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**Technical Description of Candidate
Fluorescence Compounds and
Radioisotopes for a Nuclear
Smuggling Deterrence Tag (IL500E)**

**S.D. Hartenstein
R. Aryaeinejad**

MASTER

Lockheed
Idaho Technologies Company

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(IL500E)**

**S. D. Hartenstein
R. Aryaeinejad**

Published March 1996

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**INEL-96/0087
March 1996**

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ABSTRACT

This report summarizes the efforts completed in identifying candidate fluorescence compounds and radioisotopes for a developing tagging system. The tagging system is being developed as a deterrent to nuclear smuggling, by providing a means of: 1) tracing materials and pilferers to the facility of origin for any recovered special nuclear materials, 2) inventory control of long-term stored items containing special nuclear materials, and 3) tracking materials transferred between facilities. The tagging system uses four types of tagging materials to cover a range of applications intended to prevent the pilfering of special nuclear materials. One material, fluorescent compounds which are invisible without ultraviolet or near-infrared detection systems, is marked on controlled items with a tracking pattern that corresponds to a specified item in a specified location in the data control system. The tagging system uses an invisible, fluorescent dusting powder to mark equipment and personnel who inappropriately handle the tagged material. The tagging system also uses unique combinations of radionuclides to identify the facility of origin for any special nuclear material. Currently, 18 long-lived radioisotopes, 38 short-live radioisotopes and 10 fluorescent compounds have been selected as candidate materials for the tagging system.

EXECUTIVE SUMMARY

This document represents the first deliverable for the Idaho National Engineering Laboratory's Warhead Dismantlement/Special Nuclear Materials Control Program (IL500) Task E "Radioisotope Cocktail for Nuclear Smuggling Deterrence." This project is funded through the Department of Energy Office of Nonproliferation and National Security's Office of Research and Development (NN-20). The purpose of this project is to develop a tag which will deter the pilfering and illegal trafficking of special nuclear material (SNM). The deterrence to pilfering of SNM achieved by using the tag to improve the accountability of SNM and to provide readily traceable forensic evidence to trace facilities and personnel involved with the pilfering. The described tag is being developed for use by the international nuclear community.

This report describes the justification for this project, the technical requirements driving the selection of candidate tagging materials and methods, the proposed tagging methods, and the candidate materials for the tag. The tagging system is composed of three types of tags: 1) a fluorescent tag, 2) a radioisotope tag, and 3) fluorescent and radioisotopic tampering tag. These tags' relative importance and function are determined by their respective applications. In combination, the tags will uniquely identify the facility of origin for materials and their storage containers, will uniquely tag each container for inventory control, and will provide forensic information to assist in the identification of equipment and personnel involved in the handling or transport of pilfered SNM. The tag is based on the ability of solvents, containing fluorescent compounds and radionuclides, to become embedded into the SNM or metal storage containers as the solvent penetrates into the metal's surface microcracks. The tag will be colorless under normal lighting conditions and will be detectable by: 1) a gamma detector, 2) an ultraviolet light, or 3) a near infrared camera.

Design criteria for the tagging system cover the materials to be tagged, location of use, and physical characteristics of the tag. The tagging system will be designed to tag components and containers of at least 500 g of plutonium and uranium metals and salts; warhead components; unirradiated nuclear fuel rods; and storage containers. The tags will be applied in normal process areas at SNM handling facilities and be detectable with portable equipment throughout the handling facility and transport processes. The tag will be designed to be difficult to remove all traceable evidence of the tag from the SNM and the personnel and equipment used to remove the tag. Currently, 18 long-lived radioisotopes, 38 short-lived radioisotopes and 10 fluorescent compounds have been selected as candidate materials for the tagging system.

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ACRONYMS

CPM	counts per minute
DOE	United States Department of Energy
DOT	United States Department of Transportation
EPA	United States Environmental Protection Agency
IAEA	International Atomic Energy Agency
INEL	Idaho National Engineering Laboratory
NDAA	National Defense Authorization Act
NIR	near infrared
NRC	United States Nuclear Regulatory Commission
OSHA	United States Occupational Safety and Health Agency
SNM	special nuclear material
UV	ultraviolet light
VIS	visible light
WMD	weapons of mass destruction

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1. INTRODUCTION

This document represents the first deliverable for the Idaho National Engineering Laboratory's Warhead Dismantlement/Special Nuclear Materials Control Program (IL500) Task E "Radioisotope Cocktail for Nuclear Smuggling Deterrence." This project is funded through the Department of Energy Office of Nonproliferation and National Security's Office of Research and Development (NN-20). The purpose of this project is to develop tagging to deter the pilfering and illegal trafficking of special nuclear material (SNM). The deterrence to pilfering of SNM achieved by using the tag will be realized by enabling the SNM handling community to: 1) improve the accountability of materials being prepared or stored at facilities; 2) provide readily available forensic information related to the facility of origin for materials recovered from smugglers; and 3) provide traceable safeguarding measures and forensic evidence to detect personnel pilfering (or attempting to pilfer) SNM.

This report describes the justification for this project, the technical requirements driving the selection of candidate tagging materials and methods, the proposed tagging methods, the candidate fluorescence compounds and the candidate radioisotope compounds.

2. PROJECT DESCRIPTION

2.1 Purpose

The purpose of this project is to develop tagging methods for SNM, dismantled nuclear weapon components and their containers to determine unambiguously the point of origin of interdicted smuggled materials and to deter pilfering of these materials by personnel from facilities missioned with manufacturing, dismantling, or storing these materials.

2.2 Participants

This project is funded through the United States Department of Energy's Office of Non-Proliferation and National Security (NN-20) and is being performed at the Idaho National Engineering Laboratory (INEL) by Lockheed Idaho Technologies Company, Managing and Operating contractor under DOE Idaho Operations Office Contract DE-AC07-94ID13223.

2.3 Policy Drivers

Federal sponsorship of this research is justified through congressional charter, Section 1502 of the FY 1995 National Defense Authorization Act (NDAA). The NDAA directs the Counter proliferation Program Review Committee, chaired by the Secretary of Defense and composed of the Secretary of Energy, Director of Central Intelligence, and the Chairman of the Joint Chiefs of Staff to "identify and review existing and proposed capabilities and technologies for support of U.S. nonproliferation and Counter proliferation policy." The DOE's initiatives address enhanced programs in: material protection, control and accounting for the physical protection of Russian nuclear materials, expansion of the DOE's support to the International Atomic Energy Agency (IAEA), strengthening of support to U.S. and international commerce, preventing a black market in nuclear materials, and providing additional intelligence products in support of U.S. Intelligence. Within these initiatives, DOE is sponsoring research to build the technical foundation for a national capability to detect and track illicitly transported weapons of mass destruction (WMD) materials, including the ability to identify the source of materials and the participants in nuclear smuggling incidents.¹

2.4 Technology Users

The described tag is being developed for use by the international nuclear community as a deterrent to pilfering of SNM. The tag will be applied directly to SNM and storage containers so that the facility of origin of the materials can be identified, storage containers can be inventoried, and individuals improperly handling the materials can be detected. Facilities using the tag include those producing usable forms of SNM (metals, fuel rods, and salts), those dismantling nuclear weapons, and those preparing and monitoring SNM for long-term secured storage. Once developed, it is envisioned that the tagging methodology will be available (potentially via the IAEA) to each country handling SNM. Each facility handling SNM will be assigned its unique tagging materials and patterns and be able to obtain the instrumentation necessary to apply and detect the tags.

3. BACKGROUND

Since the first known incident of illegal trafficking of radioactive materials in 1966, DOE has provided support to law enforcement personnel in more than 450 cases. Of these cases, more than 30% have occurred in the last five years.² This trend becomes significantly alarming in that the trend is not localized in one area but is spreading throughout the international (Russia, Germany, Poland, etc.) marketplace. As an example, the number of cases of illegal transfers of radioactive and fissile material to Germany has grown from four cases in 1990 to more than 500 cases over the next four years.³

Even though most of the cases of nuclear smuggling involve scams, there are a significant number of incidents involving actual radioactive materials. Of the cases in Germany, forty-six cases resulted in the recovery of radioactive materials and fifteen cases involved fissile material.³ The fissile material recovered generally included small quantities of natural or low-enriched uranium. Some cases have involved the recovery of plutonium and americium from smoke detectors. However, smuggling incidents can involve quantities of materials that indicate an active market for nuclear proliferation materials. One of the most significant of these cases involved the recovery of 2.7 kg of 88% enriched U-235 powder in Prague.⁴

This incident, in combinations with claims that 150 kg of plutonium has been transferred from Russia and stored in Switzerland,² indicate that there is an increasing demand to secure existing stockpiles of SNM. Improved methods of security and accountability are needed to make it more difficult to gain access to fissile material and increasingly dangerous to handle the SNM. These deterrence measures can be realized by improving the chance that a person pilfering material is more likely to be caught during the act of pilfering or eventually tracked down using the forensic information gleaned from recovered materials.

Because of the inherent similarities of SNM throughout the international nuclear community, deterrence from forensic data has significantly lagged behind deterrence accomplished through security and accountability measures. This lack of forensic information has been recognized and is being addressed through international conferences and technology development activities. The tagging system described in this report is one of the measures currently underway to provide more usable forensic information to deter the pilfering of SNM.

4. DESCRIPTION OF TAGGING SYSTEM

The developing tagging system for this project is composed of three types of tags: 1) a fluorescent tag, 2) a radioisotope tag, and 3) a tampering tag. These tags' relative importance and function are determined by their respective applications. In combination, the tags will uniquely identify the facility of origin for materials and their storage containers, will uniquely tag each container for inventory control, and will provide forensic information to assist in the identification of equipment and personnel involved in the handling or transport of pilfered SNM. The tags are based on the application of a solvent containing a mixture of radionuclides and fluorescent compounds to the surface of the metal. The solvent, radionuclides, and fluorescent compounds become embedded into the metal as they penetrate into the microcracks normally present in metal surfaces. The components of the tag will be colorless under normal lighting conditions and will be detectable by: 1) a gamma detector, 2) an ultraviolet light, or 3) a near infrared camera.

4.1 Intended Applications for the Tagging System

To design a tagging system that meets its intended purposes, there must be an understanding of the intended applications for the tagging system. For this tagging system, there are four intended applications: 1) identifying the facility of origin for SNM recovered from smugglers, 2) identifying materials for inventory control, 3) tracing the transfer of materials between facilities, and 4) identifying personnel that improperly handle controlled materials. More detail describing these applications is provided as follows:

4.1.1 Identifying Point of Origin for Recovered Materials

In most incidents in which interdicted SNM has been recovered, the source of the smuggled material is difficult to determine. Currently, many national and international efforts are devoted to forensic analysis of these recovered materials to establish the facility from which the material was taken and possibly the people, vehicles, and locations involved in the transporting of the material. The purpose of a tag which identifies the facility of origin of the material will minimize the need for the current extensive practice of particle, residue, and isotopic analyses. Developing a tag which identifies the facility of origin achieves the following benefits: 1) the tag provides evidence which may assist in the arrest of personnel responsible for pilfering the material and 2) the tag improves safeguarding and accountability at the facility from which the material was pilfered.

