

# NON-DESTRUCTIVE IDENTIFICATION OF UNIDENTIFIED RADIOACTIVE MATERIALS USING MULTIPLE DATA INPUTS

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## ABSTRACT

The Feed Materials Production Center (FMPC), is a Department of Energy (DOE) facility, that produces high purity uranium for defense programs. It also serves as the DOE's repository for temporary storage of thorium materials. The containers storing the thorium materials have suffered from extensive environmental deterioration and with the passing of time records have been lost. The net result is that a significant number of containers have lost their identifying markings. The exact type of materials in some of the containers is unknown. The containers must be repackaged for Environmental Safety and Health (ES&H) reasons. Prior to repackaging the identity of the contents must be ascertained. However, these containers can not be opened due to As Low As Reasonably Achievable (ALARA) and ES&H considerations.

A method was developed to determine the content of the containers, without physical inspection, prior to the repackaging/overpacking. This method utilizes a computer based data base management system which incorporates statistical analysis of the available input data and assigns statistical weighting to the various inputs, based on the significance of the data. The data consists of the following: historical data on the containers, if it exists; the weight of the containers and materials; the gamma energies related to the various materials; the geometry of the container and materials; the absorption coefficients of the materials and containers; the self-shielding of the materials; external contamination data, if any; and any other data obtained either prior to or during the overpacking effort. This system will work not only for the thorium materials, but the system has the capability to perform similar functions at other facilities faced with similar concerns.

## INTRODUCTION AND PROJECT OVERVIEW

### Background and Historical Information

The FMPC located also produced between the 1950's and 1970's thorium materials for use in the thorium/uranium fuel cycle. In a nuclear reactor, thorium can be converted to a fissile isotope which can be used as a nuclear fuel. The use of a thorium/uranium fuel cycle was studied extensively in the 1960's for its potential as an efficient energy source. A demonstration of this technology was done at the Shippingport Atomic Power Station. The thorium/uranium nuclear fuel is not currently in use in the United States. The thorium materials now in interim storage at the FMPC were originally slated for use as part of the thorium fuel cycle.

Since about 1972, the FMPC has served as the DOE storage site for thorium. Approximately two-thirds of the thorium materials at the FMPC were processed on-site, with the remaining one-third originating at other DOE facilities. The thorium compounds stored at the FMPC are primarily mixtures of thorium metal in pure metallic form, thorium oxides, and processing residues.

### FMPC Thorium Inventory Data

The thorium at the FMPC is in various forms. Some of the materials are stored in bulk storage in an above ground silo and double bin. The remainder of the thorium materials, approximately 975 metric tons (thorium weight), are stored in four warehouses, an outside controlled pad, and other small locations on site. There are over 13,300 + individual containers. These containers consist of 322, 208, and 144 liter drums (85, 55, and 30 gallon), 19 and 4 liter size cans ( 5 and 1 gallon), various metal and wooden boxes, and bound thorium metal rods.

The informational system for the materials consisted of, the marking of the drums, attached labels to the containers, and the inventory that was originally done on the materials. The materials have not been physically inventoried for a number of years since the buildings and locations where the materials are stored are maintained under locked conditions. These materials have been stored in these locations for over twenty years and the net result being, a loss of information.

### Thorium Container Condition

These containers have been in the storage locations since the mid 1960's and 1970's. This extended period of storage has resulted in the containers suffering from standard environmental deterioration that is normally associated from weathering over a prolonged period of time. The markings on the containers have suffered equally from this environmental deterioration. The marking on the containers is becoming unreadable and some of the containers are approaching extreme environmental deterioration.

Compounding this identification concern was the complexity involved in the handling and overpacking, resulting from the fact that 21 of the containers contain potentially pyrophoric materials. The identification of these materials is critical so as to assure that sufficient safety concerns are addressed. It was necessary to develop an identification system that would assure that these containers are identified and removed from the overpacking materials stream so that they can be dispositioned under a separate program. The net result of this is that the system for the identification of radioactive materials had to be reliable to the 99.5% criteria.

### Project Scope

It was decided that an engineering approach to the

handling, identification and overpacking system for the 13,300+ containers of thorium materials needed to be developed. The basic system had constraints including: the system had to be remote in operation, incorporate extensive shielding, assure that all ALARA considerations were met or exceeded, providing ease of operator control and operations, and be automate to the maximum extent possible; all within the prescribed criteria, schedule and cost constraints.

