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Preliminary Evaluation of a Fluorescence and Radioisotope Nuclear Smuggling Deterrence Tag - Final Report (IL500E)

S. D. Hartenstein
R. Aryaeinejad
J. R. Delmastro
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Idaho National Engineering and Environmental Laboratory
Lockheed Idaho Technologies Company
Idaho Falls, Idaho 83415

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DISCLAIMER

PRELIMINARY EVALUATION OF A FLUORESCENCE AND RADIOISOTOPE NUCLEAR SMUGGLING DETERRENCE TAG - FINAL REPORT (IL500E)

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Investigators:

4-7-97
Steven D. Hartenstein, Principal Investigator
Date

R. Aryaeinejad.
4/7/97
Rahmat Aryaeinejad, Advisory Scientist
Date

Joseph R Delmastro
4/7/97
Joseph R. Delmastro, Staff Scientist
Date

Paul L. Tremblay
4/7/97
Paul L. Tremblay, Staff Engineer
Date

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 system uses three types of 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. This report also summarizes the efforts completed in identifying hardware that will be used for the tagging system. This hardware includes the devices for applying the tagging materials, the commercially available fluorescence detection systems, and gamma ray detection systems assembled from existing, commercially available technologies.

EXECUTIVE SUMMARY

This document represents the final deliverable for the Idaho National Engineering and Environmental 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 is achieved by using the tag to improve the accountability of SNM and providing readily traceable forensic evidence. The described tag is being developed for use by the international nuclear community.

This report describes the justification for this project, the technical requirements of the materials and methods, the 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 500g 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-live radioisotopes, and 10 fluorescent compounds have been selected as candidate materials for the tagging system.

Detection of the tag is based on the concept that fluorescent indicators and gamma emitting isotopes can be applied to a surface and then detected by their characteristic emissions. The uniqueness of the tag is established by assigning unique patterns of fluorophores and unique combinations of radioisotopes to each facility. Advantages of this tag are: 1) it can be detected and read with simple detector technology, 2) parts of the tag will be detectable for a minimum of twenty years, 3) very small quantities of the tag can be detected, 4) materials inside storage containers and packaging can be identified without opening the container, and 5) efforts to remove the tag will leave significant forensic residues.

This report contains the design criteria for and a preliminary design description of the hardware that will be used for the tagging system. This hardware includes the devices necessary to apply the tagging materials, the detection systems for the fluorescence compounds, and the detection systems for the gamma-emitting radioisotopes. The design criteria describe the capabilities that the hardware must meet to implement an effective tagging system within the limitations imposed by the program sponsor and the facilities potentially using the tags.

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The criteria address issues such safety, adaptability, sensitivity, and portability. Upon continuation of further funding, prototype tagging application and detection systems will be constructed and demonstrated on SNM and storage containers.

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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
ICP-MS	Inductively coupled plasma mass spectrometry
INEEL	Idaho National Engineering and Environmental 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

1. INTRODUCTION

This document represents the final deliverable for the Idaho National Engineering and Environmental 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 tagging system 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, and the proposed tagging methods. Also described are the candidate fluorescence compounds , candidate radioisotope compounds, candidate application solvents needed to effectively apply the tagging materials, candidate hardware necessary to apply the tagging materials, and candidate hardware to detect the fluorescent and radioisotope tags.

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 and Environmental Laboratory (INEEL) 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 Join 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 many cases. Most of these cases occurred during this decade.[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 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.

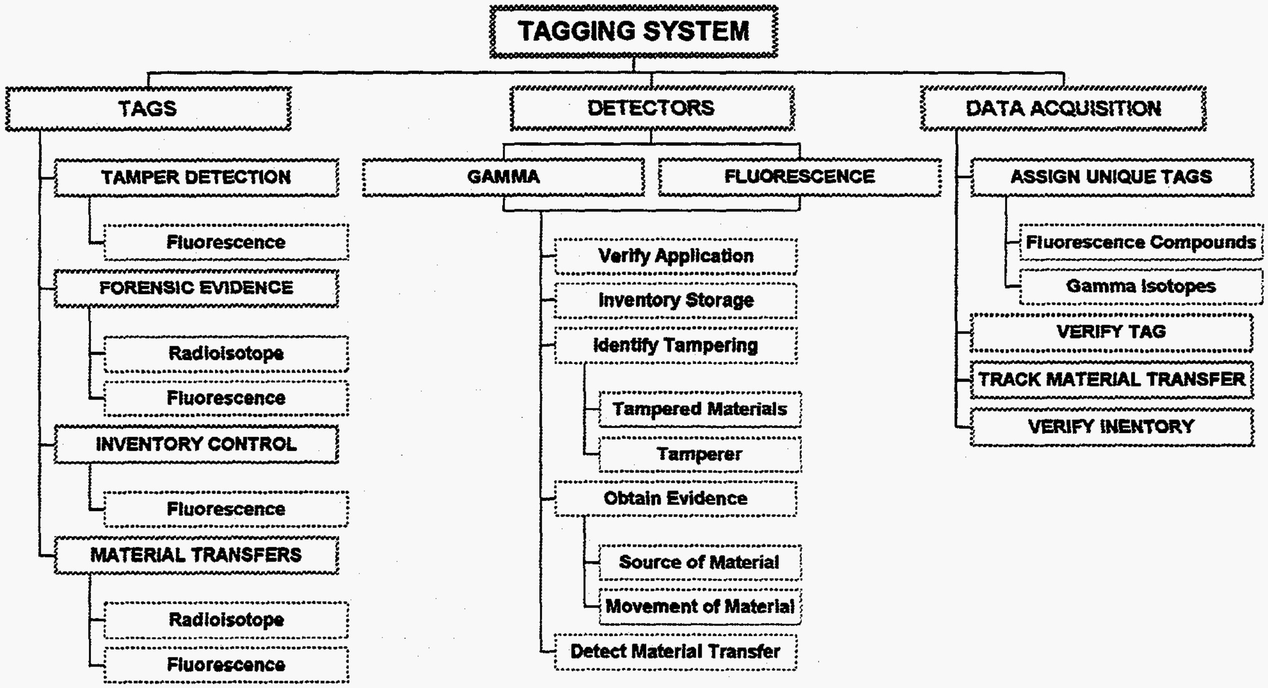

Figure 4-1. Functional features of tagging system.

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.

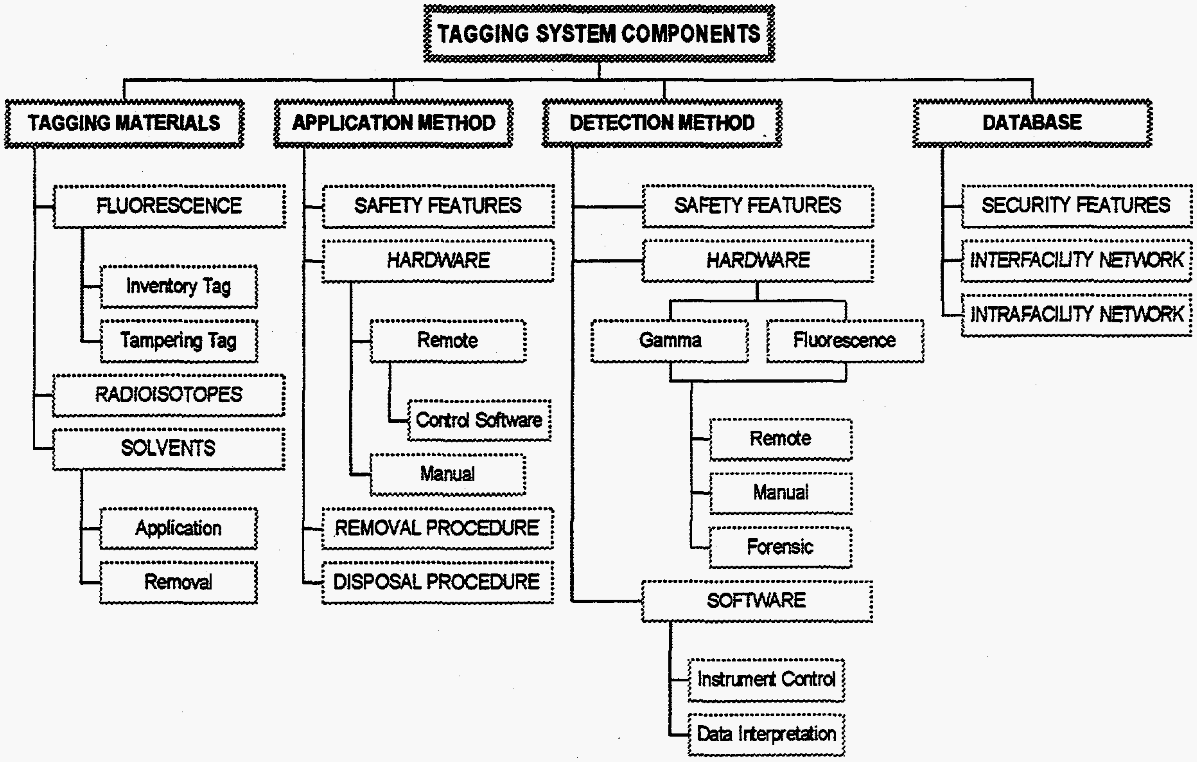

Figure 4-2. Tagging system components.

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 8, respectively.

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 ^{6}Li, 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 it 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 ($K_{effective}$); 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 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 6-1 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.

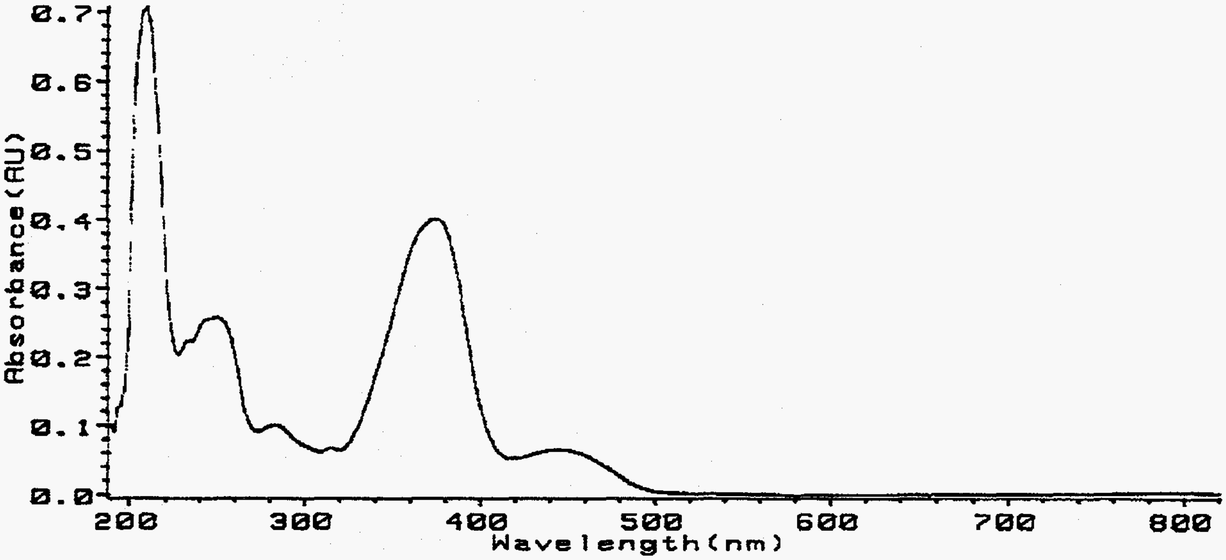

Figure 6-1. Absorbance spectrum for HM-704 in isopropanol.

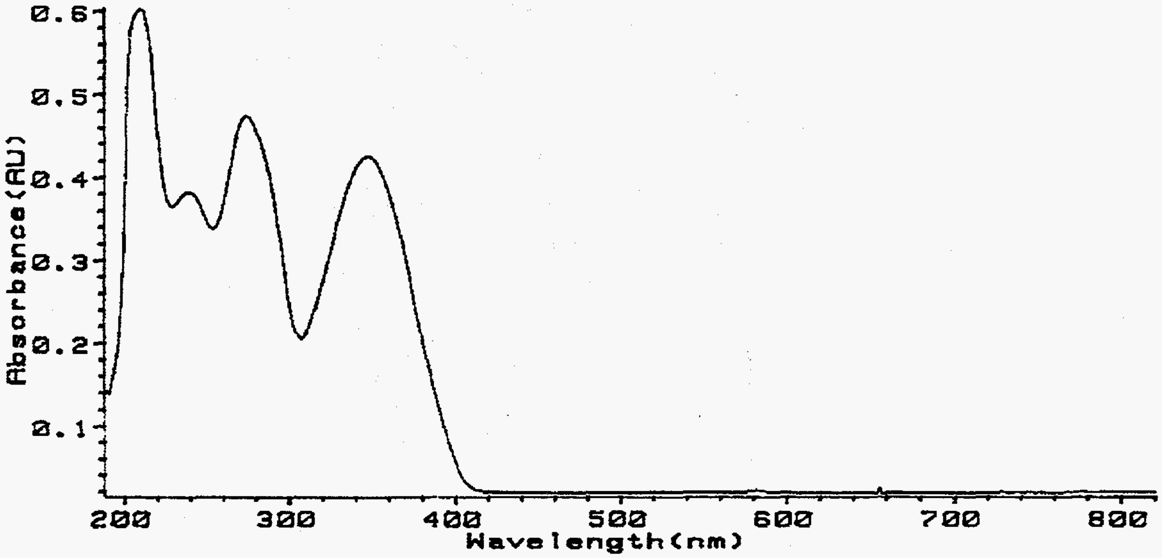

Figure 6-2. Absorbance spectrum for D-282 in isopropanol.

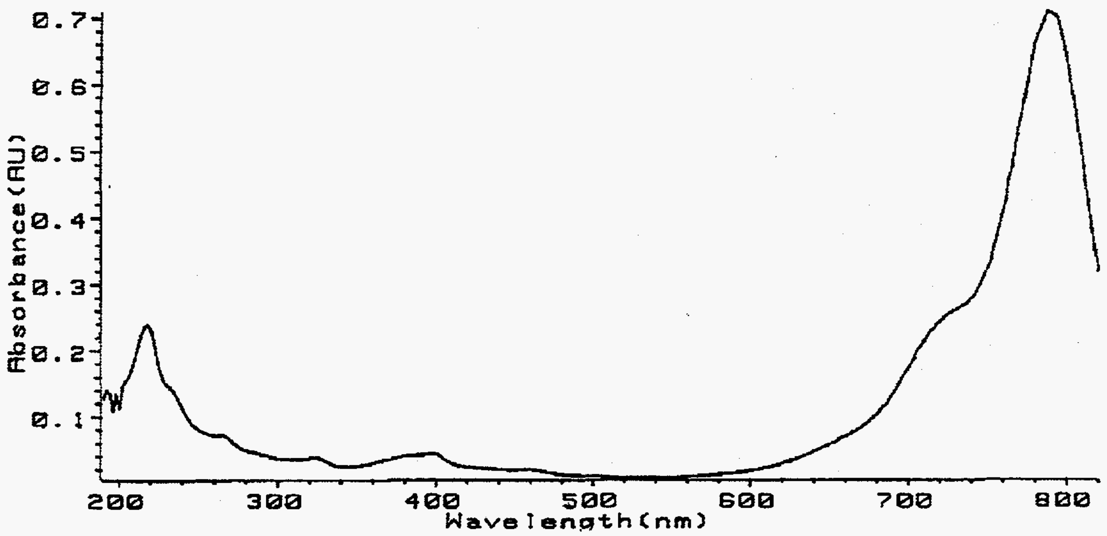

Figure 6-3. Absorbance spectrum of IR-125 in isopropanol.

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.