4.1.2 Verifying Inventory of Stored Material

A significant aspect of safeguarding SNM is controlling the inventory of material that is placed in storage. Currently it is planned that enriched uranium and weapons grade plutonium that was prepared for or removed from nuclear weapons will be packaged in metal cans and stored in secure facilities. An integral part of safeguarding these stored materials will be periodic (potentially every day) inventory checks to determine if the stored material is in the storage container and if the storage container has been disturbed. One low risk, low exposure means of performing this inventory check may be robotic sensor systems that are controlled remotely for operation in limited access storage bunkers. To use such a system, the sensor system will have to detect either unique signatures from the SNM and storage containers or unique signatures from a tag embedded on the SNM and storage containers.

Because there is a strong possibility that the SNM and containers at a specific storage facility will have similar shapes, chemical composition, isotopic composition, and contaminant composition, resolution of traditional detectable signatures (neutron or gamma spectra and intensities) will be difficult to interpret to establish the identity and position of individual items. Therefore a unique tag placed on the SNM and on the storage container will offer an opportunity to develop a remotely controlled sensor system that can verify the unique identity of the material and its locations. Use of a unique tag would improve safeguarding of the SNM by making a remote inventory monitoring system more practical. Benefits of having an implementable remote inventory system based on unique tags would then be realized through: 1) minimizing personnel access opportunities to the storage bunker, 2) minimizing radiation exposure to personnel performing the inventory, and 3) increasing the rate of periodic/random inventories.

4.1.3 Tracing Material Movement Between Facilities

Another important feature of safeguarding SNM is the ability to verify the unique identity of materials that are shipped and received between facilities. This verification is done with a variety of methods which include: 1) administrative documentation, 2) stamped markings, 3) mass, 4) energetic spectral measurements, and 5) laboratory analysis of samples. A tag which identifies specific shipments or containers that have been transferred will reduce the time and expense of the verification process if the tag: 1) uniquely identifies the material; 2) can be read with inexpensive, non-labor intensive instruments; and 3) requires minimum unpacking and handling of the shipment material.

4.1.4 Identify Tampering with Materials in Process

Because materials have been pilfered from SNM facilities, known weaknesses of material safeguarding are the vulnerability of materials that are handled manually in high traffic areas and those left in storage in low traffic areas. Normally, the handling of SNM is performed in a glove box or secured room where access/exit is controlled. However, the large number of personnel in combination with the complex operations and equipment in these areas, provide opportunities for personnel to improperly tamper with or even remove materials from these facilities. Therefore, the known presence of a tagging material which would mark personnel or equipment will deter personnel from improperly entering a controlled area or improperly handling a controlled material.

4.2 Functional Features of Tagging System

The tagging system must be designed to meet all the functional needs for the above described tagging applications. These functions (shown in Figure 4.1) are achieved by using the physical and chemical characteristics of the materials used in the tag; the sensitivity and resolution of the detectors for the tagging materials, and the flexibility and control features of the data acquisition system. The tag materials (a combination of fluorescence compounds and radioisotopes) provide the uniqueness and traceability necessary to identify individual components and storage containers, facilities of ownership, tampering efforts, and forensic evidence. The detectors provide the means to verify the existence and identity of the tag. The data acquisition system assigns the tag, controls the application systems, interprets detection data, and tracks the location of tagged items.

Figure 4-1. Functional features of tagging system.

4.3 Description of System Components

The tagging system is composed of several hardware and software subsystems which must be integrated to complete the development of the tagging system. A schematic of these subsystems is shown in Figure 4.2. The tagging system consists of four major subsystems: 1) the fluorescence compounds, radioisotopes, and solvents that will be the constituents of the tag, 2) the method of applying the tag to the SNM, 3) the method of detecting the tag, and 4) the database system for tracking the use of the tag. The design criteria and selection of candidate fluorescence compounds and radioisotopes are described in Section 6 and Section 7, respectively. The design criteria for selection of candidate methods for applying the tag, detecting the tag, and tracking the tag are described briefly below and will be addressed further in later reports.

Figure 4-2. Tagging system components.

4.3.1 Tagging Materials

The tags will consist of a variety of materials which when applied in combination will provide a system for all of the intended applications. The materials include fluorescent compounds and radioisotopes that will be applied to SNM or container surfaces. The utility of these materials is derived from the phenomena of these tagging materials to adhere to the surface or become embedded into the microcracks normally present on surfaces. For all of the intended purposes, the fluorescence compounds will be either a dye or pigment in a quick drying solvent carrier or as a dry dusting powder. The purpose of the dye or pigment will be to form a readable pattern which can be used for SNM identification for inventory control. The purpose of the dusting powder will be to transfer a traceable tag to equipment or personnel handling the SNM. The radioisotope tag will be dissolved in a solvent carrier which carries the radioisotopes within the surface of the SNM. The purpose of the radioisotope tag is to provide a more permanent tag which is difficult and hazardous to remove and which can be detected through packaging containers.

The solvent matrix for each type of tagging material may vary. The solvent for the fluorescent identification tag will be similar to a lacquer or polymer coating which will cure when applied to the surface of the material. This coating will serve two functions: 1) provide the stability to form a readable pattern and 2) provide a solution matrix which allows the compounds to maintain their fluorescence sensitivity. The solvent for the fluorescent dusting powder may include air or volatile solvent propellents. The solvent for the

radioisotopes will perform like a penetrating solvent. Solvents for the removal of the tag are also included in this subsystem.

4.3.2 Application Method

The application method subsystem includes the hardware to remotely or manually apply the tagging materials and the software to control the remote application system. This subsystem also considers: 1) the safety issues that will be necessary to consider when manufacturing, transporting, and handling the tagging materials, application solvents, and the removal solvents; 2) methods to remove the tag when it is inappropriately applied or needs updated by the processing facility and methods to minimize tag removal when pilferers attack the tag; and 3) issues to be addressed when disposing of unused tag materials, tagged materials, and tag removal solvents and supplies.

4.3.3 Detection Method

The detection subsystem includes the hardware to remotely or manually detect the applied tags and the software to interpret the collected data. The hardware will address the following methods: 1) remote and portable detection methods that can be used to validate the application of the tag and verify the identification of unadulterated tags and 2) detection methods necessary to identify SNM and personnel that have tampered with the tag. This subsystem will also consider the safety issues that may arise when handling the SNM and detection equipment during tag detection efforts.

4.3.4 Data Control Subsystem

The database subsystem will address the issues associated with securing and controlling the unique identification features of the tags. This control will be necessary to ensure that: 1) facilities maintain the integrity of their unique tags between facilities; 2) a facility can maintain the integrity of unique tags within a facility; 3) facilities can track the transfer of materials between facilities; and 4) a facility can track the transfer of material within the facility.

5. DESIGN CRITERIA FOR THE TAGGING SYSTEM

Using the description of the conceptualized tagging system and the intended applications, a series of design criteria can be composed to establish specifications for the eventual development of the tagging system. The design criteria address the expectations of the system and can be used to establish system level specifications for development, testing and verification. This section provides a description of the criteria that apply to the design of the overall tagging system. The design criteria and selection status for the fluorescence compound and radioisotopic components of the tagging system are provided in Section 6 and Section 7, respectively. Descriptions of design criteria for the solvent matrix, application method, detection method, and database control will be provided in future documents.

5.1 Material to be Tagged

There are wide variety of candidate materials that should be tagged. However, the types of material to be tagged will be dependent upon the risks associated with the loss of the material. A listing of materials from highest to lowest priority includes the following materials:

- 1) weapons grade plutonium metal, enriched uranium metal, partially dismantled warhead components containing SNM (i.e., including their storage containers);
- 2) unirradiated nuclear fuel elements and irradiated nuclear fuel elements;
- 3) tritium containers, unirradiated nuclear fuel elements, containers of plutonium salts, containers of enriched uranium salts, and cylinders of enriched uranyl gas;
- 4) depleted and natural uranium, beryllium metal, heavy water containers, enriched ^6Li , dismantled warhead components not containing SNM, and containers of specialty radioisotopes (e.g., ^{60}Co , ^{90}Sr , and ^{137}Cs).

To balance cost and handling and risk, the tag will be designed to tag components and containers of at least 500 g of plutonium metal; enriched uranium metal; plutonium or enriched uranium salts (solid oxides, chlorides, and nitrates); warhead components containing SNM; unirradiated nuclear fuel rods; and containers of plutonium, enriched uranium, and tritium liquids and gases. Even though tagging of irradiated fuel elements, especially those under IAEA control, is considered important, the cost of application and detection of the tags is considered prohibitive. The storage containers are expected to be constructed of aluminum, carbon steel, or stainless steel.

5.2 Facilities Applying the Tag

Because the purpose of using the tag is to deter the pilfering of SNM, facilities missioned with producing refined process quantities of SNM, assembly/disassembly of weapons, storage of SNM stockpiles, or preparation of SNM for storage will be considered for use of the tag. It is not expected that the different processing locations or building within a facility (localized complex) will apply different tags. Because the tag's primary objective is to relate recovered SNM to the

facility in which it was taken, the tag must provide sufficient uniqueness to be traced to the last facility responsible for the material. The number of facilities needing the tag is projected to be at least one hundred based upon the number of proliferant countries and the number of facilities in the United States and Former Soviet Union block countries. Therefore, to ensure that each participating facility has a unique tag, the tag will be designed to accommodate at least one hundred unique facilities.

5.3 Process Location for Applying the Tag

To maximize the deterrence impact of the tag, the tag should be applied directly to the SNM just prior to or during its initial exposure to manual handling. Therefore, the tag will be designed so that the tag can be applied upon receipt of the material at the facility, reapplied whenever the processing of the material significantly alters the detection of the previous tag (e.g., after machining, dissolution, recasting, or washing), prior to placing the SNM or container in long term storage, and before transferring the container to another facility.

5.4 Process Location for Detecting the Tag

To minimize the transfer and handling of the tagged materials, the tag should be detectable in the area where the SNM is received, immediately after the tag is applied, in SNM transfer areas, and in SNM storage areas. Because the tagged component will be hazardous and securely controlled, access to the material should be avoided whenever possible. Therefore the tag will be designed to have components that will be:

- 1) Detectable in the presence of storage containers and packaging, which may include, but is not limited to 0.6 cm (1/4") stainless steel, 2.5 cm paper, or 1 cm plastic; and
- 2) Detectable with technology consisting of commercially available instrumentation that is hand-held and adaptable for remote handling.

5.5 Tag Removal

The purpose of the tag is to deter pilfering of material as individuals will be less inclined to steal material that can be traced back to themselves. However because the presence of the tag will be publicized, personnel pilfering the tagged material may take actions to eliminate the trail of evidence which traces the SNM back to the pilferer and facility. Therefore, the tag will be designed to be difficult to remove all traceable evidence of the tag. The traceable evidence may include a residue of the tag on the tagged material and residue of the tag on the pilferer and removal equipment. Difficult to remove is defined as: 1) leaving a residue on the tagged material after the tag has been subjected to cleaning with water, soap, or readily available volatile organic solvents or petroleum products and 2) needing sophisticated equipment and facilities to prevent hazardous contamination after machining or etching the tagged material.

5.6 Retention of Components Functional Properties

The safe handling, performance, stability, and security of SNM components are based on rigorous standards for composition and configuration. Therefore the tag will be designed such that it does not alter the physical and functional properties of the SNM. These physical and functional properties include but are not limited to: 1) the shape; 2) the mass; 3) the SNM radioactive spectrum; 4) the criticality function ($K_{\text{effective}}$); and 5) the material strength.