The above criteria and project scope presented unique engineering problems. The method that was chosen was based on the fact that each radioactive element produces a unique gamma energy signature and that when the materials are in specific configurations they produce different fields. Specifically the higher the purity of the thorium, the higher the gamma field produced. The net result of this led to an investigation of the storage locations of the materials to verify if this contention was correct.

A detailed survey of the various storage locations was conducted and a grid of readings was done to determine a isoplot of the gamma fields (Fig. 1 & 2) within the warehouses relative to the materials that could be identified. The result of this survey showed that distinctive fields around specific type of materials was detected. Additionally the readings showed that the thorium that was mixed with various other components showed homogeneous gamma reading from the materials and was always within 3-4 percent of anticipated readings. Since over 8,000 containers of the FMPC thorium inventory are within this group it was a significant discovery. The investigation also turned up a unique, and at the time unexplained high gamma reading in one of the warehouses around a specific group of containers.

This high gamma reading was outside of the signature of the thorium materials in its intensity. A historic review of the containers within this location indicated that the materials inside had been treated with fissile  $U^{233}$ , a major factor contributing to the high gamma readings indicate the type of materials present and can assist in the verification of the content of the containers. A statistical review of all of the data, including the information obtained as to the container contents, the gamma readings, and the inventory of the materials, indicate that there is a significant correlation between the gamma readings and the material types.

The investigation continued to look at the weight of the various containers, the uniqueness of various components of the identification numbering system, the type of containers that the materials are contained within, and the location of the materials. From this information it was determined that the identification of the materials was possible using all of the above parameters and a correlation could be obtained within the 99.5 percent confidence level.

There were other correlations that could be obtained from the amount of material within the containers, the number of containers having overpacks within them and the resultant shielding that comes from both the materials and the containers. These materials produced significant self shielding in the purer thorium materials. The pure metallic

thorium shields most of the low energy gamma energies while the 1.8 to 2.4 MeV energies pass through the materials. This produced a higher energy signature with almost no low energy gamma emissions from the materials. The few that are not effected by the materials are shielded by the containers themselves. The result of this allowed for the ease of identification of the pure thorium materials. The mix materials did not have this effect and the majority of all the gamma energies were released. The fact that the fields were lower was simply related to the source term of thorium present in the matrix.

#### The Operating System and How It Interfaces

The automated system that is being used consists of a belt conveyer that receives containers from a shielded fork-lift. Once the container is placed on the end of the conveyer it is moved to various stations. These stations consist of, or perform, the following functions:

1. Weight of the container,
2. Perform a gamma reading on the container,
3. Perform an external alpha contamination scan,
4. Closed circuit television performs detailed visual investigation,
5. The containers are moved into various orientations for additional identification and investigation, and
6. The containers are moved to the overpacks for disposition, or are identified as pyrophoric and are overpacked in special containers.

All this work is done from behind a shield block wall from remote consoles. The personnel are working in full bubble suits for additional protection from potential airborne contamination.

All of the data are entered into specific data fields by the operator. This information is then cross-referenced against a data base management program, assigned weighted values for statistical evaluation, and a determination made by the computer program as to the identification of the container, its content, and disposition. The system also correlates the information as to the containers that are being placed in the six-pack and produces a data sheet as to the contents of the container, the location of the containers inside of the six-pack, the weight of the six pack, and produces all the required information for the shipping and storage documentation. A bar code is then generated and attached to the six-pack that cross-references to the original data base management records system. Data is kept on laser disc for long term retrievable data storage (2).

The data already described showed that there was a correlation between the thorium materials and their gamma fields. Each thorium material type produces a unique gamma field related to the type and amount of material in the container. The method of calculation was based on the reverse method for determining shielding calculations. The formula that is being used (Fig. 3) takes into consideration: gamma ray intensity; source strength; build-up factors; distance from the unit to the measuring point; the effective enter of the unit; sievert function; thickness of the

container; macroscopic cross-section of the source material; and the probe geometry and container geometry.

All of these factors are integrated into the data base management system, and using advanced statistical calculation methods are correlated against the known data fields. Additionally background radiation is removed from the equation with the use of a background subtraction method from a shielded gamma probe dedicated to this purpose. This allows other operations to occur around the operation as well as the ability for the operation to be moved to remote locations. The alpha scanner is used only for determination of exterior contamination is on the containers and if it is above the levels for shipping of materials, as required by Department of Transportation (DOT). If a container is found to be contaminated it is removed from the overpacking path and decontaminated prior to overpacking.

