RECEIVED
JAN 17 1995
OSTI

Treatability Study Using Prompt Gamma Neutron Activation Analysis (PGNAA) Technology Phase I

Topical Report

T.V. Congedo
A.R. Dulloo
F.H. Ruddy
D.H. Weigle
R.P. Williams
J.G. Seidel

June 1995

Work Performed Under Contract No.: DE-AC21-93MC30177

U.S. Department of Energy
Office of Environmental Management
Office of Technology Development
Washington, DC

For

U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

By
Westinghouse Science and Technology Center
Pittsburgh, Pennsylvania

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, 175 Oak Ridge Turnpike, Oak Ridge, TN 37831; prices available at (615) 576-8401.

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22161; phone orders accepted at (703) 487-4650.

Treatability Study Using Prompt Gamma Neutron Activation Analysis (PGNAA) Technology Phase I

Topical Report

T.V. Congedo
A.R. Dulloo
F.H. Ruddy
D.H. Weigle
R.P. Williams
J.G. Seidel

Work Performed Under Contract No.: DE-AC21-93MC30177

U.S. Department of Energy
Office of Environmental Management
Office of Technology Development
1000 Independence Avenue
Washington, DC 20585

For

U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
P.O. Box 880
Morgantown, West Virginia 26507-
0880

By

Westinghouse Science and Technology Center
1310 Beulah Road
Pittsburgh, Pennsylvania 15235-5098

June 1995

CONTENTS

LIST OF FIGURES	iv
LIST OF TABLES	iv
DISCLAIMER	vi
1. EXECUTIVE SUMMARY	1-1
2. INTRODUCTION	2-1
3. PGNAA AND THE N-SCAN™ APPROACH	3-1
4. PROGRAM TASKS	4-1
5. PGNAA MODELING	5-1
6. EXPERIMENTAL RESULTS AND DISCUSSION	6-1
6.1 DESCRIPTION OF THE LABORATORY SYSTEM	6-1
6.2 EXPERIMENTAL RESULTS	6-3
6.2.1 Original N-SCAN System	6-3
6.2.2 Modified N-SCAN System	6-5
6.3 PLANNED SYSTEM IMPROVEMENTS	6-15
6.4 DISCUSSION	6-16
7. LIMITATIONS OF PGNAA AND COMPARISON WITH SOME COMPETING METHODS	7-1
8. SUMMARY	8-1
9. REFERENCES	9-1

LIST OF FIGURES

Figure 3.1 — Schematic Illustration of Pulse Timing in N-SCAN PGNAA.....	3-2
Figure 4.1 — Laboratory Test Facility.....	4-2
Figure 4.2 — Modular Concrete-on-Grade Floor Mock-Up and N-SCAN Nuclear Components.....	4-3
Figure 4.3 — Illustration of the Stack of Disks Used to Simulate the Presence of Contamination at Varying Depths in the Concrete.....	4-4
Figure 6.1 — Distribution of the Thermal Neutron Flux as a Function of Radial Distance from the Neutron Generator (Measured with ^{235}U SSTR Neutron Dosimeters).....	6-2
Figure 6.2 — Interrogation Volume as Viewed by the HPGe Detector in the Modified N-SCAN Configuration.....	6-7
Figure 6.3 — Mercury Limit of Detection of the Modified N-SCAN System	6-8
Figure 6.4 — Cadmium Limit of Detection of the Modified N-SCAN System	6-9
Figure 6.5 — Uranium-238 Limit of Detection of the Modified N-SCAN System ..	6-10
Figure 6.6 — Thorium-232 Limit of Detection of the Modified N-SCAN System...	6-11
Figure 6.7 — Chromium Limit of Detection of the Modified N-SCAN System.....	6-12

LIST OF TABLES

Table 5.1. Predicted and Measured Prompt Gamma Yield from Mercury- Contaminated Cement Sample, Normalized to the Intensity Measured with the Sample at the Concrete Surface	5-2
Table 6.1. Weight of Contaminant in High-Concentration Cement Disks.....	6-3
Table 6.2. Main γ Energies and Interrogation Depths of Contaminants Listed in Table 6.1	6-4
Table 6.3. Comparison of Modified and Original N-SCAN System Performance Using Response from High-Concentration Mercury Disk.....	6-7
Table 6.4. Limits of Detection of Modified N-SCAN System Based on a 50-Minute Run	6-14
Table 6.5. Comparison Between the Projected Limits of Detection of N-SCAN in Phases I and II	6-16

1. EXECUTIVE SUMMARY

This report reviews the progress accomplished during Phase I of a two-phase project intended to demonstrate the use of Prompt Gamma Neutron Activation Analysis (PGNAA) as a technology for the characterization of hazardous and radioactive contaminants in concrete floors. A comprehensive experimental program was undertaken using the N-SCAN™ PGNAA system, which was initially developed by Westinghouse for soil characterization, to determine the sensitivity of PGNAA for several contaminants in concrete. The experiments were performed in a test facility specially designed and constructed for this project.

The lower limits of detection derived from the experimental data were encouraging for mercury, cadmium, uranium-238, thorium-232, technetium-99, chlorine, uranium-235 and chromium. These limits were achieved after modifications made to the original N-SCAN system significantly improved its sensitivity for elements located at or near the surface of concrete. With the implementation of additional performance-enhancing modifications scheduled in Phase II, the detection sensitivity of N-SCAN at the end of this project is expected to be at least one order of magnitude higher, allowing N-SCAN to become an effective characterization tool.

N-SCAN has several important advantages over current characterization methods and technologies. Its use does not generate any secondary waste because N-SCAN is a non-invasive technique that leaves negligible residual radioactivity. It is capable of providing information on the distribution of a contaminant down to a depth of several inches in concrete. The components of N-SCAN can be readily mounted on a mobile platform, thus facilitating the deployment and operation of the system in the field. Finally, in addition to the characterization of concrete floors and soil, N-SCAN can be utilized for other characterization applications such as drum assaying and surveying of construction debris from D&D activities.