5.7 Release of Information Related to the Composition and Shape of the Tagged Material

For some of the components to be tagged, the composition and the shape of the SNM will be controlled as classified information. To protect this information, the tag will be designed so that detection of the tag does not reveal additional information about the shape and composition of the material being tagged. If necessary, features can be incorporated into the detection or data acquisition subsystems to prevent the release of the SNM's gamma spectra.

5.8 Implementation Costs

The tag needs to be inexpensive to implement, because there will be a significant effort to get all facilities handling SNM to use the tag and because there will be a large number of items to be tagged. Therefore, the tag will be designed so that each tag will cost less than \$500, each tag application device will cost less than \$10K, and each tag detection system will cost less than \$100K.

5.9 Method of Application

Because there will be a significant effort to get all SNM that may be pilfered tagged, the equipment and tasks necessary to apply the tag should not prohibit its use. Therefore, the tag will be designed so that it can be quickly applied by hand or automatically during the normal processing activities.

5.10 Lifetime of the Tag

Because the material being tagged is hazardous to handle and may be placed in storage for extended periods of time, the tag should have a detectable lifetime that covers the intended storage time. Therefore, the tag will be designed with a signature that is detectable for 20 years.

5.11 Regulatory Compliance

The components of the tag and the materials tagged will be handled, shipped, stored, and disposed of in facilities complying with International, Federal, and State regulations. Therefore, the tag will be designed so that it can be used in compliance with federal regulations. This would include NRC and DOE requirements for radioactive materials; DOT requirements for shipping; EPA requirements for waste disposal and material handling; and DOE, OSHA, and FDA requirements for personnel safety.

6. SELECTION OF CANDIDATE FLUORESCENCE COMPOUNDS

The fluorescence tag is based on the concept that an invisible dye or pigment can be applied to a surface and then detected by the characteristic fluorescence emission of the dye. The uniqueness of the tag can be determined by assigning a specific dye or combination of dyes to each user facility. The tag could also be used to individually identify each container if it is applied in a unique pattern such as a bar code or serial number. Advantages to this type of tag are that it can be detected and read with simple detector technology, a large variety of dyes and pigments are commercially available at low cost, and very small quantities of the material can be detected. Because small quantities can be detected, efforts to remove the tag have a significant probability of leaving forensic detectable residues behind on the tagged material, the individual removing the tag, and the equipment used to remove the tag. This type of tag has the following disadvantages: 1) the tag cannot be detected if the tag is enclosed in a secondary container or wrap; 2) organic fluorescent compounds make the tag susceptible to deterioration or evaporation from the radiation emitted by the tagged SNM; and 3) organic fluorescent compounds are soluble (i.e., can be removed) in readily available solvents such as acetone, alcohols, or petroleum products.

There are two types of fluorescence tags addressed in the overall tagging system. These types of tags include the inventory control tag and the tampering tag. A description of the design criteria for each type of tag is addressed in Section 6.1 and the process of selecting candidate fluorescence compounds is provided in Section 6.2

6.1 Design Criteria for Fluorescence Tag

6.1.1 Safety

Because workers with the material will have the potential to be marked inadvertently with the tagging material, the amount of fluorescence compound in each tag will be limited to ensure worker safety. The tag will be designed so that amount of fluorescence material in each inventory tag will be less than the established short-term allowable exposure limit.

6.1.2 Visibility

To ensure that a pilferer will have difficulty removing all traceable portions of the tag, the location of the fluorescent tag should be difficult to identify. To increase the level of difficulty in identifying the location of the tag, the fluorescent compounds for both the inventory and tampering tag will be designed to be colorless in the visible spectrum yet have detectable fluorescence in the ultraviolet, visible, or near infrared. Each tag will contain a combination of compounds and require that at least two independent excitation sources are necessary to detect the entire fluorescence signature.

6.1.3 Solubility

To be easy to apply in a well-resolved pattern, the fluorescent compounds for the inventory tag will be designed to be soluble in the rapidly drying solvent matrix that adheres to the material to be tagged. These fluorescent compounds also will be designed to be difficult to remove from metal, ceramics, cloth, and skin using soap and water.

6.1.4 Sensitivity

6.1.4.1 Inventory Tag. To ensure that the pattern of the inventory fluorescence tag can be identified with minimal exposure, the fluorescence of an intact tag will be manually readable from a reasonable viewing distance with an excitation source held above the surface of the material. To ensure, traceability to the facility of origin, the amount of fluorescent material present in the original application of the tag will be sufficient so that 0.01% of the material originally present in the tag can be identified using optimized laboratory analysis techniques.

6.1.4.2 Tampering Tag. To ensure that personnel improperly contacting material dusted with the tampering tag can be detected, the presence of a small amount of the tampering fluorescence material will be manually detectable from a reasonable viewing distance with an excitation source held above the surface of the material. To ensure forensic traceability of personnel to any recovered SNM, one microgram of the tampering tag fluorescent material will be sufficient for identification by an optimized laboratory analysis technique.

6.1.5 Uniqueness to Facility

To uniquely identify a facility of origin, the inventory tag will be designed to provide sufficient combinations to identify at least one hundred facilities.

6.1.6 Uniqueness of Tag

To identify an individual that has improperly handled tagged SNM, that tag will contain a fluorescence material that can be transferred readily to the skin or handling equipment upon touching the tag. Because the number of fluorescence materials meeting the requirements for the tampering tag is limited, it is expected that the material for the tampering tag will be common to all facilities.

6.1.7 Traceable Across Facilities

Because it is expected that material will be transferred between several facilities using the tag, the tag will be designed to encode the chronological history of the movement of the SNM between facilities. This chronological tracing will be accomplished using the pattern of the fluorescence tag in combination with the electronic tracking of the data control system hardware.

6.1.8 Size and Pattern

To uniquely identify each SNM component or container, the inventory fluorescent tag will be designed to have a unique pattern and be of sufficient size to provide the necessary resolution so that the fluorescence response can be read from a video screen. The maximum size for the tag is expected to be 2 cm x 2 cm and consist of at least five alphanumeric or shape characters (yields more than 10^7 combinations).

6.1.9 Composition of Tag

The fluorescence compounds as received from the manufacturer will be soluble in the application solvent, concentrated enough to meet sensitivity requirements, not contain detectable quantities (when part of a tag) of impurities that may impact fluorescence compounds used in other facilities' inventory tags.

6.1.10 Viewability

Because the fluorescence tags are present to provide uniqueness for inventory control and tamper resistance, the fluorescence compounds are not expected to be detectable through additional packaging.

6.1.11 Detection Instrumentation

To minimize the cost of the tagging system, the tag will be designed to be detectable with existing automated instrumentation at locations including but not limited to: 1) immediately after the tag is applied, 2) shipping and receiving areas, and 3) remote storage locations. Based on the proposed tagging concept the tag will be designed to have sufficient fluorescence and adequate spectral resolution so that an unadulterated tag can be detectable by its surface reflectance with a fluorescence probe constructed from readily available commercial instrumentation (i.e., includes source, monochromators, optical probes, and detectors). It is expected that absolute verification of the facility of origin for material that has been recovered may require a higher resolution fluorescence detection system.

6.1.12 Lifetime

It is expected that the inventory fluorescence tag will be simple and inexpensive to apply and therefore can be replaced on a routine basis. However because of the security control and hazards associated with handling the stored materials, the inventory tag will be designed to have a readable life of at least one year.

6.1.13 Disposition of Tag

It is not the intent of the tag that it will be removed. However, provisions need to be available in situations when there are inappropriate applications of the tag or when it is necessary to replace a tag. Therefore, the tag will be designed so that the removal process does not generate any waste that cannot be disposed of through existing procedures. Also, the process of removing the tag will be designed so that there are no hazards in addition to those inherent to handling.

6.2 Candidate Fluorescence Compounds

6.2.1 Material Suppliers

Commercial suppliers of liquid and powder fluorescent compounds were contacted for products which might meet the projected needs for the tags. Based on this search, several dyes were purchased or received as samples. These materials represent a significant portion of the variety of materials that are available or can be modified to meet the majority of the specifications for the inventory and tampering tags. A list of the fluorescence compounds currently being evaluated is provided in Table 6.1.

Table 6-1. Listing of candidate fluorescence compounds.

Product	Manufacturer	Form	Color	Commercial Use
UV Blue D-282	Day-Glo Color Corp	powder	yellow	optical brighter
Columbia Blue D-298	Day-Glo Color Corp	powder	light violet	organic colorant
DUBL-CHEK W2-D3F	Sherwin Inc	organic solution	transparent, colorless	penetrant developer
DUBL-CHEK HM-704	Sherwin Inc	organic solution	transparent, light green	penetrant
IR-125	Exciton Inc	powder	brown-green	laser dye
1-hydroxypyrene	Molecular Probe, Inc	powder	light brown	biological stain
SpotCheck ZL-27A	Magnaflux Inc	aerosol	yellow	penetrant
I-2120	Shannon Luminous Materials, Inc	lacquer	transparent, colorless	marking ink
Bonn-Trace No. 191	Bonneau Dye Corp	organic solution	transparent, colorless	marker dye
PMI-B-WB	Spectronics Corp	organic solution	transparent amber	marking ink

6.2.2 Visibility

The color of the fluorescence compounds is an important criterion in evaluating the candidate fluorescence compounds because the presence of a visible color will readily indicate the location of the tag and may provide an adequate visual cue to monitor the removal of the tag. Even though all of the compounds in Table 1 are detectable in the powder form, their visibility is dictated by their absorbance in the visible light spectral range (400 nm to 700 nm). Based on this criterion, a small amount of each of the compounds was dissolved in isopropanol and scanned with an ultraviolet/visible (UV/VIS) spectrophotometer. The spectra of the compounds were evaluated for visible absorbance peaks. From these spectral scans, only HM-704 (440 nm) (Figure 6.1) and ZL-27A (450 nm) had significant peaks in the visible spectral region. The other compounds, had multiple peaks in the ultraviolet below 400 nm (e.g., Figure 6.2 for compound D-282). IR-125, a NIR fluorescence compound, (Figure 6.3) had it most significant absorbance peak at 790 nm. A description of the conditions for obtaining the spectra and the absorbance spectra for the candidate compounds are provided in Appendix A.

Figure 6-1. Absorbance spectrum for HM-704 in isopropanol.

Figure 6-2. Absorbance spectrum for D-282 in isopropanol.

Figure 6-3. Absorbance spectrum of I-125 in isopropanol.

Of additional importance from the absorbance spectra are the wavelengths of maximum absorbance. These peak absorbance wavelengths are used to excite the fluorescence of the compounds. When there is a strong absorbance peak in the UV and no peak in the visible spectrum for a candidate compound, the compound meets the criteria of being invisible under normal lighting but visible under a UV source. Similarly for IR-125, the compound has no absorbance peaks in the visible spectrum but a significant peak in the NIR. These spectral characteristics indicate that the compound is invisible under normal lighting but will detectable with a simple detector under a NIR source.