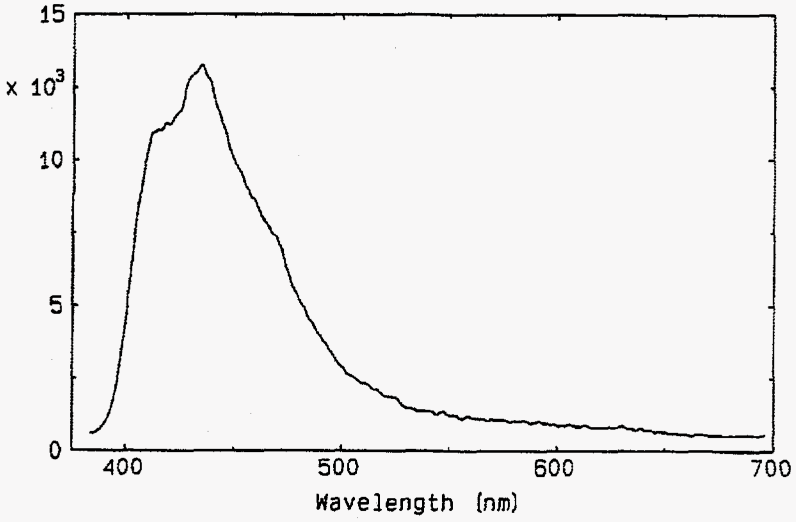

Figure 6-4. Fluorescence spectrum of D-282 under 365 nm excitation.

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.

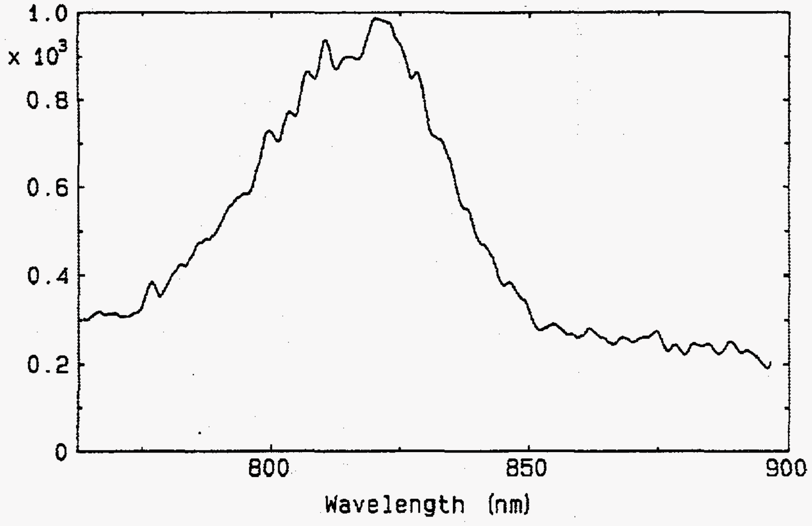

Figure 6-5. Fluorescence spectrum of IR-125 under 755 nm excitation.

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 compounds 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 cm^2 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.

6.2.6 Continuing Efforts for the Candidate Fluorescence Compounds

The current observations in the selection of candidate fluorescence compounds require continued evaluation of the materials as tagging candidates. These additional efforts should 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.

7. SELECTION OF CANDIDATE APPLICATION SOLVENTS

The tagging system 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. Design criteria for the application solvent are addressed in Section 7.1, whereas the selection process for candidate solvents is provided in Section 7.2.

7.1 Design Criteria for Application Solvent

The design criteria are listed to describe the capabilities of the solvent needed to effectively apply the tagging materials within the limitations imposed by the facilities using the tags. The criteria address issues such safety, solubility, drying time, and stability.

7.1.1 Safety

7.1.1.1 Personnel Exposure. Exposure to the application solvent will not exceed the American Conference of Governmental Industrial Hygienists' recommended threshold limit value time weighted average. The use of any carcinogens, if necessary, will be controlled to prevent personnel contamination during normal tag application operations.

7.1.1.2 Facility Hazards. The application solvent will not introduce physical hazards (i.e., fire) beyond the design specifications of the normal working environment for handling SNM.

7.1.1.3 SNM Reactions. The application solvent will not react chemically with the tagged SNM.

7.1.2 Visibility

To ensure that a pilferer will have difficulty removing all traceable portions of the tag, the location of the tag should be difficult to identify. To increase the level of difficulty in identifying the location of the tag, the application solvent for both the fluorescence compounds and radioisotopes will be colorless in the visible spectrum under ambient and ultraviolet lights.

7.1.3 Resolution

To maintain the applied pattern in a readable state, the application solvent will allow sufficient resolution between characters using either a manual implement (e.g., a marking pen) or an automated printing device. For the radioisotopic tags, the application solvent will allow for resolution between at least three 3mm x 10 mm strips (bars) of tagging materials. For the fluorescent tag, the application solvent will allow for imprinting at least five alphanumeric characters, which have a minimum characher size of five characters per cm, in the 4-cm^2 pattern space.

7.1.4 Drying Time

To maintain the integrity of the information designed into the pattern of the tag, the application solvent(s) will permit normal material handling rates at the facilities using the tag. The solvent matrix will be non-smearable within 20 minutes after applying the tag.

7.1.5 Composition

7.1.5.1 Impurities. To maintain with the fluorescence and radioisotope signatures, the solvent matrix will be of sufficient purity that it will not interfere with the detection of the fluorescence compounds and radioisotopes used in the tags.

7.1.5.2 Residue. The amount of organic residue remaining on the tagged SNM from the cured tag will conform to the SNM specification for loss-on-ignition (i.e., <0.5% total combustible mass/total mass of SNM).

7.1.5.3 Binding. To maintain specifications of the SNM being stored, the application solvent will minimize the binding between particles coated with the application solvent.

7.1.6 Solubility

7.1.6.1 Application of Tag. Because of the variety of applications for the tags, there may be two different application solvents, one for fluorescent compounds and one for radioisotopes. The application solvent(s) will have sufficient solubility to contain the necessary quantities of fluorescent compounds or radioisotopes in the solvent volume applied to the SNM surface.

7.1.6.2 Disposition of Tag. The tag must be difficult to remove to deter pilfering but possible to remove for appropriate processing purposes (e.g., transfers between facilities, inadvertent physical mishandling, aged tags, etc.,). Therefore, the application solvent will ensure that any residue from the application solvent that binds the tagging materials to the SNM will be difficult to remove when subjected to: 1) cleaning with soap and water or 2) rinsing with readily available volatile organic solvents or petroleum products. However, the solvent residue will be removable using a rinsing, chemical, or physical process that can be performed safely in a protective controlled environment. This removal process also will generate a radiological or chemical hazard to personnel removing the tag in an unprotected environment.

7.1.7 Volume

To minimize hazardous/mixed waste generation, the volume of solvent for each tag will be sufficiently large that the concentration of heavy metals or toxic organic compounds are below the Resource Conservation Recovery Act (RCRA) hazardous waste limits.

7.1.8 Lifetime

The inventory fluorescence tag will be simple and inexpensive to apply. Because of the destructive nature of radiation on organic compounds from the natural radioactive decay of the SNM, the fluorescence compounds will need to be replaced periodically. However, because of the security control and hazards associated with handling the stored materials, the application solvent for the fluorescent compounds will survive at least one year. Because the radioisotopic tag will be applied directly to SNM, which potentially could be sealed in cans indefinitely, the lifetime of the application solvent for the radioisotopes has not been determined.

7.2 Candidate Application Solvents

Because the fluorescence tag and radioisotopic tag have different functions, there could be two types of application solvents. Most organic fluorescent compounds only will fluoresce when they are in the presence of a solvent matrix whereas the radioisotopes will continue to decay in any matrix. Because of this difference, the application solvent for fluorescence compounds will consist of: 1) a liquid component which will quickly evaporate to provide for controlled application to a surface and 2) a solid residue component both to bind the fluorescent compound to the SNM and to provide a solvated environment for enhanced fluorescence. The application solvent for the radioisotopes will consist of a liquid component that can carry the radioisotopes into microcracks on the surface of the SNM. The need for the application solvent to deposit a solid residue that binds the radioisotopes to the SNM has not been established.

7.2.1 Material Suppliers

Commercial suppliers of application solvents were contacted for products which might meet the projected needs for the tags. Based on this search, several solvents 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 tags. A list of the solvents being evaluated is provided in Table 7-1.

Table 7-1. Listing of candidate application solvents.

	Manufacturer	Product Color	Commercial Use
Layout White DXX-327[a]	Dykem Company	white	layout dye
Tole Steel Blue DX-100[a]	Dykem Company	purple	layout dye
Spotcheck SKC-HF	Magnaflux Inc	transparent, colorless	cleaner, remover
Chem-Guard MP-250	Leander Lubricants	transparent, colorless	lubricant, penetrant
Dubl-Chek W-1	Sherwin, Inc	transparent, colorless	penetrant
Dubl-Chek HM-704	Sherwin, Inc	yellow-green	penetrant
Prism 454	Loctite Company	transparent, colorless	adhesive
I-2120[b]	Shannon Luminous Materials, Inc	transparent, colorless	marking ink
Bonn-Trace No. 191[b]	Bonneau Dye Corp	transparent, colorless	marker dye
PMI-B-WB[b]	Spectronics Corp	transparent, amber	marking ink

[a] The solvents for both Dykem products are the same, the color is provided by an additive.

[b] The solvent as received from the manufacturer contains a candidate fluorescent compound.

7.2.2 Visibility

The color of the application solvents is an important criterion in evaluating the candidate solvents 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. To evaluate this criterion, a small aliquot of each solvent was dropped onto an aluminum metal plate and viewed under ambient (Table 7-1) and ultraviolet light. Of the colorless candidate solvents, only Spectroline's PMI-B-WB marking ink left behind a colored residue. The visibility of this residue can be reduced significantly by minimizing the thickness of the applied solvent. The other products with visible color (Dykem layout dyes and Dubl-Chek HM-704 penetrant) were visible due to the pigments added by the manufacturer for the intended purposes of the products. In these cases, it is expected that the solvent without pigment could be purchased or the pigment could be removed through filtration, centrifugation, or column adsorption.

7.2.3 Patternability

The ability to control the location of the solvent when it is applied to a surface (patternability) will determine the amount of information that can be extracted from the tag. As an example, if the application solvent disperses over the entire 4 cm^2 area allowed for the tag, the only information available from the tag will be the fluorescent or gamma-energy spectra from the materials in the tag. Therefore, the only information that can be extracted from the tag would be the facility assigned the unique combination of materials in the tag. If a resolvable pattern of five of a set of twenty-five uniquely shaped characters can be patterned into the allowable space, a set of nearly 10^7 combinations of the patterns can be used as a traceable code to a data base which contains specific information about the tagged material.

With this criterion as a starting point, observations about the patternability of the application solvents were performed by dropping 25 to 50 μL aliquots of the solvents onto 5 cm x 5 cm plates of aluminum. Observations made for the materials are summarized in Table 7-2. From this set of experiments, the materials more patternable and easier to control (transfer cleanly and spread evenly) were those with a rapid drying time. These patternable solvents are candidates for the fluorescent tagging materials. The solvents that quickly and evenly spread over the metal, leave difficult to see residues, and dry quickly are candidate solvents for the radioisotopic tag.

7.2.4 Composition

7.2.4.1 Fluorescent Response. To optimize the use of the fluorescent compounds used to tag SNM, the application solvent cannot interfere with the fluorescence of the tagging materials. Therefore, the composition of each candidate solvent was reviewed for potential fluorophores and dried aliquots of the various candidate solvents were viewed under an ultraviolet light. Of the candidates, only those with fluorescent compounds already added as part of the product composition had any significant fluorescence.

Table 7-2. Patternability observations for the candidate application solvents.

Product	Observations	Drying Time
Layout White DXX-327[a]	Transferred cleanly and spread out evenly to a uniform height with good control after placement.	10 minutes, leaving a rough white 1 cm^2 spot.
Tole Steel Blue DX-100[a]	Transferred cleanly and spread out evenly to a uniform height with good control after placement.	4 minutes, leaving a dark violet spot 2 cm^2 spot.
Spotcheck SKC-HF	Transferred cleanly and spread out quickly as it wetted the entire surface of the coupon.	20 minutes, leaving no visible residue.
Chem-Guard MP-250	Transferred cleanly and spread out quickly as it wetted the entire surface of the coupon.	20 minutes, leaves a small residue shadow.
Dubl-Chek W-1	Transferred cleanly and spread out quickly as it wetted the entire surface of the coupon.	Did not dry.
Dubl-Chek HM-704	Transferred cleanly and spread out quickly as it wetted the entire surface of the coupon.	Did not dry.
Prism 454	Does not flow, needs to be wiped on surface. Did not move from applied location.	At least 1 hours to dry unless in thin film or under pressure. Formed an uneven, clear residue
I-2120[b]	Does not transfer smoothly. Forms long thin threads between surface and applicator. Difficult to control thickness. Did not move from applied location.	4 minutes, in thick, uneven clear residue.
Bonn-Trace No. 191[b]	Transferred cleanly and spread out quickly to a uniform height with good control after placement.	4 minutes to a clear, shadowed, 2 cm^2 residue
PMI-B-WB[b]	Transferred cleanly and formed an uneven drop which tended to hold air bubbles.	60 minutes to a amber, translucent 1 cm^2 residue.

[a] The solvents for both Dykem products are the same, the color is provided by an additive.
[b] The solvent as received from the manufacturer contains a candidate fluorescent compound.

7.2.4.2 Hazardous Components. Beyond the hazards associated with the normal handling of liquid chemicals, the candidate solvents represent several hazards which must be considered prior to their use. Most of the solvents use a rapidly evaporating flammable solvent to facilitate drying. For the candidates, these solvents include xylene (considered a carcinogen), acetone, light alcohols, butyl acetate, toluene, and ketones. Most of these solvents are flammable hazardous materials whose use in gloveboxes is not recommended. These solvents also are considered RCRA hazardous waste (F001 waste) in their pure state or RCRA characteristic flammable waste (D001) as mixtures. Because of these hazards and the potential to generate mixed waste when using the solvents, the amount of application solvent needed per tag and the configuration of the stock material will be closely controlled.

7.2.5 Retention

One significant criterion for the application solvents is in establishing that their use must make it difficult to remove the tagging materials from the SNM or containers being tagged. Ideally, the material would be impossible to remove without complicated chemistry or equipment. In reality, the task is to identify an application solvent that will leave detectable residues behind even though most of the tag can be removed with readily available solvents.

Table 7-3. Retention observations during rinses with removal solvents.

Product[a]	water with soap	Methylene Chloride	Toluene	Acetone	Isopropanol	Hexane
Tole Steel Blue DX-100[a]	resistant[b]	soluble[c]	resistant	soluble	resistant	resistant
Prism 454	insoluble[d]	resistant	resistant	resistant	resistant	not tested
I-2120	insoluble	resistant	soluble	soluble	resistant	resistant
Bonn-Trace No. 191[b]	insoluble	resistant	resistant	soluble	insoluble	resistant
PMI-B-WB[b]	soluble	resistant	resistant	soluble	resistant	resistant

[a] Approximately 50 μL of application solvent was spread over 2 to 4 cm^2, dried, and then were subjected to a 2 mL rinse (as consecutive drops over 1 minute).

[b] Some removal evident, but more than than 90% of the tagged material remained.

[C] *A significant amount of material visibly removed (>50%) with detectable residue remaining.*

[d] No material visibly removed.

To evaluate the candidate solvents for retention during removal activities, aluminum plates with dried aliquots of the solvents were rinsed with a variety of solvents to estimate their retention properties during rinse operations. The application solvents evaluated included only those that left behind a dried organic residue that binds the fluorescent material to the metal. During almost all of the tests, the rinsing solvents removed only a portion of the tag. The amount of the tag removed increased with larger volumes of the rinse solvent and with more physical wiping of the tag with the solvent. More specific results for the application solvents versus the rinsing solvents are provided in Table 7-3.

7.2.6 Continuing Efforts for the Candidate Application Solvents

Further evaluation of two candidate application solvents, Spotcheck SKC-HF - a naptha type solvent, and Prism 454 - a cyanoacrylate adhesive, was performed to determine if : 1) the necessary quantity of tagging materials can be dissolved in the small quantity of application solvent used to apply the tag; 2) the application solvent can bind the tagging materials sufficiently well to minimize inappropriate removal; and 3) the fluorescent compounds still fluoresce in the dried residue that binds them to metal surfaces.