2. INTRODUCTION

To assist the Department of Energy (DOE) in its decontamination and decommissioning efforts, the Westinghouse Science & Technology Center (WSTC) has completed the first phase of a two-phase project to develop a fully-integrated Prompt Gamma Neutron Activation Analysis (PGNAA) system for the characterization of contaminants in concrete floors and walls. The principal objective of this first phase (Phase I) was to demonstrate the non-invasive capability of the N-SCAN™ PGNAA system, initially developed by WSTC for soil characterization, to determine in situ the level of radioactive and hazardous contaminants in concrete. The ability of the PGNAA method of analysis to provide real-time results as well as its capability for automation were to be assessed in Phase I.

Over a period of three decades, Westinghouse has developed PGNAA-based technologies for application in the areas of explosive and contraband detection, trace element monitoring for process control, detection and identification of Chemical Weapons (CW) agents, and in situ detection of trace contamination in soils. In support of the latter application, the N-SCAN™ approach to PGNAA was developed, utilizing a customized pattern of neutron source pulsing correlated with gamma spectrum acquisition on a microsecond time scale to produce signal-to-background ratios of a quality unprecedented in PGNAA systems. Such performance is required in order to distinguish the presence of trace quantities of one or more contaminant elements within a massive, multi-element matrix. The present program consists of applying the N-SCAN process to the in situ detection of contaminant elements in concrete - floors or walls - in order to assist the decontamination and decommissioning of facilities within the DOE complex.

As part of the Phase I program, the sensitivity of the original N-SCAN system for a suite of contaminants was determined. Parametric adjustments were then made to the N-SCAN system to optimize its sensitivity for surface characterization of concrete (the N-SCAN system was originally intended to detect contaminants in large volumes of soil while the needs of this program is for surface/near-surface characterization of concrete). Based on measurements made with the modified system, lower limits of detection (LLD) for the selected contaminants were calculated. These LLD are presented in this report, as well as the LLD that are believed to be ultimately achievable with further modifications to the N-SCAN system. In parallel with the experimental work, modeling tasks were carried out to assist in data interpretation and depth inferencing.

The original suite of contaminants that were to be studied consisted of Hg, Cd, ^{235}U , ^{232}Th , ^{99}Tc , Pb, Ba and Cr. It was found during the course of the project that N-SCAN had good detection sensitivities for ^{235}U and Cl, which are both of interest to some sites within the DOE. As a result, LLD for these two contaminants are also reported.

Finally, the scope of work of Phase I included a demonstration of N-SCAN at the FEMP. Based on a mutual agreement between WSTC and DOE-METC, this task was postponed to Phase II of the program. A discussion of the reasons for this postponement is provided in the report.

3. PGNA AND THE N-SCAN™ APPROACH

Nuclear reactions, including neutron-induced reactions, proceed on a time scale in the range of 10^{-28} to 10^{-14} second, which is essentially instantaneous for electronic timing purposes. However, moderation of energetic ($\sim$ MeV) neutrons to thermal energies requires times of the order of microseconds. Once produced, approximately one half of the thermal neutrons will undergo nuclear reactions approximately every 200 to 300 microseconds. Therefore, the time scale of radiative capture - (n, γ) reactions - is of the order of hundreds of microseconds.

Neutron-induced reactions such as (n,n'), (n,2n), and most (n,p) reactions are produced only with fast neutrons. Since no thermalization is required, these reactions occur essentially instantaneously, or at the same time that a source of fast neutrons is present. Therefore, if a pulsed electronic source is used and data are collected on a time structure on the hundreds of microseconds scale, a time separation of thermal and fast neutron-induced gamma rays can be achieved.

Previous PGNA work had been limited by lack of sensitivity due to a combination of effects. For work employing pulsed neutron generators, the short pulse durations (1-5 microseconds) were characterized by extremely high instantaneous neutron emission rates, and the resulting gamma flux was so large that detector electronics were paralyzed. In addition, all systems - whether using electronic or isotopic neutron sources - were hampered by relatively poor signal-to-background ratios for peaks from trace constituents in a sample. In the Westinghouse system, the detection of gamma rays of interest is facilitated by exploiting the time sequence of neutron-induced reactions discussed above. This is illustrated in Figure 3.1, from which one can observe that gamma rays produced by fast neutron-induced reactions are collected separately from most of the gammas produced by thermal neutron-induced processes. One can also observe that, since the gamma rays from thermal neutron-induced nuclear reactions are collected over only a finite duty cycle (generally 10 to 20%), these gamma spectra bear only a corresponding fraction of background from ubiquitous sources, such as long-lived isotopes in the natural environment or short-lived isotopes produced by the neutron irradiation. Hence, the N-SCAN timing approach achieves high signal-to-background ratios. In addition it must be noted that the N-SCAN method avoids the intense gamma ray background produced by many isotopic neutron sources, such as ^{252}Cf , since electronic neutron generators produce virtually no associated gamma field.

N-SCAN PGNA

Figure 3.1 — Schematic Illustration of Pulse Timing in N-SCAN PGNA

A second consideration in high-sensitivity PGNA is the finite count rate throughput of the gamma ray detector and associated electronics. At present, the maximum achievable gamma ray pulse throughput is about 100,000 counts per second with a system dead time due to pulse pileup of 60-90%, depending on spectrum characteristics. In practice, the high gamma ray energies encountered in PGNA limit the count rate throughput to 20,000-30,000 counts per second. In applications where fast neutron-induced gamma rays are of primary interest, the system can be operated at lower source intensity to optimize the system throughput for data from gamma rays produced during the neutron pulses. The pulsing scheme can also be adjusted so that only a minimum of neutron thermalization occurs before collection of gamma data.

