of primary concern in this study is transit, an asbestos-cement material used for both internal and external walls in process buildings at the DOE Fernald site. Transit was used extensively as building panels at DOE sites prior to the implementation of asbestos-controlling regulations. Uranium processing at the Fernald site has resulted in the radiological contamination of these panels. In the current era of remediation, prior to the disposal of these transit panels, the multiple concerns of toxic (asbestos-containing) and radioactive (process contaminated) waste must be addressed. Such a mixed waste is much more costly to dispose of than waste which is solely toxic or radioactive. It is therefore desirable to minimize the amount of mixed wastes. In the case of Fernald transit, surface decontamination may potentially restore the bulk of the material to the simpler toxic waste form. The viability of decontamination hinges on the distribution of the contamination through the thickness of the transit panels. Consequently, characterizing this contamination distribution is the first step in the disposal of the transit. Note: the term "contamination distribution" will be used to indicate the distribution of contamination through the thickness of transit samples, and does not refer to the distribution of contamination on the surface of the samples unless specifically mentioned.

The potential exists for a variety of destructive and non-destructive techniques to be used to characterize the contamination distribution through the thickness of transit. Destructive methods primarily involve a sequential layer removal and subsequent analysis of the exposed surfaces or the removed material. Use of such destructive techniques is limited due to the difficulties in handling radiologically contaminated asbestos made friable by the destructive process. However, these destructive techniques are necessary in lieu of a suitable established non-destructive technique.

SAMPLES

Transite is the brand name of an asbestos-cement construction material consisting of chrysotile asbestos (magnesium silicate) and portland cement. Manufacturing consisted of multiple layers of material bonded in a hydraulic press to form composite laminar panels. Transite was chosen as a building material for its strength and ability to resist moisture, heat, and corrosion. At the Fernald site, these panels typically have the dimensions of 1.22 m $\times$ 3.05 m and are either flat or corrugated in form. The flat panels have a nominal thickness of 0.65 cm whereas the corrugated panels have a nominal thickness of 0.95 cm. In general, the flat panels were used for internal walls and the corrugated panels were used for external walls.

The samples used in this study to develop a methodology for determining the contamination distribution were taken from two different buildings at the Fernald site with very different process histories, representing extreme cases. One was Building 7A, a dry process area where very little process contamination occurred, and the other was Building 2A, a wet process area where more process contamination occurred. Two groups of flat panels (A, and C) were taken from these buildings for analysis. Both of these groups consisted of six panels with each panel being of approximately 25 cm $\times$ 25 cm. Group A consisted of flat panels from the interior of Building 7A. Group C consisted of flat panels from the interior of Building 2A. Each panel within the group was further labeled 1 through 6 in each group, resulting in panels A1 through A6 and C1 through C6. Furthermore, these panels were subdivided into 9 samples each, a through i. For example, panel C5 was divided into approximately 7.6 cm $\times$ 7.6 cm squares: C5a through C5i. Each of these samples represents the basic unit of analysis and has an area of approximately 60 cm². This sample size allows sufficient area for practical application of analytic methods while attempting to maximize the degree of contamination homogeneity.

EXPERIMENTAL PROCEDURES

The techniques used in the destructive analysis for this study provide information only about the amount of contamination on the surface of the sample. To obtain information about the contamination distribution, surface layers must be removed to allow measurements at various depths. In maximizing the number of successive surfaces analyzed by minimizing the thickness of each removed layer, it is possible to approximate the continuous contamination distribution.

Destructive Layer Removal

A 1.0m $\times$ 0.5m $\times$ 1.2m high efficiency particulate air (HEPA) filtered glove box [3], built specifically for this study, was used to contain radiologically contaminated friable asbestos generated during layer removal. Samples of transite were placed in the containment along with a belt sander and micrometer. Each sample was then placed in a vise, which was attached to a wooden base inside the containment, such that about half of the sample thickness extended above the top of the vise. The belt sander with rough grit sandpaper was then used to remove layers of material. Applying the sander in a variety of directions ensured as uniform a removal as possible. With extreme care, it was possible to remove a sufficiently uniform layer with a minimum thickness of approximately 0.025 cm. Variation in the thickness measured by the micrometer at eight locations on the sample provided a measure of this uniformity.

Quantitative Destructive Analysis

Prior to the initial layer removal and after each of these layer removals, the sample was analyzed with a Geiger-Mueller (G-M) detector, ultraviolet (UV) photography, and autoradiography. The G-M detector was used by placing the probe, which was about the same size as the sample, on the sample face and recording the count rate. Layer removal and analysis continue until the level of contamination is below the threshold of detection (background level).