Six fluorophores were tested with these two application solvents. Four of the fluorophores: UV Blue D-282, Columbia Blue D-298, 1-hydroxypyrene, and IR-125 had been characterized and used in our studies. The other two fluorophores selected for this study were both inorganic materials: yttrium vanadate doped with europium and zinc sulfide doped with traces of copper, aluminum, and gold.

7.2.6.1 Dissolution/Adhesion Tests with SKC-HF. One milliliter of SKC-HF was pipetted into each of six 6-mL vials and a small quantity of each of the six fluorophores added to each and mixed thoroughly. The only fluorophores which appeared to be even slightly soluble in this solvent were 1-hydroxypyrene, IR-125, and D-282. Fifty microliter aliquots of each of these solutions were pipetted on to stainless steel substrates and allowed to dry. The dried deposits were examined under UV illumination at both 254 nm and 356 nm. The colors observed under these conditions are summarized below in Table 7-4.

Table 7-4. Observations of fluorophores dissolved in SKC-HF on stainless steel substrates.

Fluorophore	Surface Color(254nm)	Surface Color(356nm)	Observations
YVO_4; Eu	Ruby red	Pale pink	Localized small spot
ZnS; Cu, Al, Au	Green	Green	Small green spot
1-hydroxypyrene	None	None	
IR-125	None	None	
D-298	Pink/rose-large area	Blue halo	Halo on perimeter
D-282	Pink- localized specks	Blue halo	Halo on perimeter

The lack of fluorescence for 1-hydroxypyrene apparently is due to the absence of an organic matrix to support the fluorescence. The localized spots for most of these fluorophores is an additional indicator of the limited solubility of the two inorganic fluorophores and the D-282 in SKC-HF. IR-125 does not fluoresce in the UV. Solutions of IR-125 fluoresce in the NIR; however we obtained some background fluorescence from the stainless steel substrate in this region when we scanned this substrate with a fluorimeter.

This experiment was repeated using pastes prepared by adding a small quantity of each bulk fluorophore to 50 µL of SKC-HF on a clean stainless steel substrate and then mixing thoroughly to form a "paste" of the mixture. The colors of the various deposits were observed while still wet at 254 nm and 356 nm and with room light after they had dried. These observations are summarized below in Table 7-5.

Table 7-5. Observations of "Pastes " of fluorophores mixed with SKC-HF on stainless steel substrates.

Fluorophore	Surface Color (254nm)	Surface Color(356nm)	Dry deposit-room light
YVO_4; Eu	Ruby red	Hot pink	White
ZnS; Cu,Al,Au	Bright lime green	Bright lime green	Pale yellow
1-hydroxypyrene	None	None	Brown
IR-125	None	None	
D-298	Sapphire blue	Sapphire blue	Pale purple
D-282	Turquoise/aquamarine		Lemon yellow

The substrates were examined again under UV irradiation after the solvent had evaporated. The colors were the same as for the wet deposits. The adhesion of the dry deposits to the stainless steel substrates was tested initially by tapping each substrate on edge three times. A significant quantity of the D-282 was removed by this treatment and some of the crystals of 1-hydroxypyrene was also removed. All of the other fluorophores appeared to remain on the substrates. This treatment was repeated a second time with similar results. It appears that the D-282 can be removed easily by tapping the substrate on edge. The bulk of the other deposits was easily removed from the substrates by brushing the substrate gently with a Q-tip or dry cotton swab or by brushing gently and then tapping the substrate on its edge. A dry Kimwipe was then used to wipe the surface of each substrate. Traces of the doped yttrium vanadate and the D-298 were still quite evident on the substrate while there were lesser amounts of the doped zinc sulfide and the D-282. All traces of the fluorophores could be removed from the substrates by wiping with a Q-tip soaked in 2-propanol. Therefore none of these fluorophores when used in SKC-HF solvent is a durable tagging marker.

7.2.6.2 Dissolution/Adhesion Tests with Prism 454. A small quantity of each of the six fluorophores was dispersed with Prism 454, a cyanoacrylate adhesive, on clean stainless steel substrates. The edge of a clean razor blade was used to mix two drops of the Prism 454 with several milligrams of each fluorophore. Although the deposits were nonuniform and contained undissolved particles, fluorescence was observed when the wet deposits were irradiated with UV light at 254 nm or 356 nm. The colors were similar to those obtained with the SKC-HF. Neither the 1-hydroxypyrene or the IR-125 exhibited fluorescence with excitation at either of these wavelengths. All these substrates were observed again with 254 nm and 356 nm excitation after drying overnight. The colors were similar to those observed with the SKC-HF fluorophore "pastes" except that the 1-hydroxypyrene fluoresces a weak blue-green in the dry deposit at both wavelengths and the D-298 fluoresces a lighter shade of blue than with SKC-HF.

Attempts were made to obtain more uniform deposits by two different techniques: (1) speading a thin layer of Prism 454 on the substrate before adding the solid fluorophores and (2) applying a solution of the fluorophore dissolved in SKC-HF, allowing it to dry, and then applying a coating of Prism 454 over the deposited fluorophore. Neither of these techniques was successful in obtaining homogeneous deposits for any of the fluorophores although better success was obtained with the 1-hydroxypyrene and the D-298. They are more soluble in the Prism 454 than are the other fluorophores investigated.

All of these deposits were more durable than those obtained with SKC-HF. None of the material was removed by tapping the substrates on edge. Gentle wiping with a Kimwipe did not remove any material although some of the fluorophore could be detected on the Kiwipe after wiping briskly. Wiping with sufficient pressure to abrade away the fluorophore which was not thoroughly covered by the film of Prism 454 still left sufficient material on the substrate to fluoresce strongly. Each substrate was scrubbed twice with 50-μL aliquots of 2-propanol and a clean Kimwipe. Although some fluorescent material was detected on the Kimwipe, the bulk of the deposit on each substrate remained intact. Although fluorescence from the 1-hydroxypyrene was weaker than for the other four UV excited fluorophores, it remained essentially unchanged after this cleaning procedure. It is likely that the material removed by cleaning with 2-PrOH is still the fluorophore which is not covered by or dissolved in the Prism 454. Some of the D-298 was transferred to clean areas of the substrate by the 2-PrOH cleaning procedure, which is probably due to the high solubility of this fluorophore in 2-PrOH.

The cleaning/removal procedure was repeated with 50 μL of acetone instead of 2-PrOH. The cyanoacrylate film was readily attacked by acetone and no trace of the doped ZnS or 1-hydroxypyrene remained on the substrates where they had been deposited; only small, isolated patches of the other fluorophores remained on the substrates. Attempts to quantify the amount of material removed by each of these treatments by weighing the substrates after each step were unsuccessful because of the small quantities initially present.

Of the six fluorophores studied, 1-hydroxypyrene and D-298 produced the most uniform, durable deposits with Prism 454. This is primarily due to their greater solubility. The quantity of fluorophore deposited in a film of Prism 454 may be increased by initially dissolving the fluorophore in a polar solvent such as acetone. Although acetone attacks films of Prism 454, it does not prevent the polymerization of this adhesive. D-298 is highly soluble in acetone so several milligrams were dissolved in acetone and then a small aliquot deposited on an aluminum substrate. Several drops of Prism 454 were deposited adjacent to this solution and the two mixed.

Although the two were not readily miscible, they were mixed and dispersed over most of the substrate. The Prism 454 dried without tackiness and the resulting film fluoresced an intense bright blue. Some of the fluorophore was readily removed by touching the surface; enough to contaminate fingertips or gloves, but the bulk of the deposit remained intact.

7.2.6.3 Visibility and Patternability of Bonn-Trace 191 Solvent. Earlier studies had indicated that Bonn-Trace 191 contains a fluorescent compound but leaves a residue on metal substrates which is visible under normal light. Attempts were made to reduce the visibility of the residue by diluting with several solvents with which it is at least partially miscible. The MSDS for this product indicated it contains approximately 14% toluene, 11% xylene, and 36% acetone. SKC-HF and several of these diluents were selected for investigation in an attempt to reduce the visibility of deposits and retain the patternability of Bonn-Trace 191. Initially Bonn-Trace 191 was mixed in several different ratios (1:1, 1:2, and 1:9) with SKC-HF and then small quantities were pipetted on to aluminum and stainless steel substrates. All three mixtures dispersed well on the substrates and the more dilute mixtures were significantly less visible on the substrates when viewed with room lighting. The 1:9 mixture was nearly invisible on aluminum substrates and showed only small changes in color near the outer edge of the deposits. The 1:1 and 1:2 mixtures deposited on stainless steel substrates consisted of two concentric rings, an inner ring which was nearly unaffected in appearance and an outer ring which made the substrate look grayer than the uncoated substrate.

The deposits on all substrates fluoresced a bright blue with stronger fluorescence at 365 nm than at 254 nm. The Bonn-Trace 191 was not entirely miscible with SKC-HF in a 1:9 volume ratio. Bonn-Trace 191 was then diluted 1:10 with toluene, acetone, and 4-methyl-2-pentanone (MIBK) and used to test its patternability on aluminum and stainless steel substrates.

Masks containing block letters or numbers 0.625 cm high were created from 3.75 cm by 5 cm Post-It notes by cutting the pattern into the adhesive-backed portion of the Post-It note with a razor blade or Exacto knife. Then neat Bonn-Trace 191 and the various 1:10 mixtures of Bonn-Trace 191 and toluene, SKC-HF, acetone, and MIBK were applied to the masks by rolling a Q-tip over the surface of the masks. Two different exposure techniques were used: (1) allowing the mixtures to dry on the masks before removing them from the substrates and (2) removing the masks from the substrates immediately after applying the fluorophore. The deposits were allowed to dry and then examined under normal room lighting and with UV illumination at 356 nm. The results of these experiments are summarized in Table 7-6.

The undiluted Bonn-Trace 191 produced more distinct pattern images for short exposure times. Thinning with acetone or MIBK clearly yielded better images for long exposure times with stainless steel substrates. If the mask is removed immediately after application of these mixtures to stainless steel, these solvents transport the fluorophore away from the region where it was originally applied, yielding a less distinct image. The aluminum substrates have a much higher degree of surface roughness than the stainless steel substrates and the diluted Bonn-Trace 191 behaves differently. The surface roughness allows the Bonn-Trace 191 diluted with acetone or MIBK to diffuse under the mask and transport the fluorophor away from the point of application, resulting in a very diffuse image (or no image at all). Bonn-Trace 191 diluted with toluene yielded the most distinct image patterns on both substrates for long exposure times and these were among the best image produced in these experiments.

Table 7-6. Patternability tests of Bonn-Trace 191 (BT 191) on aluminum and stainless steel substrates.

Mixture[a]	Substrate	Exposure Time[b]	Observations at 356nm
BT 191	Aluminum	Long	Indistinct pattern; all letters smudged
BT 191	Stainless Steel	Long	Wide line widths; letters smudged
BT 191	Aluminum	Short	Narrower line widths than for long exposure. Pattern of letters visible
BT 191	Stainless Steel	Short	Narrower line widths than for long exposure. Distinct pattern of letters visible. Image also visible with room lighting.
BT 191 SKC-HF	Aluminum	Long	Clear pattern of all letters visible
BT 191 SKC-HF	Stainless steel	Long	No pattern of letters visible
BT 191 Toluene	Stainless Steel	Long	Pattern visible. Dim outline of letters visible with room lighting.
BT 191 Toluene	Aluminum	Long	Pattern visible. Wider line widths than for dilution with SKC-HF.
BT 191 Toluene	Stainless Steel	Short	Pattern visible but of poorer quality than on aluminum substrate (wider line widths, weaker pattern.)
BT 191 Toluene	Aluminum	Short	Three of four letters clearly visible with narrow line widths.
BT 191 Acetone	Stainless Steel	Long	Distinct pattern of all four numbers.
BT 191 Acetone	Aluminum	Long	No pattern of numbers visible
BT 191 Acetone	Stainless Steel	Short	None of numbers are distinct - all smudged.
BT 191 Acetone	Aluminum	Short	Numbers not distinct; virtually illegible.
BT 191 MIBK	Stainless Steel	Long	Only two of four numbers legible.
BT 191 MIBK	Aluminum	Long	Only two of four numbers legible; wider line widths than with stainless steel.
BT 191 MIBK	Stainless Steel	Short	Only two of four numbers legible; poorer image than for long exposure.
BT 191 MIBK	Aluminum	Short	None of numbers legible.

[a] First four entries are for undiluted Bonn-Trace 191; all others are 1:10 (v/v) dilutions of Bonn-Trace 191 with solvent listed.

[b] Long exposure denotes mask left in place until mixture dried on mask; short exposure denotes mask removed immediately after applying mixture to mask.

The outline of images created on stainless steel are still visible with room lighting. This is clearly undesirable, especially if the tag consisted of numbers or letters, because it would alert a pilferer of *SNM* that the material is marked and show them the location of the tag. One alternative to using a tag consisting of numbers and/or letters would be to use a pattern composed of a series of small dots which create a readable pattern which is much less obvious as a tag.

A series of masks, containing a pattern consisting of small holes created with a fine hypodermic needle, were prepared in the adhesive-backed portion of 3.75 by 5 cm Post-It notes. The spacing between adjacent holes in a row was 0.5 cm and the holes in the adjacent rows were offset by 0.25 cm. These masks were applied to stainless steel and aluminum substrates and the neat Bonn-Trace 191 or Bonn-Trace diluted 1:10 with acetone, MIBK, or toluene applied to the masks with a Q-tip using one of these two techniques: (1) rolling the Q-tip over the entire surface of the mask or (2) applying the fluorophor only to the holes in the mask. The Bonn-Trace 191/acetone and Bonn-Trace 191/MIBK mixtures were applied to the entire surface of the masks and the masks allowed to dry before removing the masks from stainless steel substrates. When illuminated at 365 nm, the hole pattern could not be detected although some areas of the substrate contained bright spots of the fluorophor. Application of Bonn-Trace 191 and Bonn-Trace 191 diluted 1:10 with toluene by rolling the Q-tip over the entire surface of the mask and then immediately removing the masks from the stainless steel substrates replicated only a part of the mask pattern . The rest of the pattern was indistinct.

Application of Bonn-Trace 191 and Bonn-Trace 191 diluted with toluene to aluminum and stainless steel substrates by using a Q-tip to deliver the fluorophor directly on to each hole and then immediately removing the mask from the surface of the substrate yielded more satisfactory results. The dot pattern from the mask was almost totally replicated by the undiluted Bonn-Trace 191 but only part of the pattern was replicated by the Bonn-Trace 191 diluted with toluene. Some of the pattern observed with the diluted Bonn-Trace 191 was totally missing and some dots were not bright, well-formed, or distinct. The desired resolution of 0.5 cm between adjacent dots and 0.25 cm between rows was achieved with the undiluted Bonn-Trace 191. This pattern of dots was invisible on aluminum substrates with room light. However, the pattern on the stainless steel substrate was still visible with room light, due to the change in refractive index of the surface.

8. 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 8.1. The selection process for candidate radioisotopes is provided in Section 8.2.

8.1 Design Criteria for Radioisotopic Tag

8.1.1 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.

8.1.2 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.

8.1.3 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.

8.1.4 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.

8.1.5 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.

8.1.6 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.

8.1.7 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.

8.1.8 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.

8.1.9 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.

8.1.10 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.

8.1.11 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.

8.2 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 8.2.1 through 8.2.8. The candidate radioisotopes are presented in Section 8.2.9.

8.2.1 Half-life

8.2.1.1 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.

8.2.1.2 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.

8.2.2 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 $K_{effective}$ 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}%.

8.2.3 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" (0.625 cm) of stainless steel is shown in Table 8-1. 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 8-1. Efficiency of gamma-ray transmission through materials.

Energy	% of Gamma-ray Transmission	
(keV)	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

8.2.4 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.

8.2.5 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.

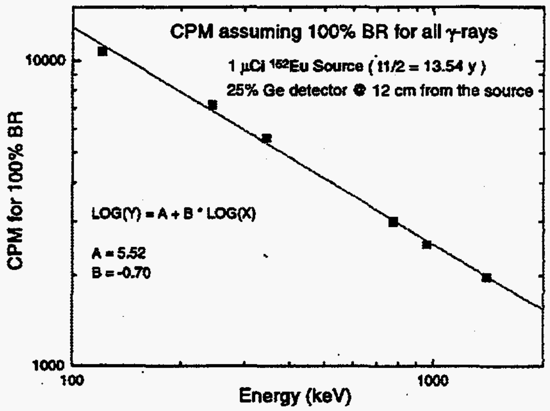

Figure 8-1. CPM versus gamma-ray energy.