In addition to the features mentioned above, the N-SCAN system utilizes the following equipment to produce high signal-to-background acquisition of high count rate gamma spectral data:

- Large-volume, neutron-resistant, high resolution gamma ray detector
- High-throughput pulse-processing electronics:
 - high speed transistor reset preamplifier
 - state-of-the-art spectroscopy amplifier
 - 450 MHz Wilkinson analog-to-digital converter
- Large-volume, high count rate, anti-Compton suppression shield
- Unique, customized pulsed neutron generator
- Custom-designed (Westinghouse proprietary) Acquisition Interface Module, governing the correlation of neutron pulsing and gamma ray data acquisition
- Computer control system utilizing Ethernet technology for:
 - system control with nanosecond timing capabilities
 - time-sequenced multi-channel analysis for gamma ray spectroscopy
 - VAX/VMS spectroscopy software, including peak search, area integration, area uncertainty evaluation, and nuclear data library match capabilities.

It should also be noted that the electronic hardware components of the N-SCAN system are readily available commercially, and are integrable to a variety of mobile platforms.

4. PROGRAM TASKS

The Statement of Work of Phase I of this program included a series of tasks to be performed. A summary of the status of each task is presented here.

Task 1.1.0. System Equipment Assembly & Checkouts

This task has been completed. A laboratory prototype of the Westinghouse Scientific Ecology Group (WSEG) Field Survey Vehicle was assembled. The Field Survey Vehicle consists of an N-SCAN sensor mounted on a Melroe 753 Bobcat skid-steer loader. The vehicle communicates via RF telemetry with its control system located in a remote Data Acquisition and Control Van. The laboratory prototype replicates the arrangement found on the Field Survey Vehicle of the neutron generator, neutron reflector, gamma ray detector and collimator.

Task 1.2.0. System Calibration for Contaminants in Concrete

Subtask 1.2.1. Assembly of Lab Mockup

This task has been completed. Figure 4.1 illustrates the test configuration, which comprises 25 concrete blocks, each of dimensions 60 cm x 60 cm x 5 cm (2 ft x 2 ft x 2 inch), laid out in a 5 x 5 square array on top of 10 cm (4 inch) of concrete floor on grade. The experimental area is an underground vault located 1.8 m (6 ft.) below ground level. The steps on the right in the photo descend down to the floor level of the experimental setup. Figure 4.2 illustrates the nuclear configuration, including on the right the neutron generator tube and a graphite neutron reflector, and on the left (proceeding from bottom to top) the (bismuth) gamma ray collimator, gamma detector and liquid nitrogen dewar. This configuration was used in the early calibration tests, and is identical to the sensor configuration on the Field Survey Vehicle. An alternate configuration was utilized in the later measurements of the Phase I program, as will be discussed later. Figure 4.3 illustrates the region in the 22.86-cm (9-inch) diameter cutout made in the block beneath the detector. As the figure indicates, blocks of cement whose density is comparable to that of the concrete are used to fill this space. Contaminated disks of 0.635 cm (0.25 inch) thickness, enclosed in a thin layer of aluminum, can be placed at varying heights in the stack, to permit measurement of the PGNAAP response of the contaminant as a function of depth in the concrete.

Figure 4.1 — Laboratory Test Facility

Figure 4.2 — Modular Concrete-on-Grade Floor Mock-Up and N-SCAN Nuclear Components

Figure 4.3 — Illustration of the Stack of Disks Used to Simulate the Presence of Contamination at Varying Depths in the Concrete

Subtask 1.2.2. Nuclear Data Compilation

This task has been completed. It was found from experimental results that gamma rays emitted by the radiative capture of thermal neutrons, as opposed to inelastic scattering of prompt neutrons, provided the most intense peaks in the N-SCAN gamma spectra, and correspondingly gave the lowest limits of detection for the majority of the contaminants. The N-SCAN timing and geometrical configurations were therefore modified to optimize the thermal neutron flux at the surface of the contaminated disks. A discussion of these modifications is given in Section 6 of this report. Compilations of thermal neutron cross section data and radiative gamma energies are readily available.¹

Subtask 1.2.3. Concrete PGNAA Spectrum

This task has been completed. A PGNAA analysis of the principal elements in the experimental vault that contribute to the gamma spectrum was performed in the absence of a contaminated sample. These elements were silicon (concrete), hydrogen (moderator and concrete), iron (structural support of detector assembly), germanium (HPGe detector) and sodium (NaI annulus).

Subtask 1.2.4. Inclusion of Concrete in the Analytical Model

This task has been completed. Originally, the Westinghouse PGNAA analytical model utilized discrete ordinate calculations performed with the DOTIII-W² code. When this program subtask was approached, it was found that the DOT code was no longer available. The successor code, DORT³, was obtained from the Radiation Shielding Information Center at Oak Ridge, and used to implement the model. This model was compared to measurements in the original sensor configuration, and the results are discussed in Section 5.

Subtask 1.2.5. Selection of Sample Set

This task has been completed. DOE settled on the following suite of target contaminants in August, 1994: mercury, enriched uranium-238, natural thorium-232, cadmium, chromium, lead, and barium.

Subtask 1.2.6. Sample Encapsulation

This task has been completed. Each of the contaminants listed in the preceding subtask was encapsulated in an aluminum disk as a dispersion in cement.

Subtask 1.2.7. Sample Response Measurement and System Calibration

The modified analytical model (Subtask 1.2.4) was benchmarked against the experimental depth response profile of the mercury sample using the original N-

SCAN configuration. The results of this benchmarking are described in Section 5. Comparison of the experimental depth response profiles of the other contaminants with the model predictions was postponed when it became clear that, in accordance to indicated DOE needs, the original N-SCAN configuration had to be modified in order to optimize the characterization of a shallow, near-surface region. The investigation of the depth response profiles of targeted contaminants using an analytical model based on the fully-optimized N-SCAN configuration will be undertaken in Phase II.

Task 1.3.0 Site Selection and Approval

Based upon the recommendation of DOE METC, the Fernald site was chosen as the location to perform the prequalification demonstration of Phase I. As part of the approval process, a briefing of Environmental Regulators was conducted. Within the U.S. Environmental Protection Administration Office of Federal Facilities Enforcement (OFFE), Westinghouse located the designated liaison with the EPA Emerging Technology Initiative. On April 26, 1994, a meeting was held at the EPA OFFE, in Washington. DOE-EM542, DOE-METC, and the Westinghouse project team were represented, along with five EPA participants. The purpose of the meeting was to acquaint the environmental regulators with this innovative technology at an early stage in the demonstration process. A key recommendation offered by EPA was that the appropriate interface for demonstrations is the EPA Remedial Project Manager at the DOE demonstration site.