Qualitative Destructive Analysis

When uranium is exposed to an oxygen-containing environment, such as air, uranyl ions (UO₂²⁺) are formed. These ions fluoresce visible green light when exposed to UV light. Thus, when a sample is contaminated with uranium, exposing that sample to UV light provides a technique for localizing the presence and extent of contamination as the intensity at which the sample fluoresces is proportional to the uranium concentration. Consequently, the presence of uranium contamination can be determined qualitatively by direct observation. Furthermore, through the use of standards to determine relative

intensities, image analysis could likely produce quantitative results. However, such a quantitative analysis would require extremely consistent image processing. Because UV light interacts with just the surface of the sample, this method provides information about contamination on the surface, and is very quick, simple, and inexpensive [4,5]. For the UV photography in this study, the transite samples were placed in a light box equipped with ultraviolet fluorescent bulbs which emitted light at a 254 nm wavelength. A 35 mm camera was attached to the viewport with 200 speed film and an exposure duration of 4 seconds. For comparative purposes, consistent settings and development were crucial. Different levels of contamination resulted in varied intensities and hues of green. Optically scanning the image into a digital format allowed some image enhancement to emphasize the actual contamination. However, an accurate interpretation of the UV photography images often required comparison to the equivalent autoradiograph.

Autoradiography differs from typical radiography only in that the source of radiation is provided by the sample itself and a separate radioactive source is not required. Classical radiography involves capturing the image of the sample on a photographic plate based on the attenuation of radiation from an external source by the intervening sample, while autoradiography captures the actual pattern and intensity of radiation from a sample. Consequently, the only requirements for autoradiography are radiosensitive film and a means of handling the light sensitive media for exposure purposes. Since autoradiography also provided a spatial representation of contamination across the surface of the sample, it was used for comparison to the UV photography results for verifying which regions contain uranium contamination. Autoradiography provides surface or near surface information. Therefore, autoradiography requires destructive analysis to provide information about the contamination distribution through the thickness of the transite panels. The autoradiographs were produced using standard X-ray imaging film with a central polymeric base coated on both sides with a thin emulsion covered with an anti-scratch layer. The film was placed in direct contact with a sample and stored in a light-tight box for an appropriate exposure time. Consistent developing was provided by a standard automated developer.

Non-Destructive Analysis

The contamination distribution results from the destructive analysis were used to both characterize the transite and to provide verification of a novel non-destructive technique using gamma-ray spectrometry proposed by Chung, et al.[6]. This non-destructive method was based on measuring the gamma-ray spectra from both sides of the sample. Fig. 1 illustrates the implementation of this technique. Note the different photopeak areas recorded by the two detectors in the two gamma-ray spectra. In conjunction with knowledge of the gamma-ray linear attenuation coefficient for the material and proper diffusion or leaching models representative of how the material was contaminated, the ratio of photopeak areas at several energies from these spectra can be used to infer the most-probable contamination distribution. Use of a high-purity germanium (HPGe) detector provides sufficient energy resolution to discern all of the photopeaks of interest. Comparing the measured ratio of the respective photopeak areas from both sides of the panel (Fig. 1-b) over a range of energies to computer-generated ratios for possible contamination distributions provides a prediction of the most-probable distribution (Fig. 1-c). The computer generated ratios were based on distributions predicted by diffusion theory for given types of exposures, or initial conditions. In particular, for a single instantaneous exposure, the distribution was based on a gaussian function, and for a constant exposure, the distribution was based on a complementary error function [7]. For this study, the condition of constant exposure existed and the complementary error function proved to be applicable.

RESULTS

Destructive analysis was conducted on both sides of one A sample and eleven C samples using the G-M detector to obtain quantitative contamination distributions. Approximately six layer removals were required per side to reach background level. As representative G-M detector data, sample C5a produced the results shown in Table 1 and Figure 2. Figure 2 also illustrates the correlation with the contamination distribution predicted by the non-destructive technique. To determine how the contamination distributions vary from sample to sample, the depth at which count rates from the G-M detector approached background were recorded. Table 2 lists the sample sides analyzed and their corresponding depth to background level.

To further verify the penetration of uranium concentration into the thickness of the transite panels, both UV photographic and autoradiographic analyses were performed on several samples. The autoradiography required an exposure time of either 24 or 48 hours, depending on the contamination level. It was

empirically determined that a G-M detector reading of about 1000 counts per minute (cpm) or greater could be exposed for 24 hours to produce a sufficient image. Lesser activity required a 48 hour exposure.