8.2.6 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 8-1), 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 5 cm (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 8-1). 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).

8.2.7 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 8-2). These data were then fitted using a two-exponential decay method to derive the formula for calculating the attenuation:

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

Where $Y =$ photon cross-section in barns
$X =$ gamma-ray energy in keV

and the values for the other parameters were equal to:

$X_0 = 136.4$ $Y_0 = 3.984$
$A_1 = 8.714$ $T_1 = 40.61$
$A_2 = 8.409$ $T_2 = 488.0$

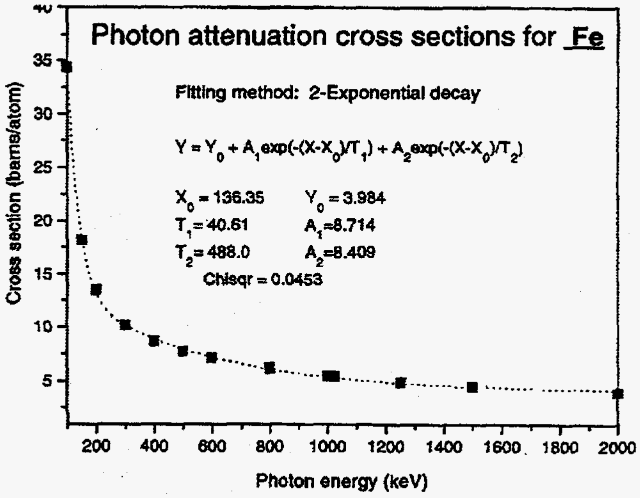

Figure 8-2. Photon attenuation versus energy.

8.2.8 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 7.5 cm x 7.5 cm (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.

8.2.9 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 8-2 and 8-3, respectively. This tables list isotopes with acceptable half-lives, gamma-ray energies of ≥200 keV, and branching ratios ≥1%.[7,8,9] Tables 8-2 and 8-3 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 8.2.1 through 8.2.8, 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.

8.2.10 Biological Tampering Tag

There are several short-lived radioisotopes such as ^{95m}Tc, ^{203}Hg, ^{105m}Ag, ^{114m}In, and ^{85}Sr in Table 8-3 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.

8.2.11 Continuing Efforts for the Candidate Radioisotopes

The current list of candidate isotopes for the tagging systems will require further 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 8-2. 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 8-3. 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

8.2.11.1 Persistence of Cesium and Strontium Salt Residues on Metal Substrates.

Earlier experiments indicated that SKC-HF dries without leaving a residue on metallic substrates. To determine if sufficient quantities of cesium and strontium salts dissolved in this solvent would penetrate the surface of substrates and survive various removal techniques, the following experiments were performed with the non-radioactive salts. Stable Cs and Sr salts were used for ease of handling although the quantities required for detection by chemical analysis are many orders of magnitude more than the quantities determinable radiochemically. Due to the limited solubility of cesium and strontium chlorides in pure SKC and their moderate solubility in ethanol, we initially planned to dissolve these salts in EtOH and then dilute with SKC. However EtOH and SKC are immiscible, whereas 2-PrOH and 4-methyl-2-pentanone (MIBK) are each miscible with SKC when mixed in a 1:1 volume ratio. These two solvents were each used to dissolve small quantities of cesium and strontium chlorides and SKC was then added in a 1:1 volume ratio.

Small quantities of the salts were also added to pure Bonn-Trace 191, which is another excellent penetrating solvent. The solubility of both salts in each of these three solvent mixtures (SKC/2-PrOH, SKC/MIBK, and Bonn-Trace 191) was still quite limited so that the concentrations of these solutions could not be determined directly. The concentrations were determined by analyzing the solutions resulting from leaching deposits of 25-µL quantities of each salt/solvent mixture on glass microscope slides. These deposits were allowed to dry and then leached for 48 hr in a known volumes of DI water. These leachates were analyzed for Cs and Sr by ICP-MS. The same volumes of these three solutions were pipetted on to stainless steel and aluminum substrates and allowed to dry. The following sequence of removal techniques was then used sequentially on these substrates.

Dry wipe step. The surface of each substrate on which the salt was deposited was wiped with half a dry laboratory Kimwipe (actual size 10.6 by 11.3 cm) and then the material removed was rinsed from this wipe by placing it in a small polyethylene funnel and slowly passing 20 mL of DI water through the wipe and collecting it in a 30-mL poly bottle. The excess water was removed from the Kimwipe by pressing it with the bulb of a disposable poly dropper.

Wet wipe step. Half a standard Kimwipe was wetted with 200 µL of water and then the surface of each substrate on which the salt had been deposited was wiped with this wet Kimwipe. The material removed from each substrate was rinsed from the wipe by placing it in a small polyethylene funnel and then slowly passing 20 mL of DI water through the wipe. The rinsings from wiping each substrate were collected in 30-mL poly bottles. The excess water was removed from the Kimwipe by pressing it with the bulb of a disposable dropper.

Static 15-min leach. Each substrate was placed with the deposit side up in 20 mL DI water in a plastic weighing boat and allowed to remain for 15 min. Then the leachate from each substrate was transferred to 30-mL poly bottles.

Static 24-hr leach. Each substrate was placed, with the side on which the material had originally been deposited facing up, in a plastic weighing boat containing 20 mL of DI water. The substrate was allowed to remain in the boat for 24 hr and then the leachate was transferred to 30-mL poly bottles.

All samples were acidified with nitric acid to a concentration of 1% HNO_3 and then most of the samples were analyzed by ICP-MS. The samples generated by leaching Cs and Sr from the glass microscope slides were analyzed in order to estimate the total quantity of material originally deposited on each substrate. Both the cesium and strontium chlorides were significantly more soluble in the 1:1 mixture of 2-PrOH and SKC than in the other two solvents. All the analysis results are presented below in Tables 8-4 and 8-5. Table 8-4 includes all the results for stainless steel substrates while Table 8-5 presents only the results for the SKC/2PrOH and SKC/MIBK mixtures on aluminum substrates. The aluminum substrates on which the Cs and Sr salts dissolved Bonn-Trace 191 were deposited were not analyzed because the solubilities were very low in this mixture.

Table 8-4. Concentrations of Cs and Sr Deposited On and Removed from Stainless Steel Substrates

Solvent	SKC/2-PrOH	SKC/MIBK	BT-191	SKC/2-PrOH	SKC/MIBK	BT-191
Solute	Cs ng/mL	Cs ng/mL	Cs ng/mL	Sr ng/mL	Sr ng/mL	Sr ng/mL
Glass slide[a]	330	3.5	2.6	78	20	3.6
Dry wipe	80	<[b]1.3	<1.3	6.2	3.5	2. 6
Wet wipe	220	3.3	<1.3	13	5.5	3.8
15 min leach	<1.3	1.7	<1.3	2.3	<[c]0.87	<0.87
24 hr leach	2	<1.3	<1.3	<0.87	<0.87	<0.87

[a]Calculated from concentrations determined by ICP-MS and dilution factor.
[b]Denotes measured detection limit for the determination of Cs by ICP-MS.
[c]Denotes measured detection limit for the determination of Sr by ICP-MS.

Table 8-5. Concentrations of Cs and Sr Deposited On and Removed from Aluminum Substrates

Solvent	SKC/2-PrOH	SKC/MIBK	SKC/2-PrOH	SKC/MIBK
Solute	Cs ng/mL	Cs ng/mL	Sr ng/mL	Sr ng/mL
Glass slide[a]	330	3.5	77	20
Dry wipe	61	<[b]1.3	3.7	2.9
Wet wipe	270	1.9	42	5.1
15 min leach	29	1.8	16	<[c]0.87
24 hr leach	2.6	1.8	14	<0.87

Symbols for footnotes are the same as for Table 8-4.

These results indicate that although the bulk of the cesium and strontium is removed in the dry wipe and wet wipe procedures, there are still nanogram quantities remaining on the substrates which are difficult to remove and require additional washing or leaching. This is most evident for the results obtained with SKC/2-PrOH where the quantities deposited are significantly greater than for the other mixtures. (The actual nanogram quantities of Cs and Sr removed from the 5 cm x 5 cm substrates are twenty times the tabulated concentrations because the leachate volumes for all surface treatments was 20 mL.) These results indicate that it would be difficult to remove all traces of radioisotopic Cs and Sr tags from metal surfaces.

9. SELECTION OF CANDIDATE TAGGING MATERIAL APPLICATION SYSTEM

The fluorescence inventory tag and the radioisotope identification tag will be applied to the surfaces of SNM and the SNM storage containers. Because of the cost for application technology, the potential need for high throughput, and the potential hazards associated with the tagging materials; the tagging material application system will require both manual and automated techniques to "paint" the tagging materials onto the tagged materials. Section 9.1 describes the design criteria for the application techniques and Section 9.2 describes some candidate technologies for applying the tag.

9.1 Design Criteria for the Tag Application Technology

9.1.1 Safety

The taggant supply to the printing or painting mechanisms will minimize a worker's exposure to the hazards of the taggant and solvent materials. The refill of taggant reservoirs will be via sealed containers or solid encapsulant.

9.1.2 Compatibility with Taggant Materials

The materials of construction for the tagging application technologies will not degrade when exposed to the taggant materials,

9.1.3 Waste Generation

The taggant application systems will lessen waste generation through designs that minimize dry-out of taggant reservoirs and excessive dispersal of solvents.

9.1.4 Size Constraints

9.1.4.1 Size of Tag. The taggant application systems will be designed to apply the necessary tagging material patterns within an area of 2 cm x 2cm.

9.1.4.2 Size of Material to be Tagged. The taggant application systems will be designed to paint tags on metals and powders. For metals, the tagged surface is expected to vary from flat surfaces to spherical surfaces. It is not expected that resolvable patterns will be deposited onto powders.

9.1.5 Lifetime of Components

The taggant application systems will be designed for maximum life with modularity of critical elements.

9.2 Candidate Mechanical Printing Methods

In its simplest forms, the tag application system will be a set of manually operated pens containing the individual taggant species in a volatile carrier solvent. More complex printing mechanisms for automated/remote use in process boxes will be designed for hazardous environments or extensive processes with multiple types of items to be tagged.

9.2.1 Manual Tagging Application System

To minimize cost and for ease of application, the option to manually apply the tagging materials to surfaces must be available. To accomplish this, the tagging material could be applied as a solvent and painted onto the surface or sprayed onto the surface as an aerosol. However, this option would introduce several significant safety issues. Painting on the surfaces with a brush probably would require the use of open reservoirs of flammable solvents. Spraying of the tagging materials would lead to widespread contamination and exposure throughout the operating environment. For these reasons, the manual tagging application will use a system similar to an ink marking pen for metals.

Hand manipulated pens containing individual species of taggants will provide flexibility in an out-of-process environment for tagging. Pen types suitable for handling taggants in a volatile solvent include fountain, drafting, ball point, and fiber tipped, all of which, when constructed of suitable materials, will accommodate any desirable solvent. Fountain pens, the oldest technology in this set, are likely to leak solvent and use a hardened applicator tip which could permanently mar the surface to be marked. Ball point and drafting pens have similarly hardened tips. Fiber tipped pens, similar to the commercially available "Sharpie"©, use a fiber wick applicator and are unlikely to leak solvent or mar surfaces. Examples of commercially available models containing visible and invisible inks are shown in Figure 9-1. One pen contains a thick resin ink that is squeezed onto a surface, another is a wax crayon, and the other is a marking pen containing a rapidly drying solvent.

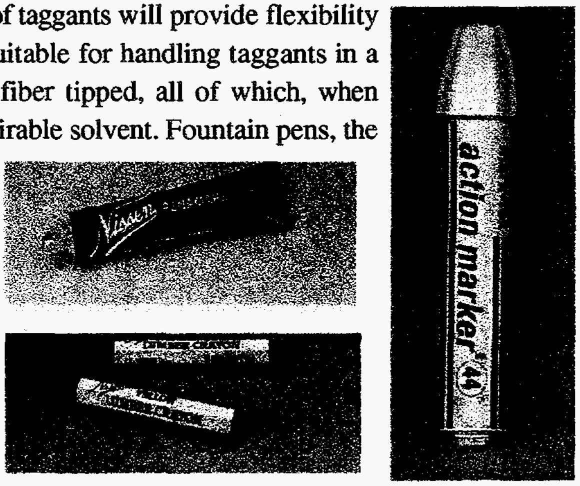

Figure 9-1. Examples of manual marking devices.

9.2.2 Mechanical Tagging Application System

Because the nuclear tagging system will use a variety of tagging materials, the mechanical tagging application system is envisioned to be similar to those systems that are used for automated color printing. However, the tagging application system will differ from the commercial systems in that the inks will be invisible or radioactive and the printing surfaces will be metal.

There are four different mechanisms in the current state-of-the-art of color printing which are candidate methods for applying the tags. These methods have proven themselves in the marketplace, and they differ significantly from the historical methods of relief, intaglio, surface, and stencil processes common to commercial printing. These methods include: 1) bubble (or wax) jet, 2) laser electrostatic, 3) toner jet and 4) wax transfer systems. Of these, the bubble jet systems offer the only practical solution for mechanized application systems, because they require neither configured substrates nor accessory materials.

The pumping mechanism (Figure 9-2) for bubble jet printers relies on a thermal process which forms a minute bubble of gas within a chamber of the nozzle.[11,12] Expansion of the gas drives a picoliter sized drop of liquid out of the nozzle. All of the bubble jet systems can print densities well within the five characters per 4 cm^2 constraint assigned in the design criteria. This process has the potential disadvantage of sequestering taggant materials within the bubble chamber resulting in unsatisfactory variation of taggant concentrations on the tagged surfaces.

As an alternative to the thermal process, the bubble jets can use a piezoelectric pump. The piezoelectric pump uses mechanical force to propel small quantities of the ink onto surfaces. This process does not separate solvent and ink additives and therefore avoids concentration variability. Another significant advantage of the piezoelectric pump models is that there are three nonexclusive taggant options available with a piezo-pumped system: water-based taggants, volatile organic solvent-based taggants, and solid encapsulant materials (waxes).

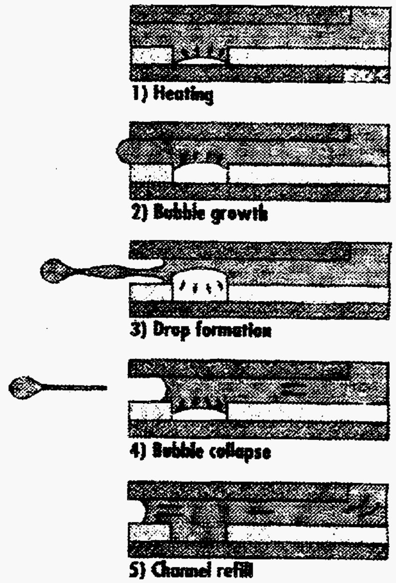

Figure 9-2. Bubble-jet nozzle.