Task 1.4.0 Phase I Prequalification Demonstration

Based on a mutual decision by WSTC and DOE-METC, this task was postponed to Phase II of the program. The two principal reasons for the postponement are 1) the lack of sensitivity of N-SCAN in its original configuration for some of the contaminants at Fernald (notably barium and lead), and 2) a reduction in the 1995 funding level available to the Fernald program for sponsoring the demonstration of characterization technologies at the site. As a result, it was jointly decided by DOE-METC and WSTC to postpone an N-SCAN demonstration to Phase II, when the final configuration of the N-SCAN system for near-surface characterization will be in place. This postponement will also allow DOE-METC to identify another source of funding for a demonstration.

5. PGNAA MODELING

In applying PGNAA to the characterization of a contaminant in soil fields or concrete floors, it is necessary to irradiate an extended medium with energetic neutrons, and then to interpret the yields of signature gamma rays from the contaminant. If the gamma rays are sufficiently energetic, trace concentrations located at significant depths (i.e. many inches, depending on gamma ray energy) into the medium can contribute appreciably to the observed yield. In addition, the natural profile of the thermal neutron flux in the medium, resulting from the medium's moderating characteristics (average atomic weight, elemental identities, density), provides an inherent spatial bias to the prompt gamma neutron activation process.

In cases where the thermal neutron flux has been configured to have significant intensity only in a near-surface region, or where the energy of the signature gamma ray is small (i.e. less than 1 MeV), the interpretation of the gamma data is relatively simple, and an experimental calibration is sufficient to determine both the contaminant concentration and its location. Since the indicated needs of the DOE are focused on the characterization of concrete in a near-surface region (1/8" to 1/4" in depth), the original N-SCAN configuration, which was intended to detect contaminants in large volumes of soil, was modified to maximize the intensity of the thermal neutron flux in the near-surface region so as to improve the system sensitivity for this application (see Section 6). Hence, as mentioned above, the calibration of this modified system can be performed experimentally in a straightforward manner.

In cases where the contaminant has penetrated to a considerable depth below the surface, the interpretation of the gamma data to determine the concentration and location of the contaminant requires radiation transport modeling. Such "deep" contamination can occur either if cracks in the concrete allow surface contamination to spread to deeper regions, or if a contaminant can diffuse rapidly through concrete (e.g. ⁹⁹Tc). Ideally, a depth profile of the distribution of the contamination in concrete would be inferred from a single measurement. This process was developed by Westinghouse⁴ for soil contamination prior to undertaking the present program, incorporating a neutron transport model to predict the spatial distribution of the reaction rate of interest, and a gamma transport post-processor to predict gamma detection rates for diverse spatial distributions of contaminants. In the present work, it was originally planned to implement this process for a concrete matrix. For this reason, the radiation transport model was modified to accommodate a concrete-on-grade floor, assuming the sensor configuration of the Field Survey Vehicle, as depicted in Figure 4.2. During the program, it was learned that the DOTIII-W²

code, with which the earlier soil model had been developed, was no longer available. It was therefore necessary to obtain the DORT^a code from the Radiation Shielding Information Center at Oak Ridge, implement the code on a local RS6000 workstation, transport the original model to the DORT format, and then address the concrete and specific sensor configuration within the DORT model. These tasks were successfully completed.

The gamma ray detection rates measured for three different gamma rays from thermal neutron radiative capture in mercury are considered for the comparison between the model and the experimental data. These gamma ray energies are 368.1 keV, 1693.9 keV, and 5966.2 keV. The sample was a thin (0.635 cm, 0.25 inch) disk of mercury-contaminated cement, covered on top and bottom with a thin (0.079375 cm, 0.03125 inch) layer of aluminum. (The aluminum thickness was greater in the radial direction, but did not represent a significant perturbation of neutron behavior because the moderating properties of aluminum are similar to those of concrete). Table 5.1 illustrates the predicted and measured ratio of gamma yield for each of the three gamma rays with the sample at the top and bottom of the two-inch stack of cement.

Table 5.1. Predicted and Measured Prompt Gamma Yield from Mercury-Contaminated Cement Sample, Normalized to the Intensity Measured with the Sample at the Concrete Surface

Sample center depth cm (inch)	Normalized Gamma Ray Yield					
	368.1 keV		1693.9 keV		5966.2 keV	
	Exp.	Pred	Exp.	Pred	Exp	Pred
0.397 (0.156)	1.0	1.0	1.0	1.0	1.0	1.0
4.683 (1.844)	0.45	0.69	0.62	1.02	1.11	1.21
Calc. / Exp.	1.54		1.64		1.09	

As can be seen from the table, the experimental results and model predictions for the dependence of response on sample depth agree within a factor of 2 for each gamma ray. Modeling with higher order directional quadrature and finer depth mesh near this interface would provide better agreement, as a result of better prediction of the observed yield from the disk in the surface position. Note that in the generation of depth profile algorithms, the theory curve of response vs. depth is anchored to experiment at a point beneath the surface because the air/concrete discontinuity prevents the calculation from converging until several centimeters beneath the surface. Furthermore, the presence of normalized yields greater than one for the higher energy gamma rays reflects the fact that, with the sample placed at the greater depth, the system collimator is not cutting off the detector view of the outer radial region of the disk. When the disk is in the surface position, this effect reduces the observed yield by approximately 60%.

Although DOE needs are principally focused on the characterization of a near-surface region, it is important to realize that N-SCAN, unlike most real-time characterization methods, has the capability to determine contaminant levels in deeper regions below the surface, and that the transport model to support analysis of these deeper interrogation regions has already been developed.