6.2.3 Fluorescence in Solution

After the visual inspection of the sample materials, a fluorescence evaluation was completed by viewing the isopropanol solutions of the fluorescence materials under UV radiation. This was done by holding a 4-watt UV lamp over the isopropanol solutions and recording the visible color of the solutions for both 254-nm and 365-nm UV lamps. Under the UV lamp, the compounds demonstrated sufficient sensitivity to be seen easily with the unaided eye at concentrations of less than 0.01% with the lamp held at a distance greater than 20 cm. The color of the visible fluorescence from each compound is summarized in Table 6.2. Of the candidate compounds, the IR-125 was the only dissolved compound that did not produce visible fluorescence under the UV lamps. This lack of response was expected, because IR-125 does not have a significant absorbance peak in the UV.

Table 6-2. Fluorescence color of the fluorescence compounds.

Compound	Solution Color Under 254 nm	Solution Color Under 365 nm	Surface Color Under 254 nm	Surface Color Under 365 nm
UV Blue D-282	violet	violet	green	green
Columbia Blue D-298	blue	blue	white	white
W2-D3F	violet	blue	blue	blue
HM-704	white	white	yellow-green	yellow-green
IR-125	no fluorescence	no fluorescence	no fluorescence	no fluorescence
1-hydroxypyrene	no fluorescence	purple-red	no fluorescence	no fluorescence
ZL-27A	yellow-green	yellow-green	yellow-green	yellow-green
I-2120	violet	blue	blue	blue
Bonn No. 191	violet	blue	gray	gray
PMI-B-WB	violet	blue-violet	gray	gray

Solution colors are for <0.1% of compound in isopropanol (D-282, PMI-B-WB, and Bonn No. 191 required a small amount of water to dissolve in the isopropanol).

Surface colors are for an aliquot of the commercial product dried on a stainless steel surface. The compounds received as a powder were applied to the surface as isopropanol solutions.

The isopropanol solutions used in the absorbance evaluations were scanned using a spectrofluorometer to obtain fluorescence spectra of the candidate compounds. The scans were performed using a 365-nm excitation source for all compounds except IR-125. The IR-125 solution fluorescence spectrum was obtained using a 750-nm excitation source. A description of the conditions for obtaining the fluorescence spectra and the fluorescence spectra for the candidate compounds are provided in Appendix B. Conclusions for the selection of the candidate compounds from the absorbance and fluorescence spectra follow:

6.2.3.1 D-282. Day-Glo's UV Blue D-282 compound has some absorbance structure with UV peaks at 220 nm, 240 nm, 280 nm, and 350 nm (Figure 6.2). This structure is slightly apparent in the fluorescence spectrum (Figure 6.4) but is not well resolved under the 365-nm excitation source with 10-nm bandwidth. However, there are indications of peaks at 420 nm, 440 nm, and 470 nm. The combination of UV absorbance peaks, no visible absorbance peaks, and the visible fluorescence peaks provides indications that this compound should be considered as a candidate compound for the fluorescence tagging compound. The fluorescence spectrum for D-282 is similar in shape to compounds I-2120, Bonn-Trace 191, and PMI-B-WB.

Figure 6-4. Fluorescence spectrum of D-282 under 365 nm excitation.

6.2.3.2 D-298. Day-Glo's Columbia Blue D-298 compound has some absorbance structure with UV peaks at 210 nm, 245 nm, 375 nm. This structure is not apparent in the fluorescence spectrum with the one large smooth peak at 445 nm. The combination of UV absorbance peaks, no visible absorbance peaks, and the visible fluorescence peaks provides indications that this compound should be considered as a candidate compound for the fluorescence tagging compound. The fluorescence spectrum for D-298 is similar in shape to compounds W2-D3F and HM-704.

6.2.3.3 W2-D3F and HM-704. DUBL-CHEK W2-D3F and HM-704 appear to have the same absorbance and fluorescence spectra except for the visible absorbance peak at 450 nm for HM-704 (Figure 6.1). This visible peak provides a significant difference in the observed color of the fluorescence between the two candidate compounds. With the addition of the visible yellow peak of the HM-704, the fluorescence from the HM-704 appears white rather than the violet seen with W2-D3F (Table 6.2). The combination of UV absorbance peaks, no visible absorbance peaks, and the visible fluorescence peaks provides indications that W2-D3F should be considered as a candidate compound for the fluorescence tagging compound. However, the visible yellow color of HM-704 should eliminate this compound as a candidate for the fluorescence tag.

6.2.3.4 IR-125. Exciton's IR-125 has a small absorbance peak in the UV at 220 nm and absorbance peaks at 720 nm and 795 nm. The absorbance peaks in the NIR were used to obtain a NIR fluorescence spectrum using an excitation wavelength of 755 nm (Figure 6.5). IR-125 was included in the list of candidate fluorescence compounds because its characteristic NIR fluorescence may provide a tagging compound that yields a fluorescence response in a spectral region not readily measured by a pilferer. From the spectral observations of IR-125's UV absorbance peak, no visible absorbance peaks, and the NIR fluorescence peak, this compound should be considered as a candidate compound for the fluorescence tag.

Figure 6-5. Fluorescence spectrum of IR-125 under 755 nm excitation.

6.2.3.5 1-Hydroxypyrene. Molecular Probes' 1-hydroxypyrene compound has a complex absorbance structure with multiple peaks below 400 nm. This complicated structure is repeated in its fluorescence spectrum with peaks at 390 nm, 410 nm, and 440 nm. The combination of UV absorbance peaks, no visible absorbance peaks, and the visible fluorescence peaks provides indications that this compound should be considered a candidate compound for the fluorescence tagging compound. In addition, the complexity of the fluorescence spectrum makes 1-hydroxypyrene an excellent candidate to add to other candidate compounds to provide a unique fluorescence spectral signature for a tag.

6.2.3.6 ZL-27A. Magnaflux's AL-27A has both UV and visible absorbance peaks and a large fluorescence peak at 525 nm. Even though this compound's bright yellow-green fluorescence would make a unique spectral contribution when in combination with the other candidate compounds, the visible yellow color of SL-27A should eliminate this compound as a candidate for the fluorescence tag.

6.2.3.7 I-2120, Bonn-Trace 191, and PMI-B-WB. Shannon's I-2120, Bonneau's No. 191, and Spectroline's PMI-B-WB all have similar UV absorbance spectra and visible fluorescence spectra and similar fluorescence spectra to D-282. These similarities may indicate that they are the same compound or slight modifications to the same organic-base structure. The combination of UV absorbance peaks, no visible absorbance peaks, and the visible fluorescence peaks provides indications that these compounds should be considered as candidate compounds for the fluorescence tagging compound.

6.2.4 Fluorescence from a Metal Surface

After observing the fluorescence of the isopropanol solutions, the fluorescent compounds dissolved in a commercial matrix were brushed directly onto a 316-stainless steel surface. Compounds received as a powder were dissolved in isopropanol and were aliquoted onto the stainless steel surface. After the solutions had dried, the surfaces were exposed to the UV lamps. For most of the compounds, the fluorescence from the greater than 4 cm² spots was visible when the lamp was held from at least 20 cm from the metal surface. Of

the compounds deposited, only IR-125 and 1-hydroxypyrene did not produce a visible fluorescence response. The lack of fluorescence for 1-hydroxypyrene was due to the absence of an organic matrix to support the fluorescence of the solvated compound (i.e., the dry compound is not fluorescent and needs an organic substrate to produce visible fluorescence). The D-282 and D-298, even though present as dry powders after the isopropanol evaporated, continued to be naturally fluorescent in the powder state. The fluorescent colors of the compounds deposited on the steel surface are listed in Table 6.2.

6.2.5 Fluorescence in Coating

Some of the fluorescence materials were received in a matrix which leaves a shellac or polymeric residue. This matrix serves three functions: 1) it provides a solvent to carry the fluorescence compound into the application surface, 2) it provides a coating substrate which binds the fluorescence compound to the application surface, and 3) it provides a matrix to support the fluorescence of the compounds. Generally, this matrix leaves a colorless transparent coating with an apparent darkened surface caused by a change in the reflectivity of the application surface.

This impact of the matrix on the fluorescence is seen in several ways. Without the matrix, 1-hydroxypyrene was no longer visibly fluorescent. Other compounds exhibited changes in the visible color and intensity of the fluorescence. The most significant observation about the fluorescence from the coated surfaces is that the compounds with a supplied matrix emitted a more sensitive fluorescence response. The reason for this behavior is that most fluorescence organic compounds are dependent upon a solvent to distribute/absorb excess energy to allow fluorescent transitions to occur.

Based on these observations, the organic fluorescent compounds that will be used as an inventory tag will need a solution matrix that will remain with the compounds throughout the life of the fluorescent tag. For the tampering tag, the fluorescent compounds chosen will have to be fluorescent in the powder form. A complete evaluation of the choice and properties of the solvent matrix will be provided in a later report.

6.2.6 Continuing Efforts for the Candidate Fluorescence Compounds

The current observations in the selection of candidate fluorescence compounds require the continued evaluation of the materials as tagging candidates. These additional efforts will focus on providing conclusions for the fluorescence inventory tag by determining if: 1) the candidate compounds can be resolved from each other when present in combinations; 2) the compounds will retain their fluorescence properties when combined with the selected application solvent; 3) the compounds can survive for the intended lifetime of the tag in the presence of the background radiation from the tagged SNM; and 4) the compounds will penetrate into and respond from the microcracks of metal surfaces. Additional efforts that will be completed to provide conclusions for the fluorescence tampering tag will focus on determining whether: 1) the compounds can be applied in a solvent that generates a readily transferrable powder upon contact; 2) the compounds provide sufficient sensitivity without a solvent to be detectable under a screening detector; and 3) the compounds bind to equipment and personnel handling the materials.

6.2.7 Issues to be Resolved for the Candidate Fluorescence Compounds

Because of the variety of applications for the tag and the limits imposed by the requirements, several issues will need to be addressed, before full implementation of the fluorescent tags can be accomplished. These issues include and will be addressed as:

6.2.7.1 Impact on SNM Handling. Will the application of the compounds to the surface of the SNM or dusted throughout the handling facilities impact the safety and performance properties of the SNM being tagged? This issue will be addressed by consulting with facilities currently missioned with warhead dismantlement and SNM storage. Recommendations on the applicability of using the fluorescent tagging materials will be based on the acceptability of this tagging application to these facilities.

6.2.7.2 Acceptability of Tagging Personnel. Will the transfer of tagging compounds to the skin of employees pose an acceptable risk? This issue will be addressed by presenting the application to industrial hygienists and submitting a human experimentation plan to appropriate Contractor/DOE review committees.

7. SELECTION OF CANDIDATE RADIOISOTOPE COMPOUNDS

The radioisotope tag is based on the concept that unique isotopes can be applied to a surface and then detected by their characteristic gamma decay. The uniqueness of the tag will be established by assigning unique combinations of the radioisotopes to each user facility. Advantages of this tag are that it can be detected and read with simple detector technology, the tag will be detectable for a minimum of twenty years, very small quantities of the tag can be detected, and materials inside storage containers and packaging can be tagged and identified without opening the container. As with the fluorescent tag, efforts to remove the tag will leave significant forensic detectable residues. This radioisotopic will have several disadvantages: 1) the special precautions and handling procedures required when using the radioisotopes; 2) the cost of the limited number of available radioisotopes; and 3) the potential detection difficulty of the radioisotopes in the presence of the background radiation from the SNM. Design criteria for the radioisotopic tag is addressed in Section 7.1. The selection process for candidate radioisotopes is provided in Section 7.2.