9.2.2.1 Solvent Resistance Tests for Print Cartridges and Printing Mechanisms.

Sample ink-jet systems from Hewlett-Packard and Canon were tested for their resistance to the solvents expected to be used with the fluorescent and radioisotope taggants. The ink cartridges were tested by applying a 10-µL aliquot of each solvent tested to a readily accessible surface of the ink-jet cartridge and continually observing the surface until the solvent had completely evaporated. Both brands of ink-jet cartridges resisted attack from SKC-HF, a xylol-based solvent, for a minimum of 5 min. Close examination of the Hewlett-Packard cartridge revealed that the main bladder is made of neoprene rubber, which may not resist this solvent for long periods of time. The Canon cartridge is made of molding grade polyethylene and should be resistant to most organic solvents below 50°C. When tested with Bonn-Trace 191, the surface of the HP cartridge melted immediately upon challenge with this solvent/solids mixture. None of the deposit could be removed from the

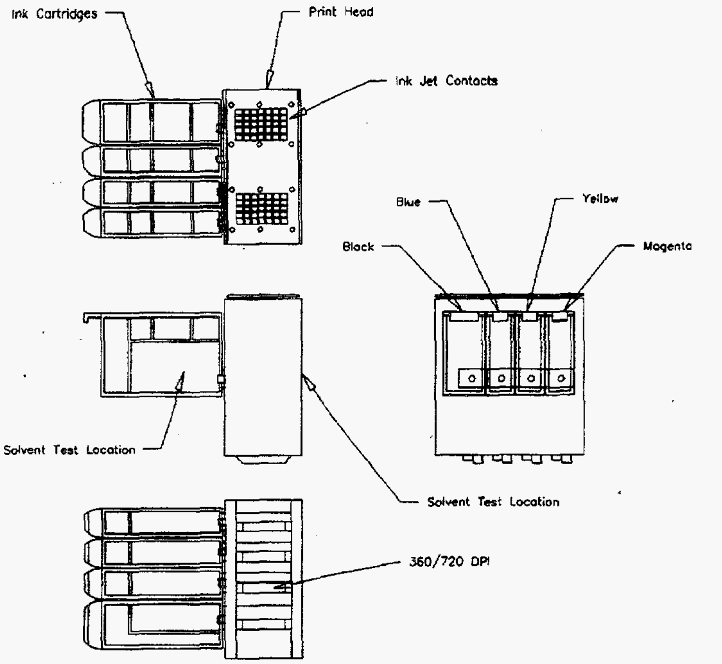

Figure 9-3. Sketch of Canon ink-jet cartridge and print head mechanisms.

surface after the Bonn-Trace 191 had dried. The Canon cartridge resisted the Bonn-Trace 191 entirely. The solids deposit containing the UV fluorescent material could be removed completely by simply peeling it off the surface with a fingernail. No traces of fluorescence were detectable on the cartridge after the deposit was peeled off. No further testing of the HP system was performed because the cartridge did not have adequate solvent resistance.

The Canon ink-jet printing mechanism is composed of a different type of plastic than the ink cartridge so it was necessary to also test its solvent resistance. It appeared resistant to SKC-HF for a longer time than was required for complete evaporation of this solvent. The area where the 10-μL aliquot was deposited was inspected more carefully under 50X magnification and revealed no evidence of damage. Testing of this mechanism with Bonn-Trace 191 revealed a slight attack at the periphery of the solvent blot, where traces of fluorescent material were detected with a UV lamp after removal of the dried deposit. Under 50X magnification, this attack appeared as filled-in areas between asperities of the surface molding markings. No dye was fould in the area where the bulk of the solvent aliquot was initially deposited. This may be due to a differential created in the solvents by the positive meniscus at the periphery. This would result in the more volatile components of the mixture escaping in this region and thus having a greater effect on the plastic body with concomitant transport and deposition of the fluorescent material.

Figure 9-3 illustrates the Canon ink-jet cartridge and print head mechanisms. It is annotated to indicate the locations where the solvents were applied for the tests described above.

9.2.2.3 Modification of Canon BJC-240 Printer for Use with Taggant Materials. Following successful solvent resistance tests of the Canon printing system, a Canon BJC-240 printer was procured for follow-on testing. This device would be reasonably simple to modify for process marking of components with taggant materials. Figure 9-4 shows the printer as received. Figure 9-5 illustrates the modifications necessary to remove and replace the manufacturer's ink with the selected taggant/solvent. Two 1/8 in. holes are drilled into one of the ink chambers(in this case, the yellow dye chamber). These are threaded for 8-32 screws to preclude leakage and drying of the ink following replacement. Figure 9-6 illustrates the method of removing and replacing the ink in a cartridge chamber. The pipette is used to pierce a foam filter within the cartridge on the left side allowing removal and replacement of the commercial ink. There is 8 mL of ink in each chamber. It is composed of approximately 70% isopropyl alcohol, 26% glycerine and 4% of an oil based colorant. The replacement ink used in the tests was D-298 fluorescent dye, dissolved in isopropanol. Figure 9-7 illustrates the placement of the ink cartridge within the printer. Use of flat headed screws would allow all three chambers of the cartridge to be used for dye replacement. Figure 9-8 shows an experimental placement of test coupons for test printing. The colors are individually addressable by the software provided with the printer, so printing from one or a combination of chambers is possible. Observation of the printing process reveals that a a mixture of colors to produce black is accomplished in multiple passes of

Figure 9-4. Canon BJC-240 printer.

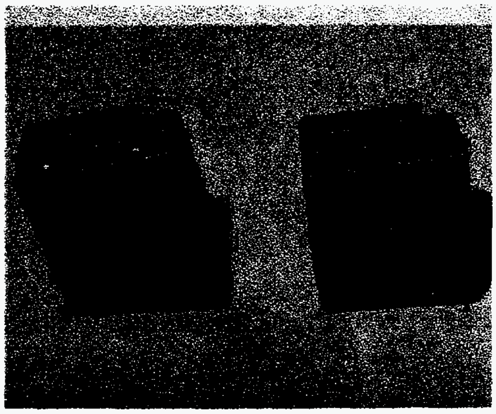

Figure 9-5. Unmodified and modified Canon ink cartridges.

Figure 9-6. Filling ink cartridge.

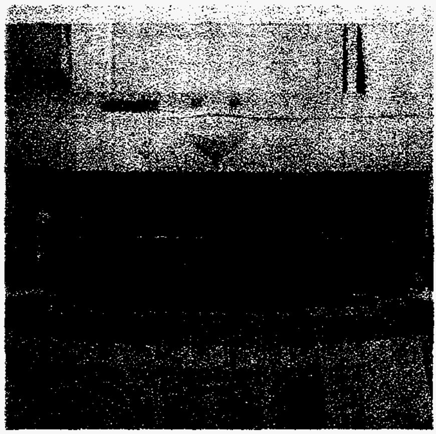

Figure 9-7. Placement of modified ink cartridge within the printer.

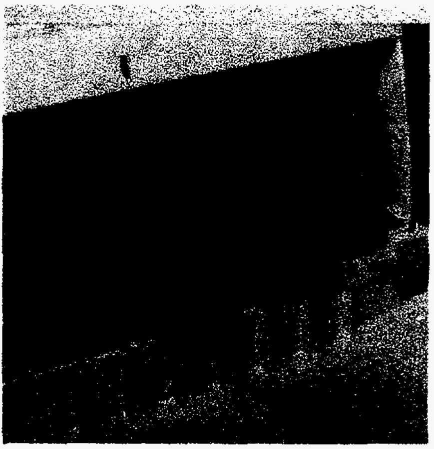

Figure 9-8. Placement of coupons for printing.

the cartridge with a minute translation of the printed surface between passes. This complicates the modification of this particular printer for use with mixed colors on production line blocks of metal as the means of translation of the blocks for correct registry to each color head would have to be accomodated.

9.2.2.4 Test Results with Canon BJC-240 Printer. The printer was cycled through a test program provided by the manufacturer which paints a full page of a rotating alphabet, numerals and punctuation characters for each phase of the testing. Following replacement of 1.5 mL of ink with D-298, fluorescence from the characters printed was not observed after several pages of various papers had been printed. With one exception, it was found that paper and the transparency films tested contained a fluorescent dye apparently used for brightening. Its spectrum matches D-298 and would seem to be nearly ubiquitous in papers, cloth and projection films.

After a paper was identified (a black construction paper) that had not been treated with a UV fluorescent brightener, it was determined that the lack of fluorescence of the characters was due to interference by the original ink remaining in the cartridge. Replacement of greater proportions of ink with the taggant dye resulted in successful excitation of the D-298 characters under UV challenge.

Tests were performed on aluminum foil "pages" as well as the coupons shown above. Without modification, the printer does not accept samples greater than 25 mils in thickness. Presence of any of the original colorant prevents the printed characters from drying on a metal surface. Any trace of oil (from even a fingerprint) on the surface of the metal will result in a beading up of the characters so as to become unreadable. If the metal surface is free of oil, the characters are clear and sharp.

9.2.2.5 Conclusions from Work with the Canon Printer. The Canon BJC-240 is a simple 3-color printer, easily modified to accept solvent-based dyes for various tagging applications. It was not tested against multiple solvent taggants such as Bonn-Trace 191. It would have to be extensively modified to print characters on production line blocks of metal. More appropriate commercial printing systems using the same heat driven ink jet are available which are configured for this task.

While it appears that it could be modified to overcome its liabilities, the Canon BJC-240 system is inadequate in its present form for use as a production line tagging device. Its printing mechanism would have to be modified to accommodate materials thicker than 25 mils. A taggant material which is not normally present in substrate materials must be used with an ink-jet printer system. Metal surfaces to be tagged must be free from all traces of oil, which interfere with the correct formation and registry of characters. Suitable roughening of the surface of metal substrates may enable more successful tagging with imprinted letters and characters. A tagging/solvent system which dries more rapidly than the D-298 must be used to minimize smudging of the printed tag.

10. SELECTION OF FLUORESCENT TAG DETECTION SYSTEM

The fluorescence detection system includes the hardware to detect the applied tags manually, remotely, and forensically. 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.

10.1 Design Criteria for Fluorescence Tag Detection System

10.1.1 Safety

10.1.1.1 Personnel Radiation Exposure. The configuration of the fluorescence detection system will minimize personnel exposure to the ionizing radiation characteristic of the materials being tagged. This will be accomplished by minimizing the time needed to collect the fluorescence information, maximizing the distance (remoting) and barriers (shielding) between operators and tagged materials.

10.1.1.2 Personnel Light Exposure. The configuration of the fluorescence detection system will minimize personnel exposure to the fluorescence excitation energy needed to generate the fluorescence response.

10.1.1.3 Facility Hazards. The detection system will not introduce physical hazards (i.e., energy source, neutron moderator) beyond the design specifications of the normal SNM working environment.

10.1.2 Configuration

Based on the variety of planned uses of the fluorescence tags, there will have to be several types of fluorescence detector configurations. These configurations include systems for: 1) detection in process locations; 2) detection in storage locations, and 3) detection of forensic evidence. A sketch of how this detection hardware may appear is shown in Figure 10-1. For implementation in process locations or storage locations, the detection probe could be held manually or scanned robotically, with the fluorescence pattern read manually or automatically from a video screen. For forensic evidence, the residual tamper detection fluorescence dusting powders or residual amounts of fluorescent compounds will be detectable with a hand-held probe or with a commercially available fluorescence spectrometer.

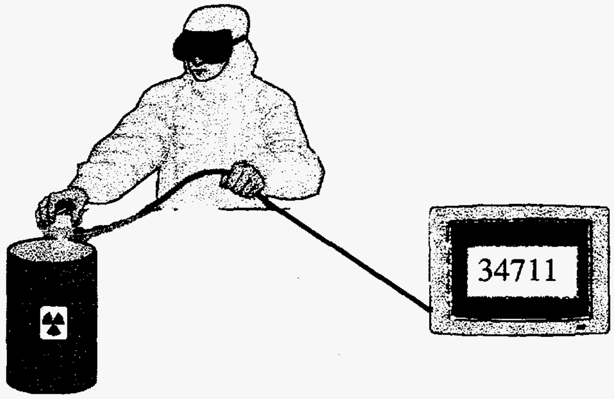

Figure 10-1. Fluorescence Detection System.

10.1.2.1 Process Validation of Tag Application. The configurations of the fluorescence detection systems will allow for both automated and hand-held detection of the applied fluorescence tag at a processing location (i.e., glovebox) to validate the application of the fluorescence tag and to track the movement of material through a facility's processes.

10.1.2.2 Inventory Control. The configurations of the fluorescence detection systems will allow for both automated and manual detection of the applied fluorescence tag in a storage area (i.e., bunker or vault) to inventory the tagged material.

10.1.2.3 Forensic Detection of Tagging Materials. The configurations of the fluorescence detection systems will allow for both field and laboratory identification of fluorescence residue serving as forensic evidence.

10.1.3 Spectral Regions

To ensure that the number of fluorophores available for use as tags is not limited by the system's hardware, the fluorescence detection systems will have the flexibility to excite or detect ultraviolet, visible and near infrared fluorescence.

10.1.4 Resolution

The detection system will resolve a fluorescence pattern of at least five alphanumeric characters, which have a minimum character size of five characters per cm, in a 4-cm^2 pattern space. This resolution will allow for visual reading of the pattern by directly observing the tag and by imaging on a video screen.

10.1.5 Detection Surfaces

The fluorescence detection system will be able to resolve the fluorescence pattern of the tagged material from a flat or rounded metal surface. Even though there will be no pattern on tagged powders, the detection systems will be designed to identify the presence and composition of the fluorescence tagging material.

10.1.6 Sensitivity

10.1.6.1 Inventory Tag. To ensure that the pattern of the inventory fluorescence tag can be identified with minimal personnel exposure and no radioactive cross-contamination, the process and inventory control fluorescence detection systems will have sufficient sensitivity to read a tag without contacting the tagged surface. Because it is expected that the radiation from the SNM will degrade the intensity of the fluorescence inventory tag's emission, the detection system will have sufficient sensitivity to read a tag that contains at least 5% of the original tagging material.

10.1.6.2 Forensic Evidence. To ensure traceability to the facility of origin when a tag has been tampered with, the forensic fluorescence detection system will have sufficient sensitivity to detect and identify the fluorescent compound when at least 0.01% of the fluorescent material is present.

10.1.7 Availability to Users

To ensure that all potential users of the tagging system can obtain the fluorescence detection systems, no component in the detection systems will be limited by export restrictions.

10.2 Fluorescence Detection Systems

To provide detection of the fluorescence materials there will be three types of detection systems. These systems include: 1) hand-held devices for quick, manual detection of the fluorescence pattern or fluorescence contamination, 2) remote imaging systems for fluorescence pattern recognition and recording, and 3) laboratory fluorescence systems for forensic evaluation. To limit the development costs of the tagging system, the fluorescence detection systems will be based on commercially available hardware with limited modifications as necessary.

10.2.1 Hand-held Fluorescence Detection Systems

The hand-held detection systems provide an inexpensive option for verifying the application of fluorescence tags to surfaces, performing manual inventories, and screening areas and personnel for tampering. These systems would be used if the implementing facility chooses to have personnel manually observing and recording the tagging information. To cover both the ultraviolet and near infrared spectral regions, two types of systems will be necessary.

10.2.1.1 Ultraviolet Fluorescence. Ultraviolet (UV) lamps will be used as the excitation source to cause the fluorescent tags to emit visible light. This light will be detected by personnel via direct observation of the illuminated area. These lamps are readily available in a variety of hand-held configurations(Figures 10-2 and 10-3). Generally a 4-Watt lamp costing less than $200 can excite visibly detectable fluorescence both in solutions containing high parts per million levels of fluorophores and in microgram quantities of fluorescing powders. For glovebox applications, a free standing lamp using a 15-Watt bulb will allow the tag to be read from a greater distance and also will free an operator's hands for manipulation of the tagged material. This hand-held hardware is inexpensive, flexible, and readily available. However, its disadvantages include:
1) the use of mercury lamps to generate ultraviolet excitation light may result in the generation of mixed waste if the device becomes contaminated. 2) Manual detection of the fluorescence tags will result in increased radiation doses to personnel, because of the decreased distance between personnel making the measurement and the material with the tag. 3) The ultraviolet light used to excite the visible fluorescence may result in increased safety hazards such as UV exposure to the eyes, degradation of organic materials in gloveboxes, or a spark/ignition source.

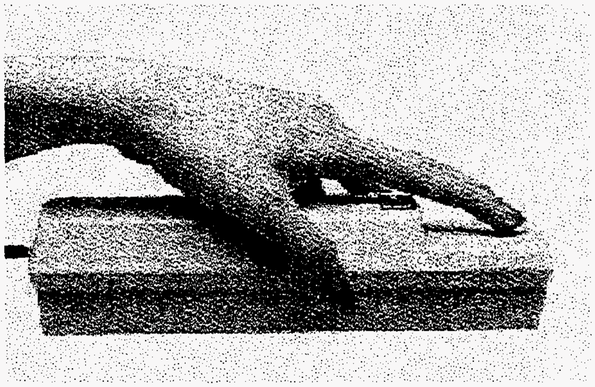

Figure 10-2. Hand-held 4-Watt UV lamp.