6. EXPERIMENTAL RESULTS AND DISCUSSION

A comprehensive experimental program was carried out to evaluate and improve the detection sensitivity of the N-SCAN PGNAA system for selected contaminants in concrete. Eight sealed samples of contaminated cement were utilized in a total of approximately 150 measurements, which included both active PGNAA interrogation and passive measurement of gammas from natural radioactive decay. This program had the following objectives:

1. Determine the sensitivity of the original N-SCAN system for the selected contaminants in concrete (the original N-SCAN system had been designed for detection of contaminants at greater depths in large volumes of soil).
2. Adjust the system sensitivity in response to DOE's stated detection goals by modifying key parameters of N-SCAN (DOE goals emphasize detection of near-surface contaminants in concrete).

6.1 DESCRIPTION OF THE LABORATORY SYSTEM

In order to perform laboratory experiments designed to emulate field, in situ PGNAA measurements, a laboratory concrete test bed was assembled (see Figures 4.1, 4.2 and 4.3). The components of the concrete test bed were identified in Section 4. The concrete test bed was designed to have modular components which would allow the placement of known volumes of concrete with known concentrations of key contaminants at known depths and locations relative to the gamma ray detector and neutron generator. Using this facility, response calibration data were obtained for the PGNAA system in order to provide input to benchmark calculations. The data that are obtained can be used to provide pointwise response information or can be summed to approximate distributions of interest.

Neutron transport in the concrete test bed was measured using Solid State Track Recorder (SSTR) neutron dosimeters which were placed within the modular concrete structure and exposed to neutrons from the neutron generator. Examples of thermal neutron distributions obtained with ^{235}U SSTRs are shown in Figure 6.1. A reaction rate of 1.0×10^{-26} fissions per atom per source neutron corresponds to a thermal neutron fluence of 1.2×10^5 neutrons per cm^2 per source neutron. These experimental neutron transport data are also used as input to the calculations.

The concrete test bed is a mockup of a 15-cm (6-inch) thick concrete floor. The upper layer is made up of 5-cm (2-inch) thick concrete slabs. The slab placed directly beneath the gamma ray detector has a 22.9-cm (9-inch) diameter circular section removed from its center. Cement disks, uniformly contaminated with known

Figure 6.1 — Distribution of the Thermal Neutron Flux as a Function of Radial Distance from the Neutron Generator (Measured with ^{235}U SSTR Neutron Dosimeters)

amounts of selected contaminants, are inserted into this circular section as illustrated in Figure 4.3. The contaminant disks are 20.32-cm (8-inch) diameter, 0.635-cm (0.25-inch) high cylinders which are encapsulated by a thin layer of aluminum to prevent contamination of the test bed and surroundings. The center of the contaminant disk is located below the axis of the cylindrical HPGe detector. The contaminant disks can be placed at various depths in the concrete test bed to determine system response as a function of depth and to provide data which can be used to simulate depth distributions by summing combinations of the individual depth responses.

6.2 EXPERIMENTAL RESULTS

6.2.1 Original N-SCAN System

The original N-SCAN system was configured for detecting contaminant elements in large volumes of soil. The first series of experiments was designed to obtain sensitivities of the original configuration for the contaminants listed in Table 6.1. In these measurements, high concentrations of the contaminant elements were used in order to clearly identify the useful gamma rays produced and to obtain count rate data with low statistical uncertainties. Measurements were done with the contaminant at several depths to provide sensitivity data as a function of depth and also to provide a data set which could be summed to simulate various depth versus concentration profiles.

The actual disk contaminant concentrations are listed in Table 6.1 as well as corresponding volume-averaged concentrations. The volume-averaged concentration is obtained by assuming that the weight of contaminant in the disk has been

Table 6.1. Weight of Contaminant in High-Concentration Cement Disks

Contaminant	Weight in Disk, g	Disk Concentration, %	Volume-Averaged Concentration, %
Hg	59.0	12.9	1.6
²³⁸ U	55.5	11.4	0.8
²³² Th	54.2	11.5	1.8
Cr	68.1	16.5	1.2
Pb	79.5	22.5	0.6
Ba	78.1	17.6	0.9
Cd	10.3	2.4	0.2

averaged over a volume determined by the collimator field of view and an effective interrogation depth. The effective interrogation depth is a function of gamma ray energy and is defined as the depth at which the gamma ray intensity of a planar source is 25% of the intensity of the same source at the surface. This definition is based on previous experience in developing depth algorithms for soil where it was found that, for a uniform distribution of contaminant, the contribution from depths below the effective interrogation depth is less than a few percent.

The effective interrogation depths for the two principal gamma rays from each of the isotopes in Table 6.1 as well as from Cl , ^{235}U and ^{99}Tc are given in Table 6.2 with the corresponding gamma ray branching ratios. Note that, because the effective interrogation depths are generally in the range of 5-15 cm (2-6 inches), the effective volume sampled is much larger than the volume of the concrete contaminant disk, and the volume-averaged concentrations are correspondingly lower than the actual concentrations.

As expected, detection sensitivities obtained with the original N-SCAN system for near-surface concrete contaminants were generally low because this system had been optimized for interrogation over larger depths and volumes. However, the following could be concluded based on these measurements:

Table 6.2. Main γ Energies and Interrogation Depths of Contaminants Listed in Table 6.1

Contaminant	First γ Energy (BK), keV	Interrogation Depth in Concrete, in.	Second γ Energy (BR), keV	Interrogation Depth in Concrete, in.
Fg	368.1 (0.814)	2.5	1693.9 (0.141)	4.3
$^{235}\text{U}^a$	1001 ^b (0.006)	3.5	765 ^b (0.003)	3.0
$^{232}\text{Th}^a$	239 ^c (0.47)	2.1	583 ^d (0.86)	3.0
Cr	835.1 (0.269)	3.3	749.2 (0.110)	3.0
Pb	7367.7 (0.941)	6.0	6736.4 (0.050)	6.0
Ba	1435.3 (0.141)	4.0	627.5 (0.123)	3.0
Cd	558.6 (0.727)	3.0	651.3 (0.139)	3.0
Cl ^e	1164.7 (0.199)	3.5	1950.9 (0.217)	4.5
$^{99}\text{Tc}^e$	172.2 (0.488)	2.0	299.5 (0.115)	2.3
$^{235}\text{U}^e$	642.2 (0.58)	3.0	687.5 (0.18)	3.0

a Passive mode counting is assumed for ^{235}U and ^{232}Th .