7.1 Design Criteria for Radioisotopic Tag

7.1.1 Safety

The tag will be designed to limit the radioactive dose from the applied tag to add less than a few percent to the existing background of the tagged SNM and will not cause an unacceptable exposure during normal handling operations. Any radioisotope that may be taken into the body of an individual handling the tag will provide a less-than-lethal dose, have well-defined biological uptake processes, and be treatable with established methods.

7.1.2 Uniqueness to Facility

To provide capabilities to identify each facility handling SNM, the number of useful tagging radioisotopes will be sufficient to identify uniquely at least one hundred facilities.

7.1.3 Uniqueness of Tampering Tag

To identify an individual that has improperly handled tagged SNM, that tag will be designed to contain a radioisotope that can be taken up by a person, has a long residence time, and normally is not found at the targeted handling facilities. Because the number of radioisotopes with these characteristics is limited, it is expected that the tampering tag will be common to all facilities.

7.1.4 Traceable Across Facilities

Because it is expected that material will be transferred between facilities using the tag, the tag will be designed to contain a component or pattern that will allow the material to be tagged chronologically so that the history of the tag can be traced. This may be accomplished by monitoring the intensity ratio between isotopes as they decay, using short-lived isotopes that will survive long enough to cover the transport time, or by using encoding information into the pattern in which the isotopes are applied.

7.1.5 Lifetime

Because the SNM to be tagged may be sealed in long-term storage containers, the radioisotope tag will be designed to be detectable over at least twenty years. A short-lived isotopic tag for interfacility transfer will be designed to be detectable for at least 30 days.

7.1.6 Size and Pattern

To accommodate the expected size of materials to be tagged, the maximum size for the tag is expected to be 2 cm x 2 cm. To provide uniqueness between tags, the radioisotopes may be applied in resolvable patterns.

7.1.7 Composition of Tag

The radioisotope as received from the manufacturer will be in a chemical form that is soluble in the application solvent, sufficiently concentrated to meet sensitivity requirements, and sufficiently pure to not contain detectable quantities of other tagging radioisotopes.

7.1.8 Detection Instrumentation

To minimize the cost of the tagging system, the tag will be detectable with existing portable instrumentation at locations including but not limited to: 1) immediately after the tag is applied, 2) shipping and receiving areas, and 3) remote storage locations. Based on the proposed tagging concept the tag will be designed to have sufficient activity and adequate separation between emission lines so that the tag can be detectable with a conventional sodium iodide and/or germanium gamma detector. It is expected that the detector will consist of a standard 5-cm diameter crystal, the type found in commercially available hand held detection systems, and will be adequate for detecting unadulterated tags that have recently been applied. It is also expected that absolute verification of the facility of origin for material that has been recovered may require a higher resolution, low background germanium detection system.

7.1.9 Detection of Recently Applied Tag

To verify the existence of an unadulterated tag, the tag will be designed to be detected from a reasonable and safe distance through 0.6 cm of stainless steel, with an average count time of 15 minutes. The tag will be considered detectable if the count rate is statistically significant when compared to the background of the SNM being tagged.

7.1.10 Detection of Adulterated Tag

To verify the existence of a tag that has been through a 99.9% removal effort, the tag will be designed to provide at least one count per minute (CPM) above background with a high resolution, low background, a germanium detector exposed directly to the tag from as close as reasonably possible. The maximum count time for this type of detection is estimated to be 600 minutes.

7.1.11 Disposition of Tag

It is not the intent of the tag that it will be removed. However, provisions need to be available in case inappropriate applications of the tag occur. For this reason, the tag when embedded will not consist of any quantity of hazardous material beyond the expected radioactive hazards if physical methods of removal are used. The dissolution of the tag will be possible using a solvent that can be disposed of with available mixed waste treatment technology.

7.2 Candidate Radioisotopes

The tagging system will use three different sets of radioisotopes for three different tagging applications: 1) long-lived high gamma energy radioisotopes for the identification of the facility of origin, 2) short-lived, biologically active radioisotopes for tagging an individual involved in proliferation of SNM, and 3) short-lived high gamma energy radioisotopes for tracing the transfer of SNM from one facility to another facility. Each of these applications requires the tagging materials to meet specific criteria identified in Section 5. How the candidate radioisotopes were evaluated against these criteria is described throughout this section. The criteria evaluated in this section include the half-life, gamma-ray energy, gamma-ray branching ratio, and the number of gamma-rays. These criteria are described in Sections 7.2.1 through 7.2.8. The candidate radioisotopes are presented in Section 7.2.9.

7.2.1 Half-life

7.2.1.1 Tracing Facility of Origin. To have a tag that will identify the facility of origin for the SNM or its containers for at least 20 years, gamma emitting radioisotopes with a half-life of 5 years to 1500 years were identified for further evaluation. This selection criterion ensured that at least 5% of any isotope in the original tag will remain 20 years after being applied. This half-life selection range also ensures that none of the candidate radionuclides can be part of the natural decay chain of uranium or plutonium isotopes and that the tags will need only a minimal amount of mass to achieve the requisite activity.

7.2.2.2 Tracking Interfacility Movement. To have a tag that can be used to track the facility of origin while materials are transferred from one facility to another, gamma emitting radioisotopes with half-lives from two weeks to three months were identified. This range provides a wide choice of lifetimes to allow a choice of a specific half-life which is long enough to finish the processing and transferring tasks, yet short enough to not require tag removal when the transfer is complete. This range of half-lives ensures that at least 5% of the tag will remain for transfers completed within 10 to 50 weeks and 99.9% of the tag to decay away for transfer times ranging from 20 to 120 weeks. Because the short half-life will minimize exposure hazards to contaminated pilferers, this identified group of short-lived isotopes also was used to select eligible biologically active isotopes for the tampering tag.

7.2.2 Isotope Decay Mode

The candidate radioisotopes were chosen to avoid the natural decay chain for uranium and plutonium to minimize ambiguity between the background radiation from the SNM and the radioisotopes in the tag. In addition, the mode of decay of each candidate radioisotope and its daughter, if they exist, was limited to gamma-rays from short-lived daughters formed from parent beta- or alpha-decay. Gamma-rays were chosen

to provide sufficient energy to detect the tag through containers. The gamma emitters were chosen to avoid ambiguity with any background radiation that may occur during spontaneous fission from the SNM and to eliminate hazards from any $K_{\text{effective}}$ impacts due to neutron emission. If a candidate isotope had a coexisting spontaneous fission decay mode, the candidate isotope was selected only if the branching ratio for this mode was less than $10^{-3}\%$.

7.2.3 Gamma-ray Energy

To make the tagging system more tamper-proof, a part of the tag must be applied directly onto the SNM and be detectable through a typical storage container or shielding materials. Therefore, a calculation was completed to identify which gamma-ray emissions would have sufficient energy to be detectable through 1/4" (6 mm) stainless steel. The percent of transmission of different gamma-ray energies through 1 ft (30 cm) of air and 1/4" of stainless steel is shown in Table 7.1. This calculation indicates that the candidate radioisotopes should have gamma-ray energies of at least 200 keV to be detectable (>40% transmission) inside a 6 mm thick stainless steel container.

Table 7-1. Efficiency of gamma-ray transmission through materials.

Energy (keV)	% of Gamma-ray Transmission	
	1 foot of Air	1/4" Stainless Steel
120	99.3	22.6
250	99.4	52.3
350	99.5	59.8
500	99.5	65.8
780	99.6	71.3
960	99.7	73.7
1400	99.8	79.8

7.2.4 Branching Ratio

Because it will be difficult and expensive to embed a significant mass of the radioactive tag into the SNM, the efficient use of each radioisotope tagging component will be essential. For this reason, not only is the effective transmission of each gamma-ray important, the effective conversion of each decay of the radioactive isotope into a detectable gamma-ray (branching ratio) is also important. A higher branching ratio will result in a higher achievable count rate (i.e., less material needed to be detected or less time needed to detect). To achieve an acceptable count rate (the true measure of the detectability of the tag) for each isotope, there must be a balance between the mass of isotope, half-life, the energy of decay, branching ratio, and counting time.

7.2.5 Mass of Radioisotope in the Tag

The amount of radionuclide used in the tag is mainly determined by the solubility of each individual isotope. It is currently estimated that the amount of a radioisotope soluble in the application solvent will be 100 ng in the projected volume of tag applied (e.g., 25 μL of solution). This corresponds to 4 parts per million (ppm) of isotope in the solvent and maintains the concentration of any toxic heavy metal isotope below the usual 5 ppm characteristic hazardous waste concentration limit. Even though, non-hazardous metals could exceed 100 ng and not be categorized as hazardous waste, the 100 ng of material per tag was

used as an upper limit of material in a tag for estimating a count rate. In some cases, the mass of the isotope may have to exceed 100 ng to achieve the appropriate count rate.

Figure 7-1. CPM versus gamma-ray energy.

7.2.6 Count Rates for Candidate Radioisotopes

As stated above, a primary criterion for selecting candidate isotopes is the estimated count rate. The count rate can be estimated using several factors: 1) the energy efficiency of the isotope through the packaging container (Table 7.1), 2) the efficiency of a decay to generate a detectable gamma-ray (branching ratio), 3) the mass of isotope in the tag (100 ng), 4) the number of decays per unit time (half-life ranging from 5 to 1500 years), 5) the amount of time available to detect the isotope (count time limited to 15 minutes), and 6) the efficiency of the detector to detect a gamma-ray (based on detector type, spatial orientation, and distance). To obtain good statistics in detecting an interrogated gamma-ray peak, the detector must record at least 500 counts (<5% uncertainty in the total counts).

To select candidate isotopes for the tag, the above parameters were used to calculate a theoretical CPM of each emitting gamma-ray for 100 ng of each candidate radioisotope. The calculations for count rates were based on measurements made with an 11.7- μCi ^{152}Eu source placed 12 cm away from a 25%-efficient Ge detector (N-type Ge crystal with a 2" diameter and thickness). The efficiency curve was obtained by counting the ^{152}Eu source for 10 minutes. The resulting intensities were then converted to represent the expected intensities from a 1 μCi source counted for one minute (Figure 7.1). From this curve, the expected CPM was estimated for a 1- μCi source of a specific energy gamma-ray for each candidate isotope at 100% branching ratio and 12 cm distance. The calculated CPM value then was adjusted for the actual branching ratio of the gamma ray, the measurement distance (32 cm), and the mass of isotope (100 ng).

7.2.7 Count Rates Through a Storage Container for the Candidate Radioisotopes

The above measurements of the 11.7- μCi ^{152}Eu source were used to complete the CPM calculations for each of the candidate radioisotope. The effect of the 6 mm of stainless steel from a storage container was determined using the total photon attenuation cross-sections for iron^{5,6} plotted as a function of gamma-ray

energies (Figure 7.2). These data were then fitted using a two-exponential decay method to derive the formula for calculating the attenuation:

$$Y = Y_0 + A_1 \cdot \exp(-(X-X_0)/T_1) + A_2 \cdot \exp(-(X-X_0)/T_2)$$

Where Y = photon cross-section in barns

X = gamma-ray energy in keV

and the values for the other parameters were equal to:

$$X_0 = 136.4 \quad Y_0 = 3.984$$

$$A_1 = 8.714 \quad T_1 = 40.61$$

$$A_2 = 8.409 \quad T_2 = 488.0$$

Figure 7-2. Photon attenuation versus energy.