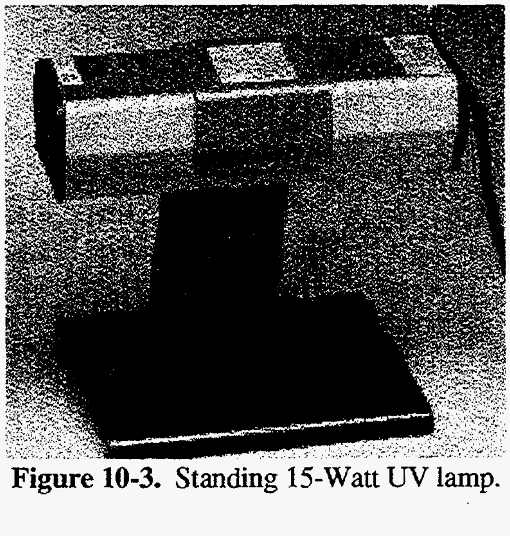

Figure 10-3. Standing 15-Watt UV lamp.

10.2.1.2 Near Infrared Fluorescence. Near Infrared (NIR) excitation sources will be used to excite NIR fluorescence that will be detectable via a monitoring screen. This screen's hardware or software will convert the NIR image into a visible display. A detection system with properties similar to the system needed for the tagging system is commercially available for aligning lasers, viewing activities in dark rooms, etc. The hand held units cost less than $2000 per unit and are very light and portable. For this type of detection system, the units would have to be modified to include an appropriate NIR excitation source.

10.2.1.3 Examples of Hand-held UV Lamp Applications. The extent of the use of a hand-held lamp detection system is demonstrated for several of the candidate fluorescence materials in Figures 10-4, 10.5, and 10-6. The sensitivity and manual readability of a hand-written fluorescence image is shown by comparing two metal coupons (one with an invisible fluorescence tag and one without) under room light and under a UV lamp (Figure 10-4). The ability to detect small quantities of tamper indicating dusting powders on parts is depicted in Figure 10-5. In this figure, milligram quantities of three different fluorescence powders were dusted on metal coupons and then compared against a blank coupon under room and UV light. The ability to identify personnel or equipment that is contaminated by contacting material coated with a dusting powder or trying to remove a tag with a solvent is demonstrated in Figure 10-6. In this figure, a glove that is contaminated with several dusting powders and a glove that has been contaminated with a fluorescent dye while trying to wash a tagged material are compared with an unused glove.

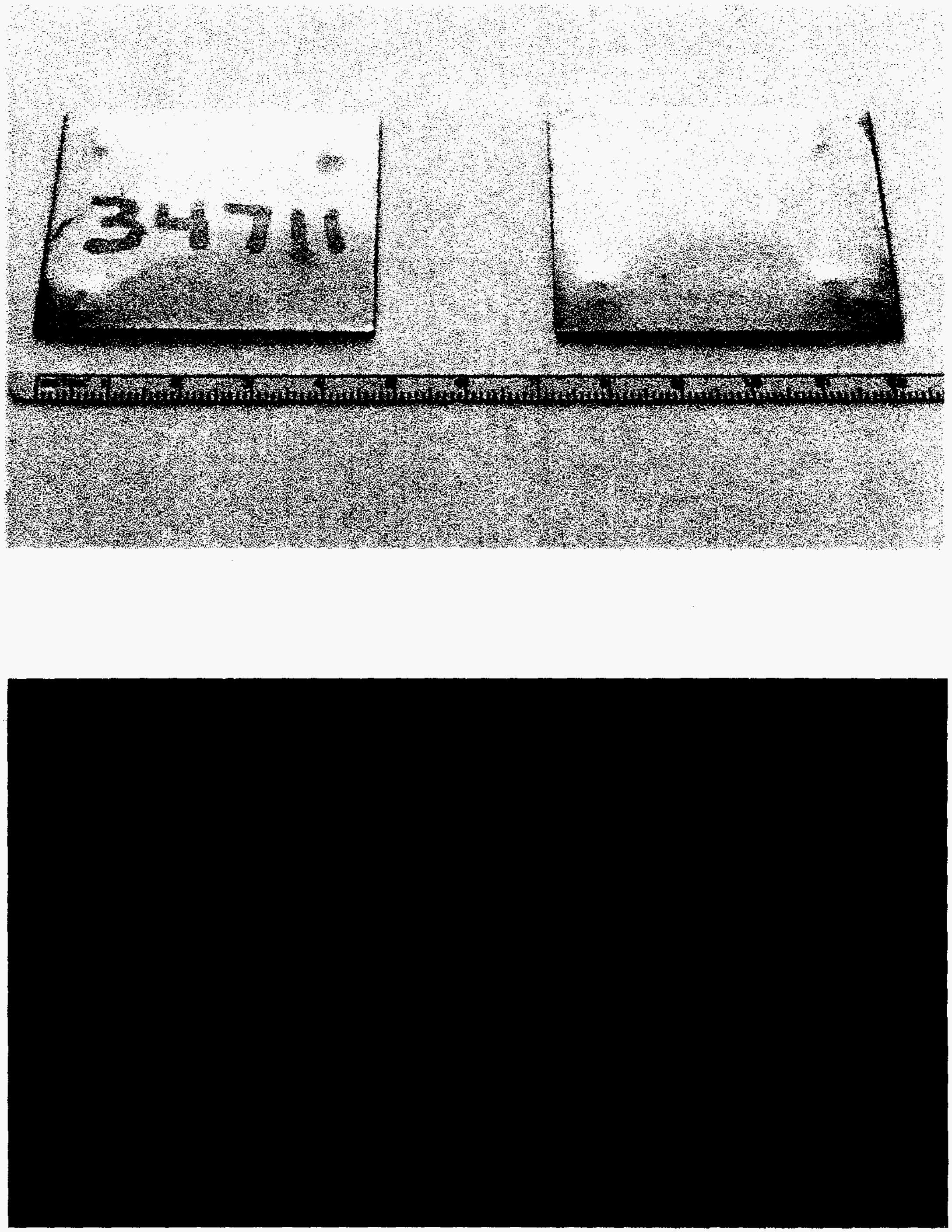

Figure 10-4. Tagged and untagged metal coupons exposed to room light (top) and UV light (bottom).

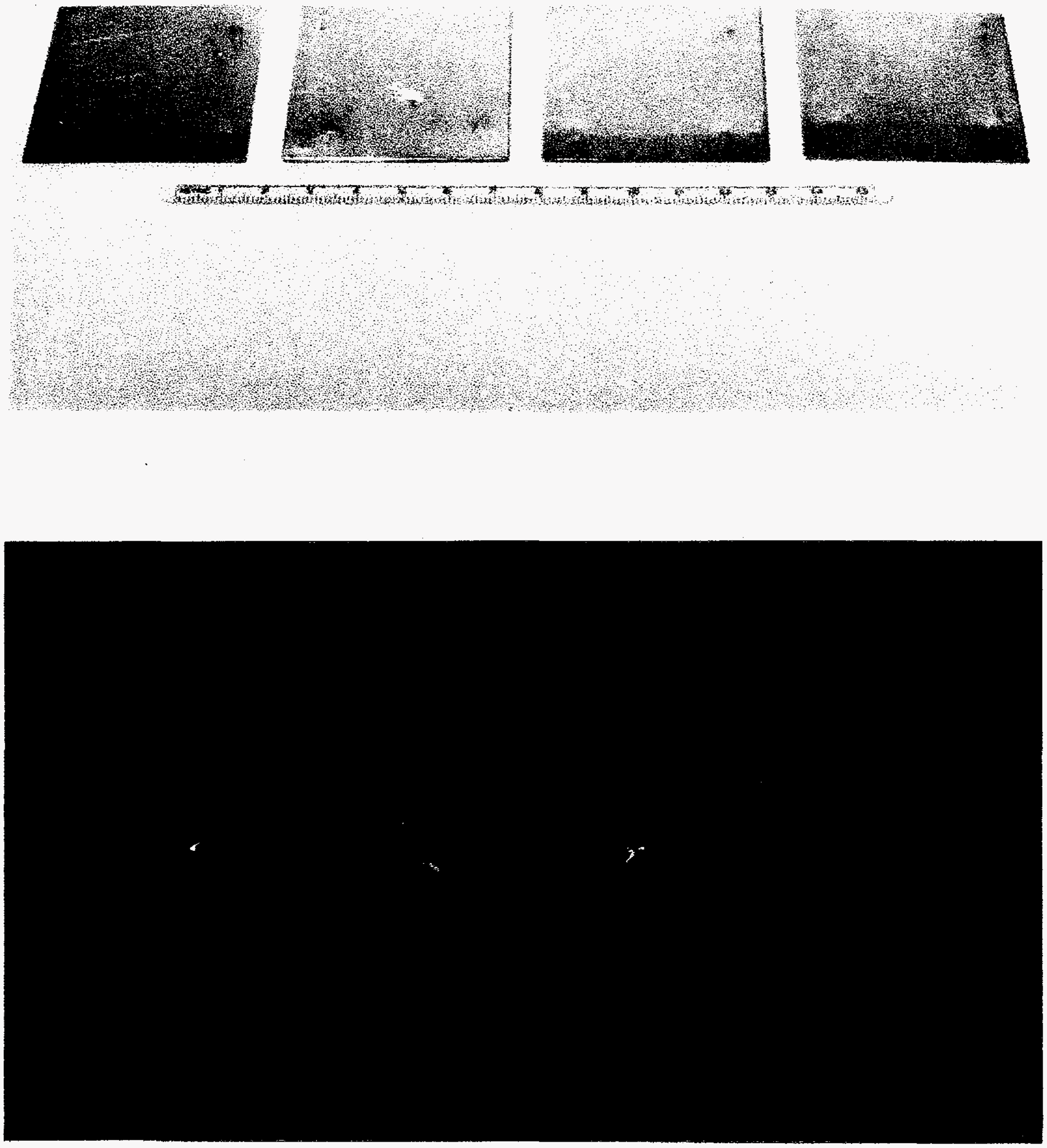

Figure 10-5. Metal coupons with small quantities of tamper indicating dusting powders exposed to room light (top) and UV light (bottom). A different dusting powder is on each coupon except the blank coupon on the right.

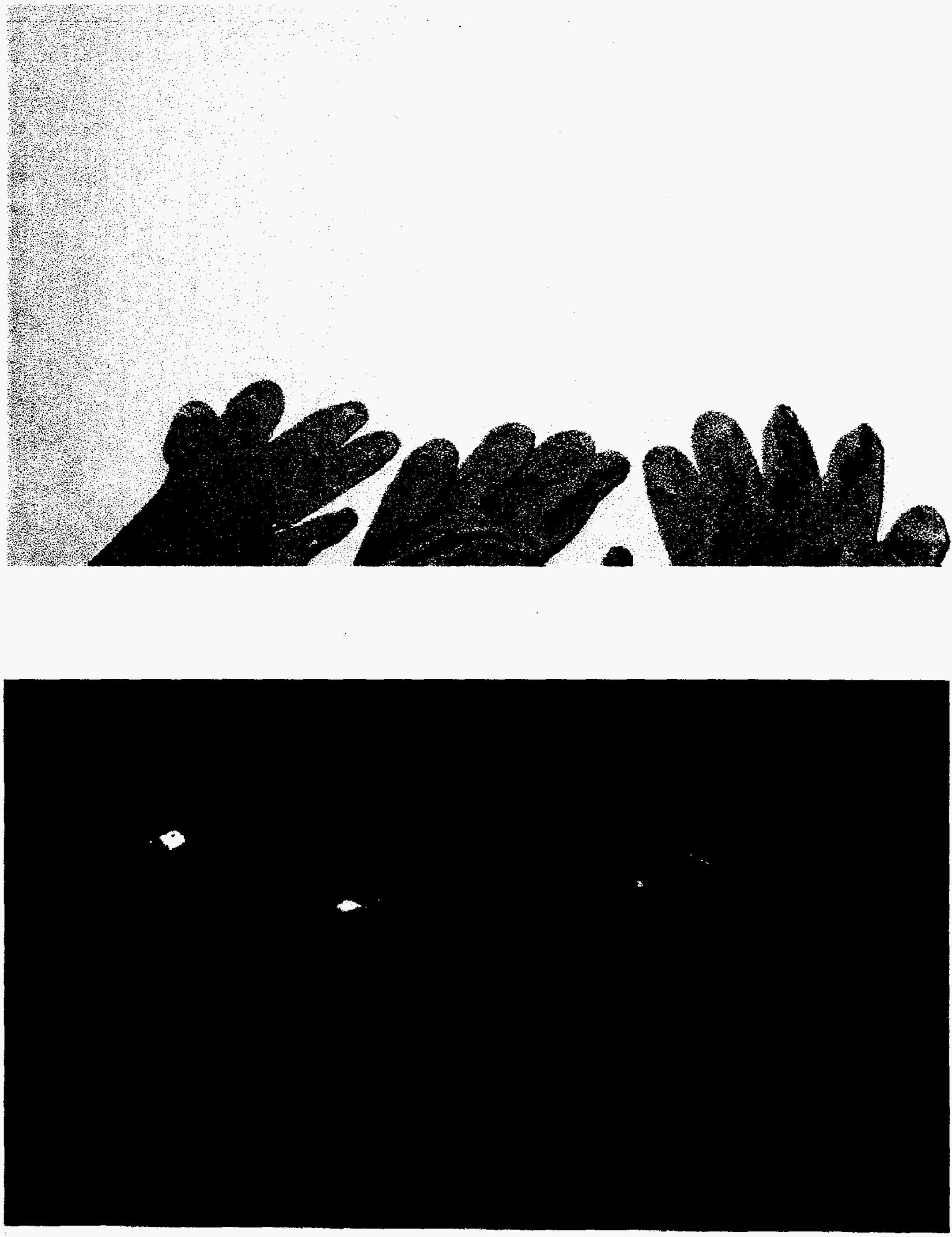

Figure 10-6. Rubber gloves exposed to room light (top) and UV light (bottom). The left glove has a different dusting powder on each finger, the center glove is uncontaminated, and the right glove was contaminated by removing a tag with isopropanol.

10.2.2 Remote Fluorescence Detection Systems

The hand-held detection systems provide the simplest option for detecting the fluorescence tags; but a remoted and automated detection system will minimize radiation exposure to personnel. Remote detection system will be designed to be used in a glovebox or in a automated surveillance system. The detection system will be constructed from: 1) a fiber optic probe that transmits excitation light to the tagged surface and returns the emission image, 2) a video display of the emission image for manual confirmation of the tag, and 3) a pattern recognition program for reduction of the fluorescent image.

As described, this type of detection system is a combination of commercially available systems for remote inspection of metal or electronic components and bar code reading systems. The expected cost of the detection system can range from $3K for a remote camera that can transfer a manually read image from a video screen to $50K for a boroscope system that can collect and store electronic digitized images. Examples of two commercially available systems that may be used for the remote detection system are shown in Figure 10-7 and Figure 10-8.

The first system (Figure 10-7), Majik-Cam manufactured by Dynatron, is a miniature video camera connected to high resolution monitor or standard video cassette recorders. For the SNM tagging system, this device system would have to be modified: 1) to incorporate a UV and NIR excitation source with either an excitation lamp placed in the glovebox or excitation light transmitted through fiber optics to the viewing area. The second system (Figure 10-8) is a boroscope manufactured by Olympus Industrial. This system ($>40K) is designed for viewing the inside of hard-to-reach areas and is available with a UV source for metal fatigue analyses. An add-on capability which allows for electronic storage of images is available.