b decay gamma from ^{234}Pa , a member of the ^{238}U series. ^{234}Pa is usually in secular equilibrium with ^{238}U , and its decay gamma radiation may be used to characterize ^{238}U .

c decay gamma from ^{232}Pb , a member of the ^{232}Th series. ^{232}Pb is usually in secular equilibrium with ^{232}Th , and its decay gamma radiation may be used to characterize ^{232}Th .

d decay gamma from ^{208}Tl , a member of the ^{232}Th series. ^{208}Tl is usually in secular equilibrium with ^{232}Th , and its decay gamma radiation may be used to characterize ^{232}Th .

e element not listed in Table 2 but discussed later

1. Cd and Hg, with the highest thermal neutron capture cross sections and gamma ray branching ratios, had the highest detection sensitivities.
2. Passive-mode sensitivity for ^{232}Th and ^{238}U is better than active-mode sensitivity with the original system.
3. Generally, lower sensitivities were obtained for Cr, Ba and Pb. The latter element has extremely low detection sensitivity because of its extremely low thermal neutron capture cross section.

With regard to elements such as Pb, it should be noted that the current N-SCAN timing is configured to optimize detection of prompt gamma rays resulting from thermal neutron capture. However, with proper timing and neutron intensity changes, N-SCAN can be optimized to detect gamma rays from fast neutron-induced prompt reactions which occur during the neutron generator burst. With the revised system optimization, much greater detection sensitivities would be obtained for elements such as lead. Due to the scope and funding of Phase I, this alternative optimization could not be pursued.

Calculations using the analytical model were compared with the thermal neutron capture results for Hg placed at three different depths, and the experimental and calculated responses agreed well. These results were described in Section 5.

6.2.2 Modified N-SCAN System

The original N-SCAN system was designed to detect contaminant elements in large volumes of soil. Since the principal need of the DOE is to detect trace contaminants located at or near the surface of concrete, the N-SCAN system configuration was modified to enhance its detection sensitivity in the surface region. A series of experiments was performed to study the effect of each modification on sensitivity using the high-concentration Hg disk as a gauge of system performance. More than one hundred experiments, consisting of several hundreds of PGNAAs runs, were carried out with the N-SCAN system in this series. The system performed without any component breakdown during this period, which attests to its high degree of reliability.

Geometric modifications were made to the system in order to maximize the thermal neutron flux at the surface of the concrete. These modifications include changes in the reflector material and configuration, collimator position, and detector-to-generator distance. The system electronics were adjusted to provide better noise suppression. Finally, adjustments were made to the N-SCAN pulse timing to better synchronize the acquisition of gamma data with neutron thermalization times.

Because of adjustments made to the detector height above the floor and to the collimator configuration, the surface diameter "viewed" by the detector, as illustrated in Figure 6.2, is 11 inches in the modified system. The corresponding

dimension in the original system is 6 inches. Therefore, the volume-averaged concentration resulting from the same weight of a contaminant evenly distributed in the interrogation volume is smaller in the modified configuration than in the original configuration due to the larger volume being interrogated in the modified configuration.

Following the implementation of these modifications, a gamma spectrum was taken with the high-concentration Hg disk located under the detector at the surface of the floor. The results of this run are compared in Table 6.3 with those of a run made with the original configuration using the Hg disk at the same location. It is evident that the modified system has greater sensitivity for the characterization of mercury in the surface region. A six-fold improvement is observed in the signal-to-background ratio of the main Hg peak, in spite of the fact that a greater volume of concrete was irradiated in the modified system. Furthermore, the modified configuration resulted in fewer thermal-neutron capture reactions in the detector material, as indicated by the reduction of the area under the germanium peak. This reduction contributed in part to the observed decrease of the system dead time, allowing the detector to record more gamma rays originating from the interrogation volume in the floor.

Building on the encouraging results obtained with the high-concentration Hg disk, an experiment with a Hg disk of intermediate concentration (1236 ppm disk, 60 ppm volume-averaged) was performed. A mercury signal with a 35% uncertainty due to counting error was established after a 50-minute run. Based on this result, the projected sensitivity for Hg as a function of time is plotted in Figure 6.3, using a standard deviation of 50% (i.e. 95% confidence) as the detection criterion.

Following the Hg experiments, the sensitivity of the system for Cd was obtained from a run with the high-concentration Cd disk. Based on the experimental result, a plot of the projected sensitivity for Cd as a function of time is shown in Figure 6.4. A comparison of Figures 6.3 and 6.4 reveals that the projected sensitivity for Cd is better than that for Hg. Due to neutron self-shielding effects in the high-concentration Cd disk, an additional five-fold improvement in the detection sensitivity for Cd at low concentrations is anticipated.

Experiments with the high-concentration ^{238}U , ^{232}Th and Cr disks were also carried out. Plots of the projected sensitivities for these three contaminants are shown in Figures 6.5, 6.6, and 6.7. Table 6.4 shows the projected limits of detection for Hg, Cd, ^{232}Th (passive mode), ^{238}U (passive mode) and Cr based on a 50-minute

Figure 6.2 — Interrogation Volume as Viewed by the HPGe Detector in the Modified N-SCAN Configuration

Figure 6.3 — Mercury Limit of Detection of the Modified N-SCAN System

Figure 6.4 — Cadmium Limit of Detection of the Modified N-SCAN System

Figure 6.5 — Uranium-238 Limit of Detection of the Modified N-SCAN System

Figure 6.6 — Thorium-232 Limit of Detection of the Modified N-SCAN System

Figure 6.7 — Chromium Limit of Detection of the Modified N-SCAN System

Table 6.3. Comparison of Modified and Original N-SCAN System Performance Using Response from High-Concentration Mercury Disk