#### Applicability of This Method to Other Waste Operations

It has been demonstrated that there exists a capability for the identification of radioactive materials by their form, integration of historical records, and with the use of the gamma energy extrapolation. The question then posed is that since this system will work for thorium materials will it work for other radioactive materials. The key to this is the uniqueness of the materials being identified, the gamma spectrum that is exhibited from the materials and the effects of materials combined with other radioactive materials and how many radioactive elements are present in each of the containers. Materials such as plutonium, radium, and thorium have some very distinctive gamma energies and fields that exist around them. These would be prime candidates for this type of process. The problems that can occur is if the detection of various types of uranium are required. This can cause problems since the gamma energies that are coming off of related uranium materials are close enough that a clear distinction can not be obtained.

If this is not the case, than this method will greatly enhance the ability for the identification of the materials and the reduction of the gamma dose to the personnel involved in this operation since the entire operation is remote. The use of computers to perform this work can greatly enhance the accuracy of the findings, the speed in which the operation is performed, and the analysis of the materials that are being investigated. Special application of this method could be for the identification of contaminated soils, identification of contaminated metal and machinery, and other similar applications where known contaminants exist. It needs to be stressed that a significant amount of up-front work needs to be accomplished prior to the start of this type of operation and the computer support for the development of the methods that apply to a specific circumstance exist.

A primary benefit that can be obtained from this type of operation is the significant reduction in gamma exposure. The following is a synopsis of the gamma reductions that will be achieved during this operation.

#### Dose Reduction

One of the primary drivers during the development and design of the systems for the thorium handling and repack-

aging effort is the need to adhere to the ALARA philosophy and design a system that minimizes radiation dose to the operational personnel by over 90% compared to uncontrolled handling and overpacking operations. The total dose to operations personnel during the overpacking operations is expected to be reduced from approximately 2.7 Sv (270 rem) to less than 0.5 Sv (5 rem). A summary of the ALARA review and Analysis report is shown in Table I.

TABLE I  
ALARA Report Composite

<u>Area</u>	<u>mRem Dose</u>	<u>mRem Dose</u>
	No Controls	W/Controls
Building 1	7,944	30
Building 2	147,222	2,504
Building 3	52,924	941
Building 4	61,701	771
Total	269,791	4,356

#### SUMMARY AND CONCLUSIONS

It has been demonstrated that the use of the above described system will work for thorium materials. It is logical to assume that other radioactive materials can also be identified by similar methods. The key is the identification of the unique energies that are produced from the various materials, confidence in historical records, and extensive investigations prior to the start of work.

In addition to the simplicity of material identification, the reduction in dose in using a remote and semi-automated system cannot be understated. The reduction of a minimum of 80% should be able to be realized for the overpacking and handling operation. If shielded fork lifts and other dose reduction engineering steps are taken this can even be enhanced. The use of the laser disc provides a very long term retrievable storage medium. The use of Closed Circuit Television Camera significantly reduces personnel exposure and the conveyer and remote crane further assists in this radiological reduction.

It is realized that this system will not work for all operations it is also understood that a number of facilities with concerns similar to the type we have encountered could significantly reduce there exposure and more effectively expedite there identification of radiological materials in various materials.

#### REFERENCES

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2. Mihalovich, G.S., Heisler, Jr., S.W., "Demonstration of New Handling Equipment Configurations for the Over packing of Thorium Materials", 1988, FMPC-2141, U.S. DOE Model Conference, Oak Ridge, Tenn.