As representative of UV photographic and autoradiographic results, sample C5a produced the images shown in Figs. 3 and 4, where Fig. 3 shows the sample prior to layer removal and after a single layer removal and Fig. 4 shows the sample after two and three layer removals. The progression of decreasing contamination and image correlation for the same layer are illustrated by the dark green in the UV photographs which correspond to the dark areas in the autoradiographs.

Fig. 1: Illustration of the non-destructive gamma-ray spectrometry technique used to determine the contamination distribution in the sample with two detectors: a) the sample-detector geometry, b) the gamma-ray spectra recorded by the detectors with one of the full-energy peaks, and c) the relationship of measured response ratios as functions of gamma-ray energy compared to computer-generated ratios.

Table 1: Example of layer removal results based on G-M detector β - γ measurements
(original sample thickness: 0.653 cm)

Sample C5a - side 1		Sample C5a - side 2	
Total Thickness Removed (cm)	Activity (cpm)	Total Thickness Removed (cm)	Activity (cpm)
0	19000	0	3500
0.056	1200	0.020	700
0.081	500	0.074	300
0.117	100	0.124	100
0.145	80	0.147	50 (background)
0.188	50 (background)		

Fig. 2: Contamination distribution in one transite sample (C5a) as measured by the destructive analysis with a G-M detector and predicted by the non-destructive technique.
(contamination levels normalized)

Table 2: Background depth for transite samples analyzed

Sample Side	Background Depth	Sample Side	Background Depth	Sample Side	Background Depth
A3c side 1	0.013	C5b side 1	0.109	C5f side 1	0.150
A3c side 2	0.033	C5b side 2	0.140	C5f side 2	0.163
C5a side 1	0.188	C5c side 1	0.112	C5g side 1	0.107
C5a side 2	0.147	C5c side 2	0.155	C5g side 2	0.150
C4b side 1	0.135	C5d side 1	0.147	C5h side 1	0.122
C4b side 2	0.140	C5d side 2	0.152	C5h side 2	0.244
C4e side 1	0.152	C5e side 1	0.147	C5i side 1	0.104
C4e side 2	0.137	C5e side 2	0.155	C5i side 2	0.160

One concern with results based on layer removal using a belt sander was the possibility of cross-contamination between subsequent layers due to embedding by abrasion. The well-resolved contamination pattern images from the UV photography and autoradiography and the ability to achieve background level after several layers have been removed indicate that this effect is negligible. In addition, the shape of the measured contamination distribution is consistent with the diffusion theory predictions and the results of the non-destructive technique.

CONCLUSIONS

The contamination distribution results indicate the degree to which a given sample can be labeled as surface or volumetrically contaminated. The quantitative data provided by the G-M detector produced distributions consistent with the non-destructive technique. Therefore these G-M detector distributions adhere closely to what was predicted by diffusion theory given the appropriate initial conditions.

The UV photography technique proved to be a good qualitative tool for quickly analyzing the samples. Using a series of standard sources along with extremely consistent imaging, it would likely be possible to obtain quantitative data through digital image processing. For this study, the autoradiographs were used as a reference to provide a gross, qualitative indication of what aspects of the UV image corresponded to contamination. The autoradiographs were not limited by extraneous background imaging (visual noise), as the UV images were, since exposure of the film resulted only from radiation produced by the contamination. The threshold of detection was slightly better for the autoradiographs than for the UV photographs. However, autoradiography required lengthy exposure time, on the order of days. Autoradiography could also possibly be quantitative through image analysis.

Given the primary concern of determining whether the transite can be decontaminated such that the bulk of the material is at or below background levels, a key characteristic of interest is the depth to background or the depth of material that potentially requires removal. Should this characteristic be too large, decontamination may not be viable for a given method of removal. The majority of the samples had a background depth of approximately 0.140 ± 0.012 cm on both sides, all of which had a high level contamination process history (Building 2A- extensive use/ wet process). With an average total initial thickness of 0.658 ± 0.026 cm and contamination on both sides of all samples, this represents about 40% of the total material. For samples with a process history that resulted in significantly less contamination (Building 7A- limited use/ dry process), the background depth was approximately 0.025 ± 0.107 cm which would represent about 8% of the total material. The viability of decontamination is illustrated to be heavily dependent on the process history. For future studies, it is recommended that a greater variety of sample contamination exposure histories be used for a stronger correlation to background depth.

This study established a reliable means of determining the penetration of contamination in flat transite panels by destructive layer removal and analysis. In addition, the novel non-destructive technique using gamma-ray spectrometry has been validated. Specific sample contamination distributions have been shown to be dependent on exposure history, as consistent with diffusion theory, and should be applicable to materials other than transite.

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