7.2.8 Interference of Gamma-ray Energies from Other Candidate Radioisotopes

To ensure no ambiguity between the gamma-ray signatures of tags from different facilities, a radioisotope tag must not contain radioisotopes that have gamma-ray energies that overlap with radioisotopes of another tag. However, an interference between gamma-rays of two different isotopes will not be significant if each radioisotope has at least one unique (non-overlapping) gamma-ray peak among all of the gamma-rays emitted from other radioisotopes in the tag. For the purposes of selecting candidate radioisotopes, a germanium detector with gamma-ray resolution of 2.0 keV at the 1332 keV gamma-ray of ^{60}Co was assumed.

However, since a 3"x3" NaI or CsI detector with about 7% resolution (662 keV ^{137}Cs) can only resolve two gamma-rays if they are separated by approximately 50 keV, other overlapping gamma-ray energies may eliminate candidate radioisotopes. Another potential source of interferences, to be evaluated later, will be the

gamma-ray energy background from the Compton events of radiation emitted from both the SNM and other tagging radioisotopes.

7.2.9 List of Candidate Radioisotopes.

After the elimination of radioisotopes not meeting all the criteria, lists for both long- and short-lived radionuclide were compiled. These candidate isotopes are summarized in Table 7.2 and 7.3, respectively. These tables list isotopes with acceptable half-lives, gamma-ray energies of ≥ 200 keV, and branching ratios $\geq 1\%$.^{7,8,9} Tables 7.2 and 7.3 also include an indication of the methods of production, the number of gamma rays (>200 keV) available for detection, the number of potentially interfering gamma rays from the other candidate isotopes (if all other candidate isotopes were present simultaneously), and the count rate for the most intense gamma-ray (with no conflicting gamma-ray) of the isotope. The parent isotope is listed as the long-lived candidate isotope when a gamma-ray is emitted by a short-lived daughter isotope. An example of this, is Cs-137 which decays 100% by beta and produces a short-lived Ba-137m daughter which produces the gamma-ray. The candidate isotopes meet all the criteria from Sections 7.2.1 through 7.2.8, except for Nb-94. Nb-94 has a half-life outside the 1500 year limit, but meets the specification for count rate because of its gamma-ray energy and branching ratio.

The detailed list of information about the candidate isotopes, including all gamma-ray energies, conflicting gamma rays, and estimated CPM for each gamma ray through air and a 1/4 inch stainless steel container are given in Appendices C and D. There are 18 long-lived radionuclides which can be used as tagging isotopes for identifying the facility of origin. There are 38 short-lived isotopes which can be used for monitoring SNM transferred between user facilities.

7.2.10 Biological Tampering Tag

There are several short-lived radioisotopes such as ^{95m}Tc , ^{203}Hg , ^{105m}Ag , ^{114m}In , and ^{85}Sr in Table 7.3 which can be used to trace and identify the individual tampering with the tag.¹⁰ Under normal working conditions, authorized workers handling the tags in the controlled environment would be protected from contamination with the tagging materials. However, unauthorized handling would expose the worker to these isotopes and consequently to eventual uptake of the isotopes by direct touching. This list contains isotopes which have long enough half-lives to insure traceability over time and yet short-enough half-lives to provide non-lethal doses to the individual. The methods for neutralizing the biological uptake from accidental exposure would be defined and available.

7.2.11 Continuing Efforts for the Candidate Radioisotopes

The current list of candidate isotopes for the tagging systems will require continued evaluation to complete the selection of the isotopes that can be used in the final tagging system. Efforts to complete this selection are: 1) an evaluation of the cost of production of the candidate isotopes; 2) an evaluation of the gamma-ray energies versus the background radiation from the SNM that will be tagged; 3) a review of the exposure to authorized personnel handling the radioisotopes; 4) a review of the biological uptake properties for the candidate isotopes; and 5) an evaluation of the actual expected count rate with respect to the detection system chosen for the detection system.

Table 7-2. Long-lived candidate radioisotopes.

Isotope	Half Life (y)	Production Reactions *	# gamma rays (>200keV)	# conflicting gamma- rays	Non-conflicting gamma with highest CPM Energy (keV) CPM (air)	
Ag-108m	418.0**	T, C	3	1	433.70	503
Ba-133	10.54	T, F	4	1	355.86	12828
Bi-207	31.54	C	3	none	569.67	3122
Bk-247	1380.0	C	1	none	265.0	31
Cf-249	351.0	T	3	1	387.95	206
Cf-251	898.0	T	1	none	285.0	2
Cm-243	29.1	T	3	1	228.19	618
Co-60	5.27	T, F	1	none	1173.23	39478
Cs-137	30.07	T, F, I	1	none	661.62	3861
Eu-150n	36.9	C, F	15	4	439.39	3522
Eu-152	13.54	T, F	11	3	344.30	3895
Eu-154	8.6	T, F, I, C	10	1	1274.80	3111
Ho-166m	1200.0	T, F	5	3	810.31	49
Hf-178n	31.0	T, C	13	2	213.44	6110
Nb-94	20300.0	T, I	2	1	702.50	10
Pm-146	5.53	C	4	3	747.40	7230
Po-209	102.0	C	2	none	260.50	4
Tb-158	180.0	F, C	5	2	944.20	225

* T: Thermal neutron activation, F: Fast neutron activation, C: Charge particle reaction, I: Fission product, P: Photon reaction

Conflicting gamma-ray is the only one present

Table 7-3. Short-lived candidate radioisotopes.

Isotope	Half Life (y)	Production Reactions *	# gamma rays (>200keV)	# conflicting gamma- rays	Non-conflicting gamma with highest CPM Energy	
					(keV)	CPM (air)
Ag-105	41.29	P, C	12	8	331.50	295
As-74	17.77	F, P	3	1	595.70	9195
Cd-115m	44.6	T, F, I	1	1	933.60#	37
Cm-241	32.8	C	2	1	474.50	2904
Co-56	77.27	C	8	2	846.75	3632
Co-58	70.82	C, F	2	1	810.75	3906
Cr-51	27.7	T, F	1	1	320.07#	2168
Eu-147	24.1	C	8	4	677.60	623
Fe-59	44.5	T, F	2	none	1099.22	2788
Hf-175	70.1	T, F	2	2	343.40#	2109
Hf-179n	25.05	T, F, C	8	2	453.70	3650
Hf-181	42.39	T, F	2	1	482.16	2623
Hg-203	46.61	T, F	1	1	279.17#	2948
In-114m	49.51	T, F	2	1	558.27	176
Ir-192	73.83	T, F	9	5	468.06	877
Nb-91m	60.86	C	1	none	1205.00#	75
Nb-95	34.98	I, T, F	1	1	765.82#	5027
Pm-148m	41.29	F, I	13	7	550.20	3174
Rb-83	86.2	P, F	3	1	520.35	2758
Rb-84	32.77	F, P	2	1	881.50	3941
Rb-86	18.63	T, F	1	1	1076.63#	729
Re-183	70.0	C, F	3	1	291.72	86
Re-184	38.0	F	5	1	792.07	896
Rh-99	16.1	C	8	5	352.6	5678
Ru-103	39.2	T, I, F	2	1	497.08	4871
Sb-124	60.2	T, F	11	5	1691.04	646
Sc-46	83.79	T, F	1	none	889.26	3346
Sr-85	64.84	T, F, C	1	none	513.99	4018
Tb-160	72.3	T, F	13	5	298.57	766
Tc-95m	61.0	C	7	4	582.15	1272
Te-121	16.7	T, F, C	3	none	543.08	8059
Te-129m	33.6	T, F, I	1	none	695.98	121
Tm-168	93.1	F, C	12	4	815.95	474
V-48	15.97	P, C	5	2	983.50	18446
Xe-127	36.34	T, F, C	2	1	374.96	1090
Yb-169	32.03	T, F	1	1	307.68#	653
Zr-88	83.4	C	1	1	392.85#	3464
Zr-95	64.02	I, T, F	2	1	756.72	1539

* T: Thermal neutron activation, F: Fast neutron activation, C: Charge particle reaction, I: Fission product, P: Photon reaction

Conflicting gamma-ray is the only one present

7.2.12 Issues to be Resolved for the Candidate Radioisotopes

Because of the variety of applications for the tag and the limits imposed by the requirements, several issues will need to be addressed, before full implementation of the radioisotopic tags can be accomplished. These issues include and will be addressed as:

7.2.12.1 Impact on SNM Handling. Will the application of the radioisotopes to the surface of the SNM impact the safety and performance properties of the SNM being tagged? This issue will be addressed by consulting with facilities currently missioned with warhead dismantlement and SNM storage. Recommendations on the applicability of using the radioisotope tagging materials will be based on the acceptability of this tagging application to these facilities.

7.2.12.2 Acceptability of Exposure to Authorized Personnel. Can the tagging isotopes be used with an acceptable risk to personnel handling SNM in an authorized manner? This issue will be addressed by presenting the application to radiological engineers for dose calculations with estimates of contamination spread through a normal work environment.

7.2.12.3 Acceptability of Tagging Personnel. Will the application of tagging isotopes to skin of pilfering personnel pose an acceptable risk? This issue will be addressed by presenting the application to radiological engineers for dose calculations with estimates of contamination exposure on the skin and through ingestion. Any further evaluation will require submitting a human experimentation plan to appropriate Contractor/DOE review committees.

7.2.12.4 Number of Facilities Needing Tags. Will the current list of candidate isotopes provide a sufficient number of unique tags to accommodate the number of user facilities? The long-lived isotope candidates allow for well over 100 unique tags if the tag consists of three radioisotopes. After cost of production and gamma-ray energy resolution factors are considered, this candidate list is expected to contain approximately 10 isotopes, allowing for 120 possible combinations of three isotopes. If more than 120 combinations are necessary, an evaluation of tags containing more or less than three isotopes will be completed or the use of a signature pattern will be evaluated.

7.1.12.5 Cost of Tagging Materials. Will the cost of producing the radioisotopes prohibit the use of the materials? This issue will be addressed by identifying vendors of radioisotopes to establish a projected cost for radioisotopes that are not normally commercially available. After obtaining these cost estimates, a review of production costs at the INEL will be completed to determine the feasibility of making test quantities of isotopes that are not available commercially. If acceptable cost projections cannot be obtained for particular isotopes, a candidate isotope may be eliminated from the consideration or a cooperative effort with international isotope production facilities may be arranged.

8. REFERENCES

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Appendix A

Absorbance Spectra for Candidate Fluorescence Compounds

The following absorbance spectra were obtained using a Hewlett Packard HP8452A diode array spectrophotometer. The candidate fluorescence compounds were dissolved in Fisher electronic grade -2 propanol (isopropanol) and scanned using a 10-mm quartz cuvette. The compounds were dissolved as received in the isopropanol at concentrations yielding absorbance of less than 1.5 absorbance units.