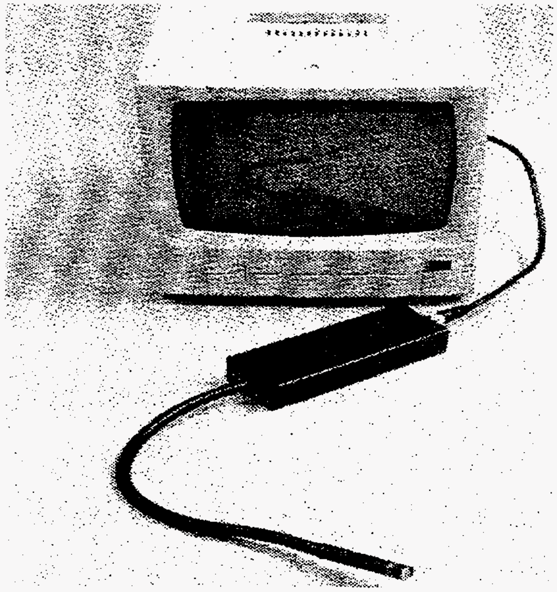

Figure 10-7. Majik-Cam video imager.

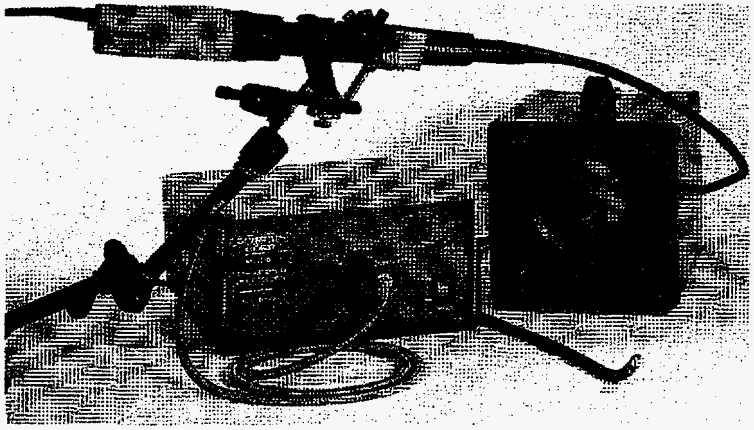

Figure 10-8. Olympus boroscope imaging system.

10.2.3 Laboratory Fluorescence Forensic Evidence Detection Systems

The hand-held and remote detection systems provide the capabilities necessary to track material for material control and accountability. However, these systems lack the flexibility and sensitivity to evaluate forensic evidence from personnel or materials involved in smuggled SNM. When these situations occur, it is expected both that the recovered material will have trace quantities of the tagging materials remaining and that the individuals illicitly handling the SNM will have been tagged with trace quantities of the tagging materials. Therefore to trace the facility of origin of the SNM using the fluorophores, it will be necessary to have detection equipment which can identify the chemical composition of the tag.

An example of how an analytical-grade fluorometer could be used is demonstrated in Figures 10-9 and 10-10. The set of candidate fluorescence compounds, currently under evaluation for the tagging system, contains several compounds that emit a blue fluorescence when exposed to UV light. The manually observed fluorescence indicates that all of the compounds are equivalent. However, when some of these compounds are evaluated with a spectrofluorometer, the observed spectra indicate that there are at least two distinct compounds (Figure 10-9). Additionally, after scanning the compounds with a different excitation wavelength, there appears to be at least three different compounds in the set (Figure 10-10). This ability to discriminate between compounds even at the ppm level will allow analysis of residues to assist in the determination of the facility of origin for recovered SNM.

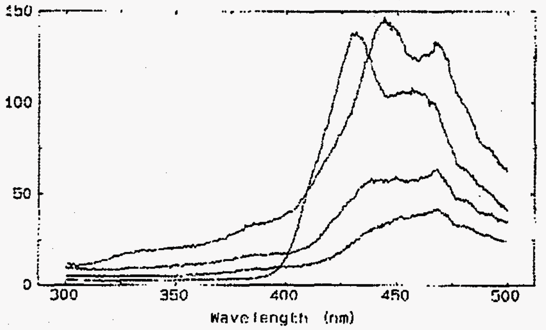

Figure 10-9. Fluorescence spectra for four of the candidate tagging materials, excitation with 254 nm.

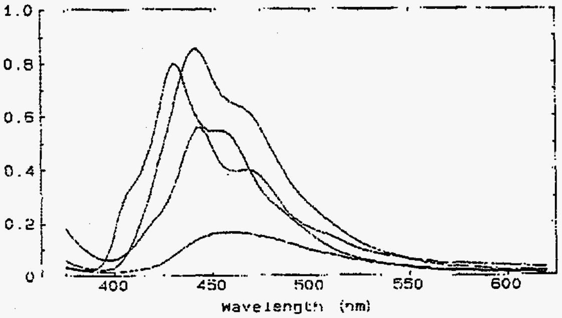

Figure 10-10. Fluorescence spectra for four candidate tagging materials, excitation at 254 nm.

To adequately provide the necessary forensic information for the range of materials and applications, the analytical-grade instruments will be designed to scan the UV, visible, and NIR spectral regions and have the capabilities to collect fluorescence spectra from solutions and solid surfaces. Typical instruments needed for the tagging system are commercially available from a variety of U.S. and international analytical instrumentation vendors at costs ranging from $15K to $30K. Detection of fluorescence from solutions can be accomplished at ppm concentrations using standard methods and equipment. Most analytical-grade instruments are modifiable to use fiber optic probes for surface reflectance measurements. Depending upon the spectral region needed for some of the candidate NIR fluorescence compounds, a separate NIR emission reflectance instrument may be necessary.

11. SELECTION OF CANDIDATE RADIOISOTOPE TAG DETECTION SYSTEMS

The radioisotope detection system includes the hardware to detect the applied tags, to validate the application of the tag, and to verify the identification of unadulterated tags. The radioisotopic tagging process involves depositing a series of small marks (three or more) on the SNM with each mark containing a different gamma emitting radioisotope. Each facility handling SNM will have a unique marking sequence of radioisotopes specifically to identify their site. Because each radioisotope has an independent gamma ray signature, gamma-ray detectors can identify each radioisotope and the sequence of radioisotopes to verify the facility of origin for the tagged SNM.

This section describes the design criteria and preliminary design for the radioisotopic detection systems that will be used for the tagging system. Also included are some preliminary test results which demonstrate the capabilities of the detection system to identify a radioisotope tag.

11.1 Design Criteria for Radioisotope Tag Detection System

11.1.1 Safety

11.1.1.1 Personnel Radiation Exposure. The configuration of the radioisotope detection system will minimize personnel exposure to the ionizing radiation characteristic of the materials being tagged. This will be accomplished by maximizing the distance and barriers between operators and tagged materials.

11.1.1.2 Facility Hazards. The detection system will not introduce physical hazards (i.e., energy source, neutron moderator) beyond the design specifications of the normal SNM working environment.

11.1.2 Security

Because of the nature of the material being tagged, the system will be designed to not report any size, shape, or isotopic composition of the tagged material.

11.1.3 Configuration

Based on the potential uses of the radioisotope tags, there will be two types of radioisotope detector configurations. These configurations include systems for: 1) a high resolution detector for absolute verification of the tag's composition; 2) a low resolution detector for rapid verification of the tag's application or process monitoring. Both systems will be designed to be portable for relative ease of use in a variety of locations at a processing or storage facility.

11.1.4 Spectral Regions

To ensure that the maximum number of radioisotopes available for use as tags is not limited by the systems' hardware, the radioisotope detection systems will have the resolution and sensitivity to detect gamma rays with energies greater than 100 keV for the candidate radioisotopes.

11.1.5 Resolution

11.1.5.1 Resolution Between Isotopes. The detection system will have gamma ray resolution capabilities of <2.5 keV (at 1332 keV) for the high resolution detector and <10% (at 662 keV) for the low resolution detector.

11.1.5.2 Resolution Between Tag and SNM. The detection system will have the capabilities to resolve >1% of an unadulterated tag against the background of 5 kg of SNM.

11.1.6 Detection Surfaces

The radioisotope detection system will resolve the radioisotope pattern of the tagged material from a flat or rounded metal surface. Even though there will be no pattern on tagged powders, the detection systems will identify the presence and composition of the radioisotope tagging material.

11.1.7 Sensitivity

11.1.7.1 Unadulterated Tag. To minimize the cost and the radiation exposure resulting from the radioisotope tag, the detection system will detect as little as 10 nCi of the radioisotope directly from the surface of the tagged material.

11.1.7.2 Tagged Material in Storage. To minimize the direct handling of stored material, the detection system will detect an unadulterated tag through 0.6 cm of stainless steel from a distance of 25 cm with a count time of 15 minutes.

11.1.7.3 Adulterated Tag. The radioisotope detection system will identify a tag that has been through a 99.9% removal effort. For this type of detection, the system will be designed for counting the tagged object directly (no storage container) for a maximum of 600 minutes.

11.1.8 Availability to Users

To ensure that all potential users of the tagging system can obtain the radioisotope detection systems, no component in the detection systems will be limited by export restrictions.

11.2 Radioisotope Detection Systems

Detection of the radioisotope tag on the SNM or on the inside of a storage container will be accomplished by using a high resolution Ge detector or low resolution NaI scintillator in combination with a multi-channel analyzer (MCA) and a laptop computer. These components of the detection systems use existing, commercially available technologies.

11.2.1 High Resolution Ge Detection System

The high resolution Ge detection system shown in Figure 11-1 consists of: 1) a portable 25%-efficient Ge detector with a 5-cm diameter, and 5.6-cm thickness, and a 3-liter liquid nitrogen dewar capacity to last 48 hours;

2) a NOMAD MCA with up to 9 hrs of battery life and a fast and high resolution ADC (8K); 3) a MAESTRO data acquisition software compatible with the MCA; and 4) a laptop computer.

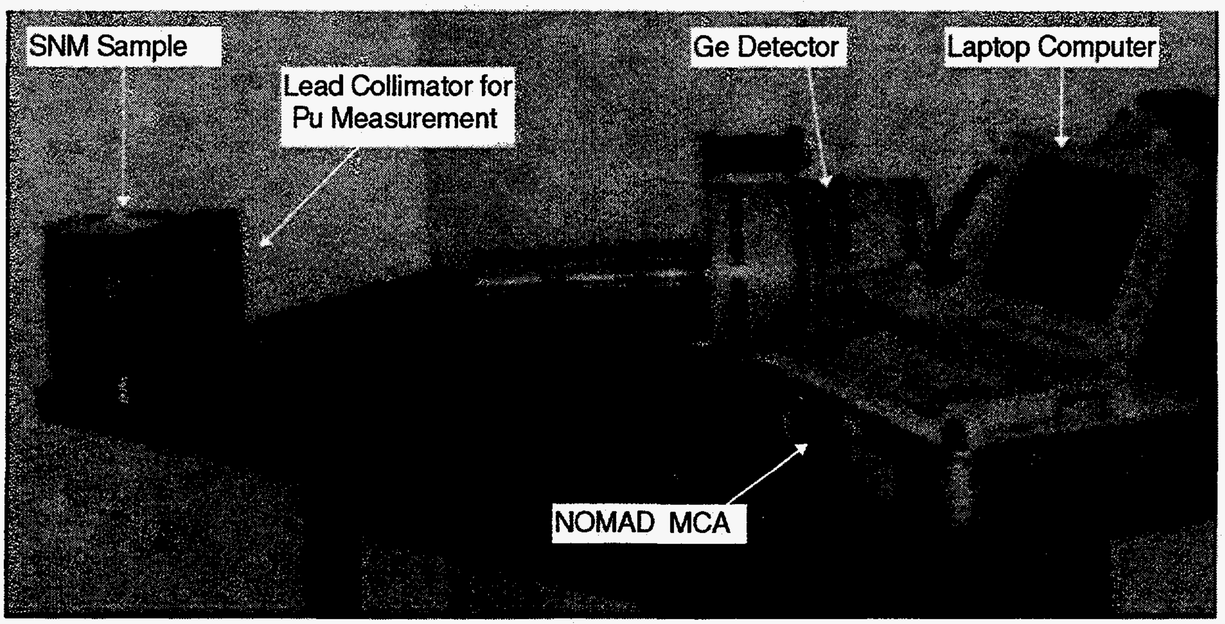

Figure 11-1. Ge high resolution detection system.

11.2.2 Low Resolution NaI Detection System

The low resolution detection system based on an inorganic scintillator (NaI or CsI) shown in Figure 11-2 consists of: 1) a detector with dimensions of 3 inches x 3 inches (no need for liquid nitrogen); 2) a micro NOMAD MCA, with dimensions of 3 inches x 3 inches x 9 inches powered with 8 AA alkaline batteries providing a full 8 hours of field operation; 3) a MAESTRO-LITE data acquisition software compatible with the MCA; and 4) a laptop computer.

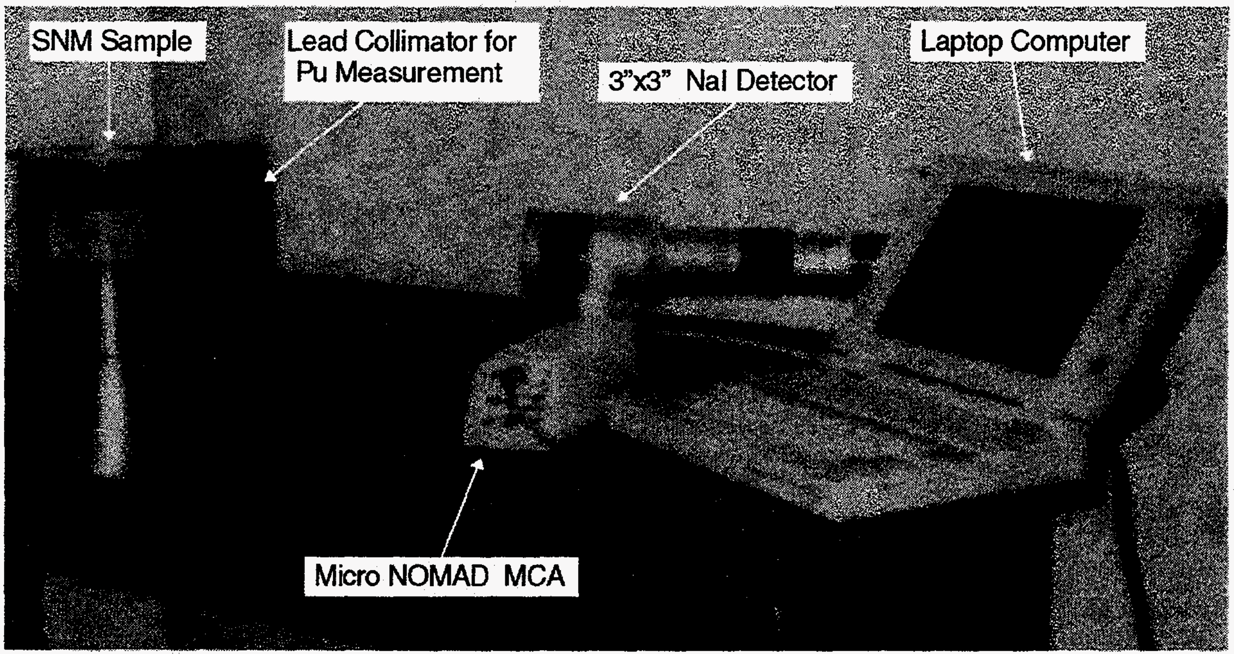

Figure 11-2. NaI detection system.

11.3 Demonstration Measurements with the Detection Systems

Before the radioisotopic tag can be implemented successfully, several detection issues must be resolved. These issues are focused on whether the background radiation from uranium or plutonium is too large to adequately identify tagging radioisotopes on the surface of SNM. To resolve these issues, a set of experiments was completed using various detection systems, radioisotope sources and SNM materials.

11.3.1 Experimental Process

The basic sequence of experimental steps to test and resolve the issues consisted of placing SNM sources 12" from the detectors with various radioisotopes placed in front of the SNM source. Gamma ray spectra were collected for each configuration, with and without the radioisotope source. After the spectra collection, the spectrum with the SNM source was subtracted from the spectrum from the SNM with radioisotope source. Ideally, the resulting subtracted spectrum would only contain the gamma ray signature information from the radioisotope source. A more detailed experimental procedure is shown in Figure 11-3.