Performance Indicator	Original System	Modified System
Net Area under 368-keV Peak (from Hg) per Source Neutron, arbitrary units	1714	3750
Background Area under 368-keV Peak per Source Neutron	6136	2148
Signal/Background Ratio of 368-keV Peak	0.28	1.75
Area under 596-keV Peak from Germanium per Source Neutron	1330	213
System Dead Time	34.6%	10.7%

run using the modified N-SCAN system in its current state. The table also shows the limits of detection for Cl, ⁹⁹Tc, ²³⁵U, Ba and Pb, which are calculated from the Hg results using the following equation:

$$PL_i = ML_{Hg} \times \frac{A_i \sigma_{Hg} BR(\gamma_{Hg}) D(\gamma_{Hg}) V(\gamma_{Hg})}{A_{Hg} \sigma_i BR(\gamma_i) D(\gamma_i) V(\gamma_i)} \quad (1)$$

where

i = Cl, ⁹⁹Tc, ²³⁵U, Ba or Pb

PL_i = projected limit of detection for element i, ppm

ML_{Hg} = measured limit of detection for Hg, ppm

A = atomic weight, g mole⁻¹

σ = thermal neutron cross section for radiative capture reaction leading to a prompt γ, cm²

BR(γ) = branching ratio of most prominent prompt γ

D(γ) = detector efficiency at energy of most prominent prompt γ

V(γ) = interrogation volume based on energy of most prominent prompt γ, cm³

The limits of detection listed in Table 6.4 are for the modified N-SCAN system achieved in Phase I. There are additional planned modifications that were not implemented in Phase I due to budgetary and time constraints which are expected to further lower these limits of detection. These modifications, discussed in the next section, will be implemented in Phase II.

Table 6.4. Limits of Detection of Modified N-SCAN System Based on a 50-Minute Run

Contaminant	Projected Limit of Detection ^a , ppm
Hg	35
Cd	10
²³² Th ^b	18
²³⁸ U ^b	140
Cr	2220
Cl ^c	370
⁹⁹ Tc ^c	1100
²³⁵ U ^c	300
Ba ^c	2.6 E 04 (2.6%)
Pb ^c	2.0 E 05 (20%)

a volume-averaged concentration

b system in passive mode

c limit of detection for this contaminant is calculated from Equation 1

6.3 PLANNED SYSTEM IMPROVEMENTS

There are two principal factors that limit the performance of the modified N-SCAN system :

1. The current system is operated with the neutron generator output set at its lowest setting because the increased background count rates resulting from higher settings of the generator overload the capacity of the current system electronics to process the data. This effect produces high detector dead times and ultimately reduces the sensitivity of the system. In order to overcome this effect and take full advantage of the maximum output of the neutron generator, the background counting rate must be suppressed.
2. Although the thermal neutron flux incident on the interrogation surface is higher in the modified than in the original system, calculations show that the incident thermal neutron flux observed in the modified system is still substantially lower than the maximum thermal flux that can be achieved based on the output of the neutron generator. Since the signal intensity resulting from thermal-neutron capture reactions in a surface contaminant is proportional to the incident thermal neutron flux, maximizing this flux at the surface will increase the sensitivity of the system.

An outline of the modifications of the N-SCAN system planned for Phase II that are expected to overcome these two limitations and improve the overall

performance of the system is presented below:

1. Adjustment of electronics to minimize pulse (gamma) pile-up and improve signal/background ratio.
2. Adjustment of the neutron pulse timing to further improve the signal/background ratio.
3. Addition of a thermal neutron absorber around the detector assembly to reduce thermal neutron reactions in the detector materials, a major source of interfering gamma rays.
4. Replacement of the NaI annulus with a smaller bismuth germanate (BGO) annulus of the same detection efficiency. A more compact HPGe-BGO detector assembly will be easier to shield from neutrons than the current HPGe-NaI detector assembly.
5. Replacement of the D-T neutron generator with a D-D generator. Although a D-D generator produces fewer source neutrons, a higher thermal neutron flux at the surface is expected with the use of a D-D source because of the lower energy of the source neutrons.
6. Additional modification of the neutron reflector to maximize the thermal neutron flux incident on the disk.

These modifications are expected to improve system sensitivity by one order of magnitude using a single detector, as in the present system. The N-SCAN system to be developed in Phase II is expected to have four detectors, which will further lower the limits of detection.

The passive-mode detection sensitivity of N-SCAN can be improved by the addition to the system of an HPGe detector with an efficiency of 160% relative to a 3" x 3" sodium iodide detector. This commercially available detector would acquire data in complete passive mode while the other detector(s) would work in active mode. This active/passive system is expected to yield an improvement of a factor of eight in passive mode sensitivity.

Other modifications intended to facilitate data collection and analysis will be implemented in Phase II. Automation of the system to collect and analyze data can be readily achieved with minor software upgrades. The software of the current N-SCAN system already includes a library of prompt gamma lines which can be used to automatically identify contaminants from signature gamma peaks. Programs to convert peak areas to contaminant concentration will be included in the software.

6.4 DISCUSSION

The modifications made to the N-SCAN system to increase its sensitivity to contaminants in a surface region of concrete have resulted in an improvement in the limits of detection of the suite of contaminants under consideration in this program. Additional improvements are expected to further lower these detection limits. Table 6 provides a comparison of projected limits of detection of the single-detector Phase I

system with the projected detection limits of an improved, four-detector system expected upon completion of Phase II. These limits of detection are for a 50-minute run, with a standard deviation of 50% (i.e. 95% confidence) used as the detection criterion.

The limits in Table 6.5 indicate that the Phase II N-SCAN system would meet regulatory requirements for the characterization of mercury, cadmium, and ²³²Th. On the other hand, the LLD of N-SCAN for lead and barium are well above the regulatory limits for these contaminants. In the case of chromium, an N-SCAN sampling time of 60 minutes will be necessary to meet the regulatory requirement. The effectiveness of N-SCAN as a characterization tool for the other contaminants listed in Table 6.5 will depend on site-specific limits for these contaminants.