The absorbance spectrum for Day-Glo Color Corporation's UV Blue D-282:

The absorbance spectrum for Day-Glo Color Corporation's Columbia Blue D-298:

The absorbance spectrum for Sherwin Incorporated DUBL-CHEK W2-D3F:

The absorbance spectrum for Sherwin Incorporated DUBL-CHEK HM-704:

The absorbance spectrum for Exciton Incorporated IR-125:

The absorbance spectrum for Molecular Probe Incorporated 1-hydroxypyrene:

The absorbance spectrum for Magnflux Incorporated SpotCheck ZL-27A:

The absorbance spectrum for Shannon Luminous Materials Incorporated I-2120:

The absorbance spectrum for Bonneau Dye Corporation Bonn-Trace No. 191:

The absorbance spectrum for Spectronics Corporation PMI-B-WB:

Appendix B

Fluorescence Spectra for Candidate Fluorescence Compounds

The following fluorescence spectra were obtained using a Photon Technology Incorporated Alphascan spectrofluorometer modified with a fiber optic dip probe. The candidate fluorescence compounds were dissolved in Fisher electronic grade 2-propanol (isopropanol) and scanned using a 365 nm excitation wavelength. The fluorescence spectrum for compound IR-125 was obtained using an excitation wavelength of 750 nm to measure its NIR fluorescence. The intensity of the fluorescence peaks are in units of relative count rates for the detector.

The fluorescence spectrum for Day-Glo Color Corporation's UV Blue D-282:

The fluorescence spectrum for Day-Glo Color Corporation's Columbia Blue D-298:

The fluorescence spectrum for Sherwin Incorporated DUBL-CHEK W2-D3F:

The fluorescence spectrum for Sherwin Incorporated DUBL-CHEK HM-704:

The fluorescence spectrum for Exciton Incorporated IR-125:

The fluorescence spectrum for Molecular Probe Incorporated 1-hydroxypyrene:

The fluorescence spectrum for Magnflux Incorporated SpotCheck ZL-27A:

The fluorescence spectrum for Shannon Luminous Materials Incorporated I-2120:

The fluorescence spectrum for Bonneau Dye Corporation Bonn-Trace No. 191:

The fluorescence spectrum for Spectronics Corporation PMI-B-WB:

Appendix C

Long-Lived Candidate Radionuclides

The following provides a listing of the properties of the candidate radioisotopes. The information^{7,8,9} also includes an estimated gamma-ray (>200keV) count rate for 100 ng of radioisotopes through the air and 1/4" of stainless steel.

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays Energy(keV) Nuclide	Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
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Ag-108m	433.70	0.90		418.00	502.79	317.15
Ag-108m	614.37	0.90		418.00	393.10	267.66
Ag-108m	722.95	0.90	722.95 Eu-154	418.00	350.40	246.80
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Ba-133	276.29	0.07	277.65 Hf-178n 277.63 Cm-243	10.540	1732.0	980.34
Ba-133	302.71	0.19		10.540	4407.3	2557.9
Ba-133	355.86	0.62		10.540	12828.	7740.5
Ba-133	383.70	0.09		10.540	1765.6	1083.7
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Bi-207	569.67	0.98		31.540	3122.1	2091.4
Bi-207	1063.62	0.77		31.540	1577.9	1190.0
Bi-207	1770.22	0.07		31.540	100.08	79.487
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Bk-247	265.00	0.30		1380.0	31.440	17.571
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Cf-249	252.88	0.03		351.00	12.674	6.9734
Cf-249	333.44	0.16	333.96 Eu-150n	351.00	55.596	33.044
Cf-249	387.95	0.66		351.00	206.06	126.79
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Cf-251	285.00	0.01		898.04	1.5054	.85968
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Cm-243	209.76	0.03		29.100	178.78	90.505
Cm-243	228.19	0.11		29.100	617.64	326.25
Cm-243	277.63	0.14	276.29 Ba-133 277.35 Hf-178n	29.100	684.35	387.91
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Co-60	1173.23	1.00		5.2700	39478.	30182.
Co-60	1332.51	1.00		5.2700	36081.	28003.
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Cs-137	661.62	0.85		30.070	3860.9	2670.4

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays		Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
			Energy(keV)	Nuclide			
Eu-150n	333.96	0.93	333.44	Cf-249	36.900	5097.1	3030.5
Eu-150n	439.39	0.78			36.900	3521.5	2227.7
Eu-150n	505.51	0.05			36.900	204.44	133.41
Eu-150n	548.26	0.51			36.900	1969.1	1308.0
Eu-150n	712.19	0.01			36.900	32.092	22.536
Eu-150n	737.44	0.09	736.20	Pm-146	36.900	281.81	199.28
Eu-150n	748.04	0.05	747.40	Pm-146	36.900	154.99	109.91
Eu-150n	751.05	0.02			36.900	61.820	43.876
Eu-150n	869.23	0.02	867.39	Eu-152	36.900	55.754	40.679
Eu-150n	1049.02	0.05			36.900	122.04	91.854
Eu-150n	1170.56	0.01			36.900	22.589	17.265
Eu-150n	1197.08	0.01			36.900	22.234	17.042
Eu-150n	1246.94	0.02			36.900	43.204	33.282
Eu-150n	1343.74	0.03			36.900	61.471	47.751
Eu-150n	1485.45	0.02			36.900	38.178	29.943
Eu-152	244.67	0.08			13.540	1469.2	798.68
Eu-152	344.30	0.27			13.540	3894.9	2332.4
Eu-152	411.09	0.02			13.540	361.58	229.26
Eu-152	778.90	0.13	780.20	Tb-158	13.540	1053.2	752.86
Eu-152	867.39	0.04	869.23	Eu-150n	13.540	300.34	219.05
Eu-152	964.00	0.15	962.20	Tb-158	13.540	1045.3	484.63
Eu-152	1089.82	0.02			13.540	127.80	96.724
Eu-152	1112.07	0.14			13.540	881.89	669.37
Eu-152	1212.89	0.01			13.540	59.245	45.486
Eu-152	1299.19	0.02			13.540	112.87	87.361
Eu-152	1408.08	0.21			13.540	1119.6	873.84
Eu-154	248.04	0.07			8.6000	1978.4	1081.0
Eu-154	591.70	0.05			8.6000	764.46	516.32
Eu-154	692.50	0.02			8.6000	273.61	191.04
Eu-154	723.30	0.20	722.99	Ag-108m	8.6000	2653.3	1869.0
Eu-154	756.70	0.04			8.6000	513.99	365.34
Eu-154	873.20	0.11			8.6000	1277.4	932.80
Eu-154	996.30	0.11			8.6000	1163.7	868.94
Eu-154	1004.80	0.18			8.6000	1892.9	1415.3
Eu-154	1274.80	0.35			8.6000	3110.9	2402.6
Eu-154	1397.80	0.02			8.6000	151.57	119.58

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays		Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
			Energy(keV)	Nuclide			
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Ho-166m	280.46	0.30			1200.0	51.686	29.383
Ho-166m	410.94	0.11	411.09	Eu-152	1200.0	14.468	9.0170
Ho-166m	711.69	0.56	712.19	Eu-150n	1200.0	49.961	35.079
Ho-166m	752.27	0.13	751.05	Eu-150n	1200.0	11.152	7.9179
Ho-166m	810.31	0.60			1200.0	48.839	35.176
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Hf-178n	213.44	0.81			31.000	6110.2	3123.1
Hf-178n	216.67	0.67			31.000	5000.7	2576.3
Hf-178n	237.40	0.10			31.000	699.71	375.91
Hf-178n	238.66	0.02			31.000	139.42	75.062
Hf-178n	257.62	0.17			31.000	1122.7	621.68
Hf-178n	277.35	0.02	276.29	Ba-133	31.000	125.37	71.044
			277.63	Cm-243			
Hf-178n	296.80	0.10			31.000	597.55	345.03
Hf-178n	325.56	0.94			31.000	5261.6	3109.5
Hf-178n	426.37	0.96			31.000	4440.9	2790.6
Hf-178n	454.05	0.18	453.60	Pm-146	31.000	796.46	507.52
Hf-178n	495.01	0.76			31.000	3163.7	2054.9
Hf-178n	535.02	0.10			31.000	394.03	260.35
Hf-178n	574.21	0.94			31.000	3523.4	2364.3
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Nb-94	702.50	1.00			20300.	9.3994	6.5821
Nb-94	871.10	1.00	869.23	Eu-150n	20300.	8.0738	5.8931
			873.20	Eu-154			
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Pm-146	453.60	0.65	454.05	Hf-178n	5.5300	19670.	12532.
Pm-146	633.30	0.02			5.5300	478.08	327.63
Pm-146	736.20	0.21	737.44	Eu-150n	5.5300	4513.2	3190.5
Pm-146	747.40	0.34	748.04	Eu-150n	5.5300	7229.5	5126.1
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Po- 209	260.50	0.002			102.00	3.9010	2.1681
Po- 209	896.60	0.003			102.00	1.8411	1.3507
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Tb-158	780.20	0.09	778.90	Eu-152	180.00	52.704	37.685
Tb-158	944.20	0.44			180.00	225.16	166.66
Tb-158	962.20	0.20	964.00	Eu-152	180.00	100.99	74.983
Tb-158	1107.70	0.02			180.00	9.1423	6.9354
Tb-158	1187.20	0.02			180.00	8.7053	6.6656

Appendix D

Short-Lived Candidate Radionuclides

The following provides a listing of the properties of the candidate radioisotopes. The information^{7,8,9} also includes an estimated gamma-ray (>200keV) count rate for 100 ng of radioisotopes through the air and 1/4" of stainless steel.

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays Energy(keV)	Nuclide	Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
Ag-105	280.30	0.30	279.17	Hg-203	41.290	2375.8	1350.4
Ag-105	319.16	0.04	320.07	Cr-51	41.290	310.67	182.72
Ag-105	331.50	0.04			41.290	295.42	175.34
Ag-105	344.20	0.41	343.40	Hf-175	41.290	2808.2	1681.5
			345.95	Hf-181			
Ag-105	392.64	0.02	392.49	Tb-160	41.290	124.82	77.006
			392.85	Zr-88			
Ag-105	443.37	0.11	442.40	Rh-99	41.290	630.00	399.33
Ag-105	617.85	0.01	616.50	Tc-99m	41.290	54.360	37.057
			618.10	Rh-99			
Ag-105	644.55	0.10	645.71	Tm-168	41.290	439.66	302.42
			645.84	Sb-124			
Ag-105	650.72	0.02			41.290	104.81	72.238
Ag-105	673.21	0.01			41.290	51.161	35.514
Ag-105	807.46	0.01	806.6	Rh-99	41.290	44.992	32.383
Ag-105	1087.94	0.04			41.290	115.41	87.325
As-74	511.00	0.59	5 other 511		17.770	10077.	6591.4
As-74	595.70	0.60			17.770	9195.4	6219.6
As-74	634.80	0.15			17.770	2197.8	1506.9
Cd-115m	933.60	0.01	933.00	Eu-147	44.600	37.183	27.470
Cm-241	474.50	0.97			32.800	2904.2	1868.8
Cm-241	640.00	0.04	641.92	Re-184	32.800	84.820	58.257
Co-56	511.00	0.48	5 other 511		77.270	2491.4	1629.6
Co-56	846.75	1.00			77.270	3632.5	2637.8
Co-56	977.46	0.01			77.270	45.949	34.204
Co-56	1037.83	0.14	1039.30	Tc-95m	77.270	440.43	330.95
Co-56	1175.13	0.02			77.270	66.275	50.679
Co-56	1238.28	0.68			77.270	1888.3	1453.4
Co-56	1360.22	0.04			77.270	111.74	86.908
Co-56	1771.49	0.16			77.270	344.96	274.00