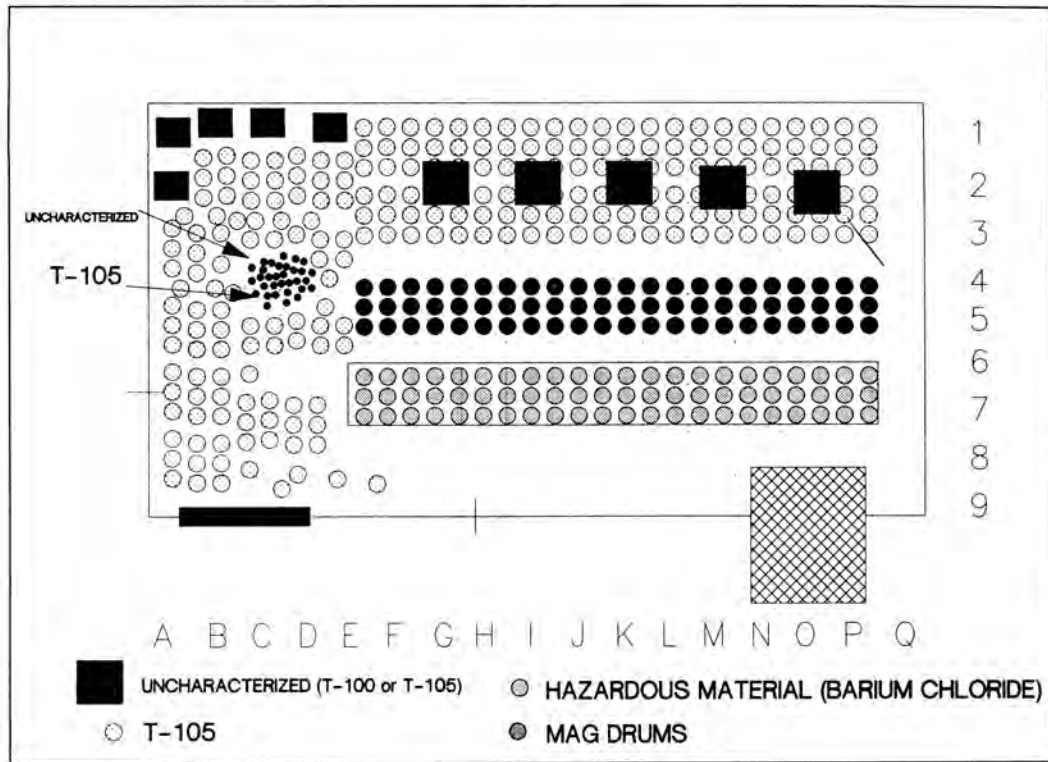


Fig. 1. Isoplot of the Gamma Fields.