A functioning N-SCAN system can either reduce or eliminate the amount of analytical sampling required to characterize large areas, depending on the type of contaminant(s) present. In these cases, the use of N-SCAN would result in a reduction of the occupational risks involved with analytical sampling. Since N-SCAN does not generate any waste, is non intrusive and is capable of depth profiling, its use will also reduce the amount of waste generated relative to other intrusive and waste-generating methods. It should be noted that, in addition to the in situ characterization of concrete, N-SCAN can also be adapted to perform real-time, non-intrusive characterization of relatively large volumes of materials in containers (ex. waste drums, construction debris generated during D&D activities). The ability to address these different applications is an important benefit of this technology.

Table 6.5. Comparison Between the Projected Limits of Detection of N-SCAN in Phases I and II

Contaminant	Projected Limit of Detection, ppm		Regulatory Limit, ppm
	Phase I	Phase II	
Hg	35	2	4
Cd	10	0.5	20
²³² Th	18	7	45.4
²³⁸ U	140	46	na
Cr	2220	110	100
Cl	370	25	na
⁹⁹ Tc	1100	63	na
²³⁵ U	300	17	na
Ba	2.6 E 04 (2.6%)	1500	100
Pb	2.0 E 05 (20%)	1.2 E 04 (1.2%)	100

7. LIMITATIONS OF PGNA AND COMPARISON WITH SOME COMPETING METHODS

The most severe limitation of PGNA as a characterization technology is that its sensitivity for an element is dependent on an optimal combination of the element's nuclear cross section, gamma ray energy and gamma ray branching ratio. Therefore, while N-SCAN's sensitivity for elements like mercury and cadmium is excellent, its sensitivity for others such as lead and barium is poor. As a result, N-SCAN cannot be the sole means of characterization at a site whose contamination history is unknown, or at a site where the sensitivity of N-SCAN for the principal contaminant is insufficient for regulatory purposes. In addition, while N-SCAN can provide information on the depth profile distribution of a contaminant, its spatial resolution is relatively poor.

These limitations suggest that N-SCAN cannot totally replace analytical sampling as a means of characterization. However, N-SCAN can reduce the amount of sampling required by serving as a screening or "first look" tool. In cases where the site contaminants are known to have good PGNA characteristics, N-SCAN may be an adequate substitute for analytical sampling, thereby avoiding the high cost and lengthy analysis time associated with sampling.

N-SCAN's main advantage over characterization technologies based solely on radiation detection is that these latter methods cannot detect any nonradioactive contaminants. Some of these methods are limited to the detection of alpha emitters and are restricted in their ability to differentiate between two or more alpha-emitting contaminants. Furthermore, most of these methods are purely for surface characterization. N-SCAN can detect a wide range of both radioactive (active and passive mode) and nonradioactive (active mode) contaminants, and has the capability to detect subsurface contamination.

PGNA systems that utilize ^{252}Cf as a neutron source are normally restricted to the detection of bulk quantities because of the high gamma background produced by this source. Furthermore, ^{252}Cf is a constant radiation hazard and the bulky shielding required for the source complicates the field-deployment of this type of PGNA system. Permits and licenses for the operation of neutron generators in the field are more readily obtained than those for the operation of transuranic sources such as ^{252}Cf .

Prompt Fast Neutron Analysis (PFNA) systems are also restricted to the detection of bulk quantities because of their limited sensitivity. PFNA is still considered to be a laboratory system, and a considerable amount of development is

needed before this technique can be used for the in situ detection of trace amounts of contamination.

8. SUMMARY

The first phase of a two-phase project to develop a fully-integrated PGNAA system for the in situ characterization of contaminants in concrete floors and walls has been completed. During this phase, a comprehensive experimental program was undertaken to determine the sensitivity of the Westinghouse N-SCAN PGNAA system for several contaminants. The experimental work was performed using a concrete floor mockup built to emulate anticipated field conditions.

The configuration of the N-SCAN system, which was initially developed to detect contaminant elements in large volumes of soil, was modified to enhance the system's sensitivity to trace contaminants located at or near the surface of concrete. These modifications resulted in a six-fold improvement in signal-to-background ratio. The projected lower limits of detection (LLD) of the modified system were determined for mercury, cadmium, uranium-238, thorium-232, chromium, barium, lead, technetium-99, uranium-235 and chlorine. These limits show that N-SCAN has good sensitivity for Hg, Cd, ^{238}U , ^{235}U , Cl and ^{232}Th . However, the sensitivity for Pb and Ba was poor. Further system improvements planned for Phase II were discussed. The projected LLD of N-SCAN after these improvements are made are expected to be at least an order of magnitude better than those of the current system.

An analytical model of the N-SCAN PGNAA response in soil was modified to model the response in concrete. The preliminary predictions of this model for the case of mercury were within a factor of two of experimental results.

9. REFERENCES

1. "Prompt Gamma Rays from Thermal-Neutron Capture," M. A. Lone, R. A. Leavitt and D. A. Harrison, Atomic Data and Nuclear Data Tables, No. 26 (1981).
2. Westinghouse Advanced Reactors Division LRA-80-719, "DOTIII-W Program Description", T.C. Chan and R.K. Disney, March 4, 1981. Non-proprietary versions of DOT are available from the Radiation Shielding Information Center (RSIC).
3. CCC-543, "TORT-DORT, Two- and Three-Dimensional Discrete Ordinates Transport, Version 2.8.14", W.A. Rhoades, contributed by Oak Ridge National Laboratory to the Radiation Shielding Information Center (RSIC); DORT is available from RSIC.
4. "Detection and Depth Profiling of Hazardous Elements Using N-SCAN™ Prompt Gamma Neutron Activation Analysis", F.H. Ruddy, T.V. Congedo, A.R. Dulloo, J.G. Seidel, Westinghouse Science and Technology Center, R.P. Williams and D.H. Weigle, Scientific Ecology Group; to be presented in June, 1995 at the American Nuclear Society Annual Meeting in Philadelphia, PA.