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays Energy(keV) Nuclide	Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
Co-58	511.00	0.30	5 other 511	70.820	1640.4	1073.0
Co-58	810.75	0.99		70.820	3906.5	2813.9
Cr-51	320.07	0.10	319.16 Ag-105	27.700	2168.5	1276.3
Eu-147	601.50	0.08	599.50 Pm-148m 602.71 Sb-124	24.100	457.63	310.18
Eu-147	677.60	0.12		24.100	623.23	433.20
Eu-147	798.80	0.07		24.100	300.52	215.86
Eu-147	857.00	0.03		24.100	123.18	89.643
Eu-147	933.00	0.04	933.6 Cd-115m	24.100	149.14	110.17
Eu-147	956.00	0.04		24.100	158.81	117.79
Eu-147	1077.00	0.07	1076.63 Rb-88	24.100	250.80	189.49
Eu-147	1256.00	0.01		24.100	33.578	25.889
Fe-59	1099.22	0.56		44.500	2788.0	2112.7
Fe-59	1291.56	0.43		44.500	1910.2	1477.5
Hf-175	343.40	0.87	344.20 Ag-105	70.100	2109.4	1262.4
Hf-175	432.80	0.02	432.60 Pm-148m	70.100	32.942	20.769
Hf-179n	217.00	0.09	215.65 Tb-160	25.050	816.59	421.03
Hf-179n	236.60	0.18		25.050	1553.6	833.52
Hf-179n	257.50	0.03		25.050	260.16	144.03
Hf-179n	268.90	0.11		25.050	867.34	486.94
Hf-179n	316.00	0.20	316.49 Ir-192	25.050	1407.0	825.47
Hf-179n	362.60	0.39		25.050	2489.5	1508.6
Hf-179n	409.80	0.21		25.050	1229.4	765.78
Hf-179n	453.70	0.67		25.050	3650.3	2325.6
Hf-181	345.95	0.14	344.20 Ag-105	42.390	539.90	323.67
Hf-181	482.16	0.86		42.390	2623.0	1693.8
Hg-203	279.17	0.81	280.30 Ag-105	46.610	2947.6	1673.4
In-114m	558.27	0.05		49.510	175.69	117.17
In-114m	725.21	0.05	724.18 Zr-95 725.7 Pm-148m	49.510	139.82	98.546

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays Energy(keV) Nuclide	Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
Ir-192	205.78	0.04	203.94 Tc-95m	73.830	105.46	52.801
Ir-192	295.95	0.30		73.830	699.24	403.44
Ir-192	308.44	0.32	307.68 Yb-169	73.830	724.38	422.43
Ir-192	316.49	0.87	316.00 Hf-179n	73.830	1933.9	1135.0
Ir-192	468.06	0.52		73.830	876.63	562.38
Ir-192	484.54	0.03		73.830	55.933	36.159
Ir-192	588.60	0.05		73.830	65.954	44.495
Ir-192	604.40	0.09	602.71 Sb-124	73.830	125.24	84.975
Ir-192	612.45	0.06	611.45 Pm-148m	73.830	80.857	55.017
Nb-91m	1205.00	0.03		60.860	75.200	57.689
Nb-95	765.82	0.99	765.3 Tb-160	34.980	5027.2	3581.7
Pm-148m	210.00	0.02	208.80 Re-183	41.290	130.91	66.317
Pm-148m	288.00	0.11		41.290	606.29	347.25
Pm-148m	311.50	0.04		41.290	198.15	115.84
Pm-148m	414.10	0.18		41.290	767.55	479.20
Pm-148m	432.60	0.07	432.80 Hf-175	41.290	272.88	172.02
Pm-148m	501.30	0.07		41.290	260.78	169.86
Pm-148m	550.20	0.91		41.290	3174.4	2110.3
Pm-148m	599.50	0.08	601.50 Eu-147	41.290	262.64	177.89
Pm-148m	611.30	0.06	610.33 Ru-103	41.290	194.29	132.15
			612.45 Ir-192			
Pm-148m	629.90	0.87	631.66 Tm-168	41.290	2758.1	1888.0
Pm-148m	725.70	0.32	725.21 In-114m	41.290	917.90	647.02
Pm-148m	915.20	0.19	914.90 Tm-168	41.290	462.59	340.58
Pm-148m	1013.80	0.20		41.290	452.97	339.14
Rb-83	520.35	0.46		86.200	1425.7	936.25
Rb-83	529.54	0.30	528.10 Rh-99	86.200	918.34	605.41
Rb-83	552.50	0.16		86.200	475.31	316.28
Rb-84	511.00	0.50	5 other 511	32.770	4079.6	2668.5
Rb-84	881.50	0.71		32.770	3940.6	2882.3
Rb-86	1076.63	0.09	1077.00 Eu-147	18.630	728.55	550.44
Re-183	208.80	0.03	210.00 Pm-148m	70.000	102.31	51.658
Re-183	246.07	0.01		70.000	41.140	22.413
Re-183	291.72	0.03		70.000	85.985	49.420

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays		Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
			Energy(keV)	Nuclide			
Re-184	252.85	0.03			38.000	158.44	87.170
Re-184	641.92	0.02	640.00	Cm-241	38.000	51.946	35.701
Re-184	792.07	0.38			38.000	895.52	642.20
Re-184	894.76	0.16			38.000	345.94	253.70
Re-184	903.28	0.38			38.000	816.11	599.51
Rh-99	322.20	0.06			16.100	1171.2	690.44
Rh-99	352.60	0.31			16.100	5677.9	3418.8
Rh-99	442.40	0.02	443.37	Ag-105	16.100	234.04	148.27
Rh-99	511.00	0.12	5 other 511		16.100	1690.9	1106.0
Rh-99	528.10	0.35	529.54	Rb-83	16.100	4818.5	3174.7
Rh-99	618.10	0.04	617.85	Ag-105	16.100	443.45	302.33
Rh-99	806.60	0.01	807.46	Ag-105	16.100	122.47	88.132
Rh-99	941.50	0.01			16.100	109.79	81.225
Ru-103	497.08	0.86			39.260	4870.7	3166.6
Ru-103	610.33	0.05	611.30	Pm-148m	39.260	259.64	176.54
Sb-124	602.71	0.98	601.5	Eu-147	60.200	2623.9	1779.3
			604.40	Ir-192			
Sb-124	645.84	0.07	644.55	Ag-105	60.200	183.59	126.34
			645.71	Tm-168			
Sb-124	709.34	0.01			60.200	33.409	23.441
Sb-124	713.82	0.02			60.200	54.642	38.389
Sb-124	722.78	0.12	724.18	Zr-95	60.200	282.59	199.03
Sb-124	968.22	0.02			60.200	36.391	27.048
Sb-124	1045.12	0.02			60.200	34.478	25.935
Sb-124	1325.53	0.02			60.200	23.011	17.849
Sb-124	1368.21	0.03			60.200	37.502	29.186
Sb-124	1436.60	0.01			60.200	15.942	12.466
Sb-124	1691.04	0.50			60.200	645.75	511.46
Sc-46	889.26	1.00			83.790	3345.6	2542.1
Sr-85	513.99	0.99			64.840	4017.8	2631.4

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays Energy(keV) Nuclide	Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
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Tb-160	215.65	0.04	217.00 Hf-179n	72.300	142.89	73.433
Tb-160	298.57	0.27		72.300	766.37	443.20
Tb-160	392.49	0.01	392.64 Ag-105	72.300	32.754	20.206
			392.85 Zr-88			
Tb-160	765.30	0.02	765.82 Nb-95	72.300	29.189	20.793
Tb-160	876.37	0.30		72.300	397.85	290.70
Tb-160	962.36	0.10		72.300	124.13	92.165
Tb-160	966.17	0.25		72.300	309.45	229.92
Tb-160	1002.89	0.01		72.300	12.056	9.0113
Tb-160	1115.12	0.02		72.300	17.896	13.589
Tb-160	1177.95	0.16		72.300	172.16	131.69
Tb-160	1199.89	0.02		72.300	25.490	19.544
Tb-160	1271.88	0.08		72.300	77.462	59.811
Tb-160	1312.12	0.03	1311.60 V-48	72.300	29.912	23.176
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Tc-95m	203.94	0.64	202.84 Xe-127	61.000	4747.3	2363.9
			205.78 Ir-192			
Tc-95m	582.15	0.36		61.000	1272.5	856.40
Tc-95m	616.50	0.02		61.000	50.914	34.692
Tc-95m	786.15	0.09		61.000	260.13	186.27
Tc-95m	820.65	0.05	821.11 Tm-168	61.000	135.88	98.100
Tc-95m	835.14	0.27		61.000	739.52	535.65
Tc-95m	1039.30	0.03	1037.83 Co-56	61.000	72.749	54.677
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Te-121	470.39	0.02		16.780	175.93	112.99
Te-121	507.54	0.19		16.780	2111.9	1379.3
Te-121	573.08	0.79		16.780	8058.8	5405.3
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Te-129m	695.98	0.03		33.600	120.80	84.429
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Tm-168	447.47	0.22		93.100	346.99	220.39
Tm-168	546.76	0.02		93.100	32.855	21.812
Tm-168	631.66	0.08	629.90 Pm-148m	93.100	95.187	65.196
Tm-168	645.71	0.01	644.55 Ag-105	93.100	17.040	11.725
			645.84 Sb-124			
Tm-168	720.32	0.11		93.100	123.93	87.225
Tm-168	730.61	0.04		93.100	49.077	34.641
Tm-168	741.32	0.11		93.100	121.44	85.966
Tm-168	815.95	0.46		93.100	474.54	342.23
Tm-168	821.11	0.11	820.65 Tc-95m	93.100	112.97	81.570
Tm-168	829.91	0.06		93.100	62.178	44.984

Isotope	Gamma Ray (keV)	Br Ratio	Conflicting Gamma-rays Energy(keV) Nuclide	Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
Tm-168	914.90	0.03	915.20 Pm-148m	93.100	27.592	20.314
Tm-168	1277.41	0.02		93.100	12.024	9.2890
V-48	511.00	0.98	5 other 511	15.970	28714.	18781.
V-48	928.20	0.01		15.970	230.60	170.19
V-48	944.30	0.08		15.970	1518.7	1124.1
V-48	983.50	1.00		15.970	18446.	13745.
V-48	1311.60	0.98	1312.12 Tb-160	15.970	14750.	11428.
Xe-127	202.84	0.68	203.94 Tc-95m	36.340	6357.8	3155.3
Xe-127	374.96	0.18		36.340	1090.2	665.67
Yb-169	307.68	0.11	308.44 Ir-192	32.030	653.22	380.70
Zr-88	392.85	0.94	392.49 Tb-160 392.64 Ag-105	83.400	3464.1	2137.4
Zr-95	724.18	0.44	722.78 Sb-124 725.21 In-114m 725.70 Pm-148m	64.020	1270.0	894.85
Zr-95	756.72	0.55		64.020	1539.0	1093.9