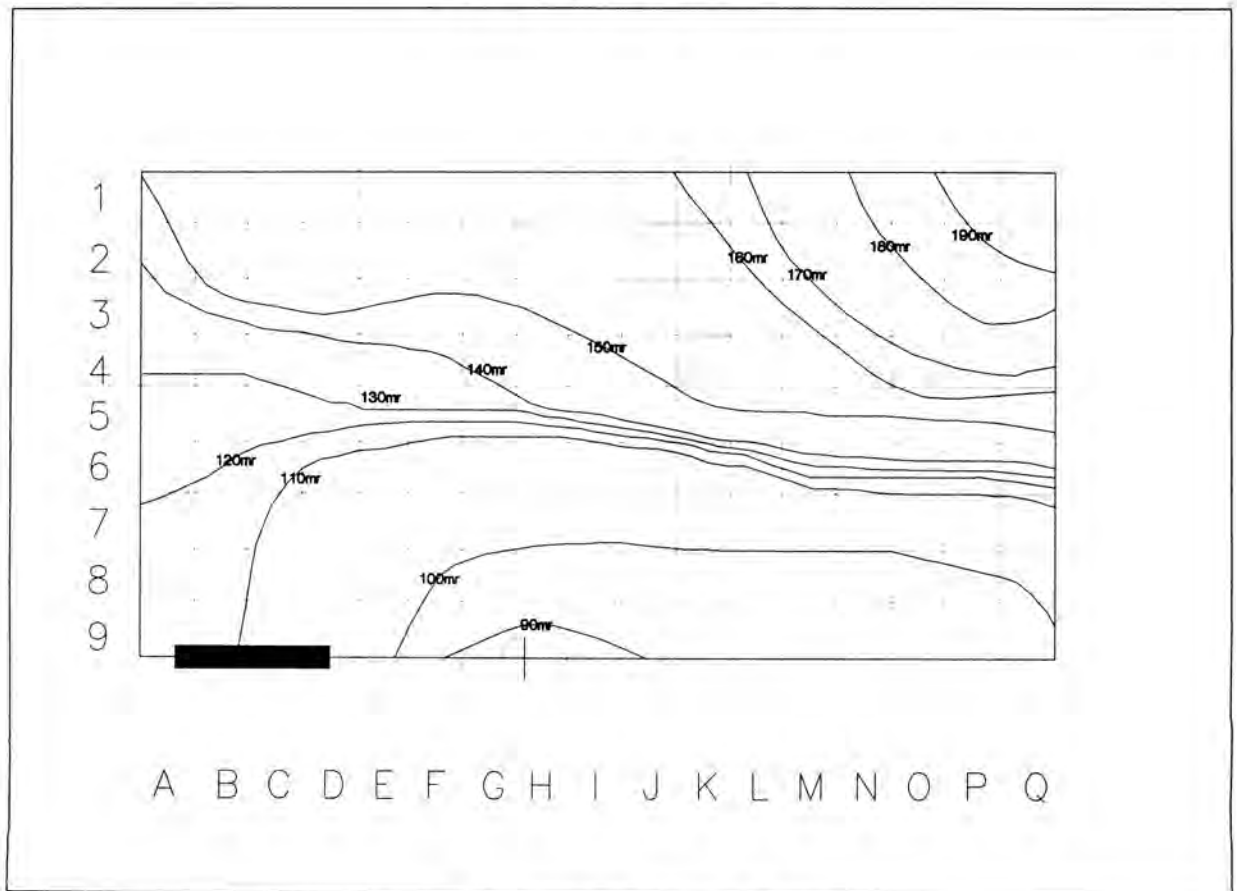


Fig. 2. Isoplot of the Gamma Fields.

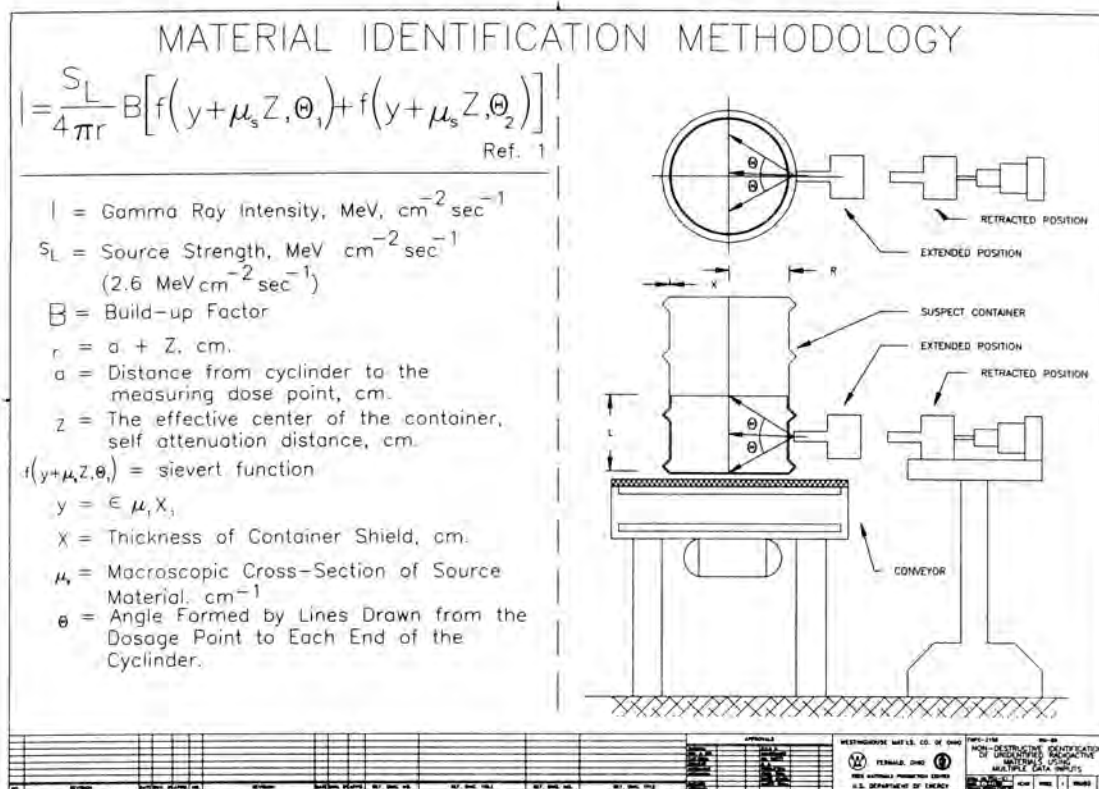


Fig. 3. Material Identification Methodology.