The radioactive sources used for various tests are listed in Table 11-1. Among them the ^{57}Co was the weakest source (130 nCi or 0.015 ng) tested, emitting two gamma rays at 122- and 136-keV. Measurements with this radioisotope were used to evaluate the detection systems' sensitivities. If this weak radioisotope source with low energy gamma rays could be detected above the SNM sources, then all other radioisotope sources would be easily detectable. Because the ^{152}Eu source (530 nCi or 3.04 ng) emits several gamma rays between 100-1500 keV, it was used to evaluate the detection systems' capabilities in the presence of a SNM gamma ray field over a wide energy range.

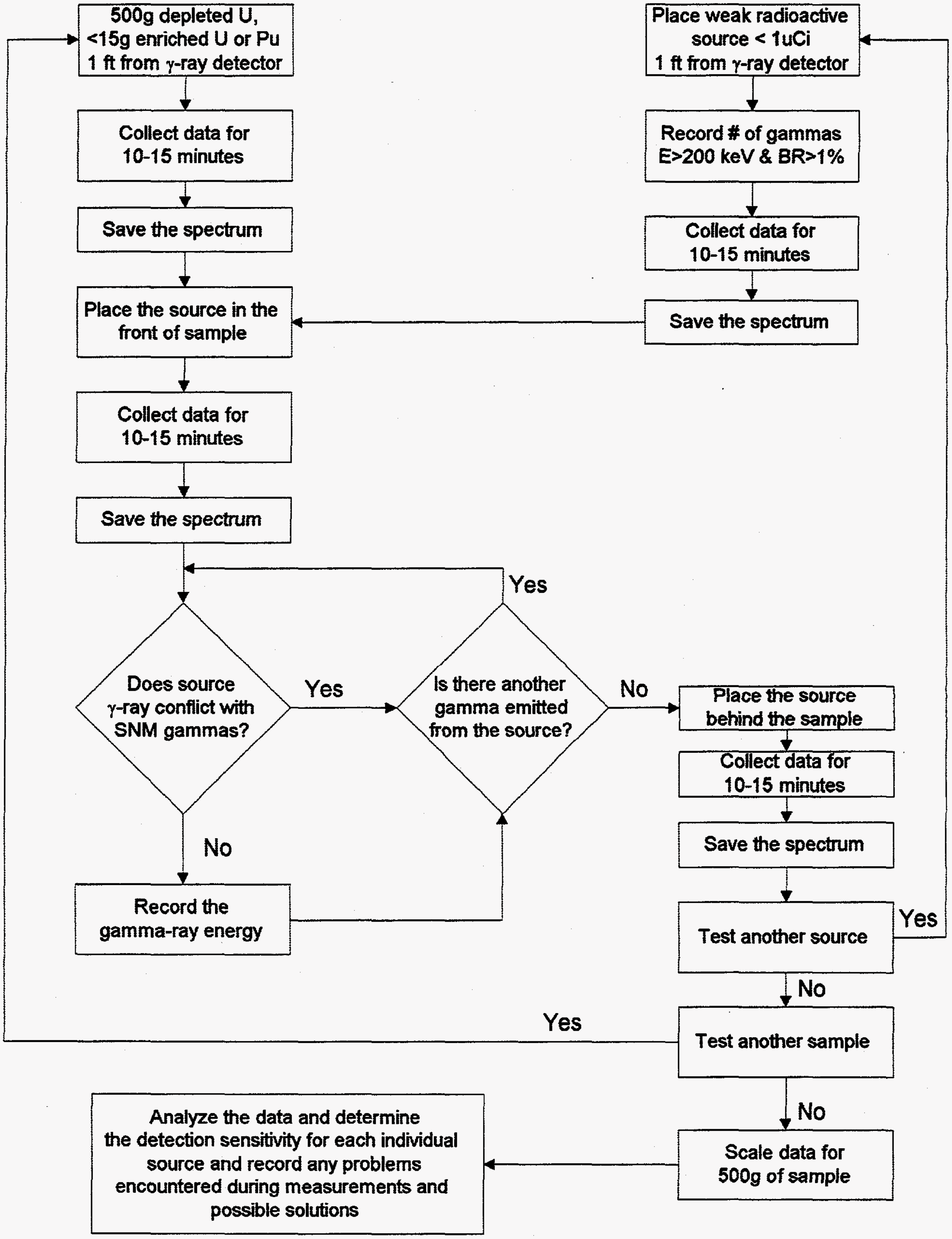

Figure 11-3. Experimental procedure followed to test the detectability of radioisotope tags.

A variety of SNM samples were used to perform the tests. These samples consisted of 500 g of depleted uranium, 47 g of 9% enriched ^{235}U, 25 g of 26% enriched ^{235}U, 25 g of 52% enriched ^{235}U, 9 g of 89% enriched ^{235}U, and 14 g of a mixture of ^{239}Pu (94%) and ^{240}Pu (6%).

Table 11-1. Radioisotope sources tested for use as a tag.

Radioisotope	Source Strength nCi	 Mass (ng)
^{57}Co	130	0.015
^{60}Co	870	0.70
^{54}Mn	140	0.018
^{133}Ba	900	3.53
^{152}Eu	530	3.04

11.3.2 Experimental Results

The gamma ray information from the radioisotope sources is obtained by subtracting the spectrum of the SNM source alone from the spectrum of both the SNM and radioisotope source, so that the resulting subtracted spectrum only contains information from the radioisotope source. With appropriate software that automatically performs this subtraction, the detection system will meet the important criteria that SNM isotopic information in the SNM is not revealed.

Experimental observations were made during four distinctive sets of laboratory analysis simulations. The first set of observations came from the Ge detector and the SNM uranium sources. The second set came from the NaI detector and the SNM uranium sources and the third set came from the Ge detector and the SNM plutonium source. Finally, the fourth set came from the NaI detector and the SNM plutonium source. Some representative spectra collected in our measurements are shown in Appendix E. The results of data analysis and the interpretation of data are discussed in the following subsections.

11.3.2.1 Uranium Measurements with a Ge Detector. Both depleted uranium (^{238}U) and enriched uranium (^{235}U) have very long half-lives of about ~10^9 years which result in very low gamma-ray backgrounds from the uranium isotopes. Therefore, measurements with the SNM uranium samples did not need the collimators shown in Figure 11-1 and 11-2.

Results of gamma scans collected from 500 g of depleted uranium with ^{152}Eu and ^{57}Co sources using a Ge gamma-ray detector are shown in the plots in Appendix E. The labeled peaks in Figure E-1 are energies in keV of the two gamma rays emitted from ^{235}U/^{238}U and gamma rays emitted from the ^{152}Eu and ^{57}Co sources. Spectra E-1.a and E-1.c are the total spectra of depleted uranium plus the radioisotope sources.

Spectra E-1.b and E-1.d are after the depleted uranium spectra were subtracted. In the latter spectra, only gamma rays from the radioisotope sources are seen, indicating both that the SNM signature can be protected and that the radioisotope tag can be resolved.

The results for the 9 g sample of 89% enriched uranium are presented in Figure E-2 for ^{152}Eu and ^{57}Co. As was the case with depleted uranium, both subtracted spectra show only gamma rays emitted from the radioisotope sources. The same results were observed for other enriched uranium samples.

Because the enriched samples were such small quantities relative to what would be experienced in a processing facility, the original pure spectra for these small sources were scaled to 500 g. Then a ^{152}Eu spectrum was added to the scaled spectra. An evaluation of the summed spectra indicated that the ^{152}Eu gamma-ray peaks remained distinguishable for different enriched uranium samples (Figure E-3). This particular scaling method can be interpreted is a "worst case scenario" since gamma ray fields do not scale directly with mass. In other words, the effect of the SNM uranium samples probably is overestimated.

11.3.2.2 Uranium Measurements with a NaI Detector. The above experiments were repeated using a low resolution NaI detector. The results for using 500 g of depleted uranium in combination with ^{152}Eu and ^{57}Co radioisotopes are shown in Figure E-4. Although the gamma rays emitted from the sources are barely distinguishable from the gamma rays of the depleted uranium in spectra E-4.a and E-4.c, they can be easily identified in the subtracted spectra of E-4.b and E-4.d, respectively. The low energy Compton edge, as well as gamma rays emitted from uranium, have completely disappeared in the latter spectra. These results indicate that the NaI detector can be used to identify the tag as long as each radioisotope has at least one gamma ray that is distinct from the other radioisotopes.

11.3.2.3 Plutonium Measurements with a Ge Detector. Measurements for plutonium samples were done using 14 g of plutonium (consisting of 14 NAD's, each containing a 1 g mixture of 94% ^{239}Pu and 6% ^{240}Pu). Since plutonium isotopes have shorter half-lives than uranium isotopes (about 10^4 years as compared to 10^9 years), plutonium isotopes produce a larger gamma ray field, gram for gram, when compared to the SNM uranium isotopes. In addition to the plutonium gamma rays, there is a significant amount of ^{241}Am present in any SNM plutonium sample. This americium isotope has a very short half-life of 14 years, decays predominately by 60 keV gamma ray, and accounts for more than 70% of the total gamma decay of SNM plutonium.

Without an absorber (gamma ray filter) in front of the detector, the gamma ray field from the SNM and its americium impurity will increase the detector dead time (time the detector is not available to record a decay) to 70% or higher and produce a pileup spectrum. To overcome this problem, thin sheets of Cu and Cd were placed in front of the detector. The combination of the cadmium and copper sheets works as gamma ray filter by using the cadmium to absorb the americium gamma ray. Upon absorbing the gamma ray, the cadmium giving off resulting x-rays which are then absorbed by the copper. The metal sheet filters reduced the 60 keV gamma ray intensity by 90% and effectively improve the dead time down to 5.5% for the NaI detector and down to 4% for the Ge detector.

In addition to using an absorber to attenuate the 60 keV gamma ray, a lead collimator was placed between the detector and the SNM Pu sample. The purpose of the collimator was to reduce the amount of Pu surface area viewed by the detector while maintaining the maximum view of the radioisotope sources. As each test was conducted, two collimators were placed in front of the SNM/radioisotope sources. Three types of spectra were taken: one spectrum without collimation, one spectrum with a 2" Pb brick that had a 1" hole, and one spectrum with the 1/4" hole in the Pb brick. The position of the Pb collimator can be seen in Figures 11-1 and 11-2.

The results of tests conducted on the 14 g of the SNM plutonium sample with the ^{152}Eu and ^{57}Co sources using the Ge gamma-ray detector are shown in Figure E-5. The peaks in the ^{152}Eu and ^{57}Co sources, as well as some peaks in the SNM sample, are labeled according to their gamma-ray energies in keV. Spectra E-5.a and E-5.c are the total spectra of the plutonium plus the radioisotope sources. Spectra E-5.b and E-5.d are the totals after the SNM plutonium spectra were subtracted. As expected, in the latter spectra only gamma rays from radioisotope sources are seen and all the other gamma rays from the SNM, including the Compton background at a low and high energy regions, have disappeared. One of the important aspects of the Pu measurements is that even with a huge Compton background below 500 keV, the 122 keV gamma ray emitted from a very weak ^{57}Co source is detectable.

***11.3.2.4 Plutonium Measurements with a NaI Detector*.** The results for measurements conducted with a low resolution NaI detector using a 1/4" collimator are shown in Figure E-6. None of the gamma rays emitted from the ^{152}Eu and ^{57}Co sources can be identified in spectra E-6.a and E-6.c. However, the peaks can be seen in the subtracted spectra E-6.b and E-6.d. These results signify that the NaI detector can also be used, in some cases, to identify the tag on a SNM plutonium source.

11.3.2.5 SNM Surface Area Effects on the Detection System. The effect of the SNM surface area exposed to the detection system was evaluated using the SNM plutonium source. These tests revealed that the detector's count rate is dependent on the surface area of the source. To quantify this effect, the 14 g of plutonium were arranged so that the NaI and Ge detectors were exposed to the SNM and then to twice as much surface area of the plutonium source. Doubling the exposed surface area increased the SNM-related count rate 1.7-fold and increased the Ge dead time to 7%. For the NaI detector, the count rate increased by 1.5-fold and the dead time increased to 9%. Therefore, the effect of increasing the surface area exposed to the detector indicates that a collimator will be necessary to ensure more sensitive and resolvable spectra when dealing with SNM plutonium sources.

To evaluate an appropriate design for a collimator, several gamma ray measurements were performed using the plutonium source, no collimator, a 1" collimator and 1/4" collimator. The results are illustrated in Figure E-7. As can be seen from these spectra, the effect of collimation allows the ^{152}Eu gamma ray peaks to be better resolved from the background. The best result was obtained with a 1/4" of lead collimator. Similar results were observed for the weak ^{57}Co source. In these tests, the collimator decreased the detectors' dead time to 0.7% and 0.5% for the NaI and Ge detectors, respectively.

11.3.2.6 Pattern Recognition with the Detection System. To establish whether the detection systems' configurations could be used to recognize a radioisotope pattern as part of a tag, four radioisotope sources (^{57}Co, ^{133}Ba, ^{54}Mn, and ^{137}Cs) were placed in order on a depleted uranium sample. The sources were placed sequentially 1/2" from each other in a line on the 500 g sample of depleted SNM uranium. Then, several gamma ray spectra were collected as the Ge detector with a 1/4" collimator was passed across the sample. The results of these testes are summarized in Table 11-2. The four positions listed in Table 11-2 correspond to where a spectrum was taken (each position corresponds to a position above one of the four radioisotope sources). These results demonstrate that the sequence of the sources correspond to the level of responses detected.

Table 11-2. Detected counts observed for various gamma rays at four positions above a uranium source.

Isotope	Energy keV	Number of Counts 1	2	3	4
^{57}Co	122	481	-	-	-
^{133}Ba	302	-	130	-	-
	356	-	366	-	-
	382	-	44	-	-
^{54}Mn	834	-	-	52	-
^{137}Cs	662	-	-	-	1719

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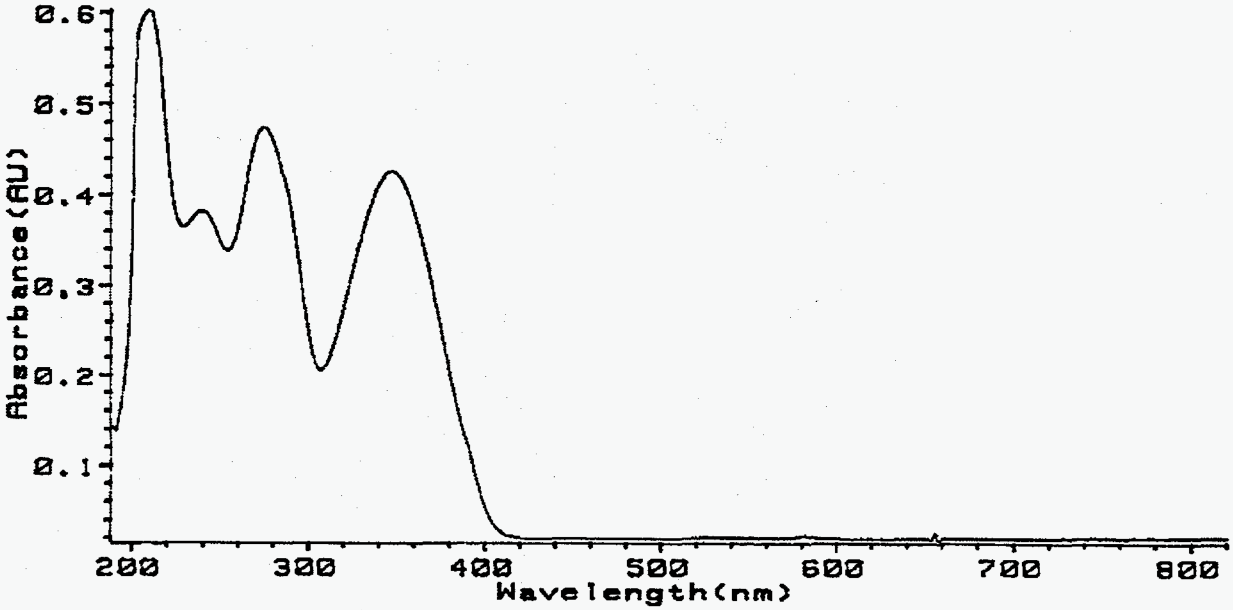
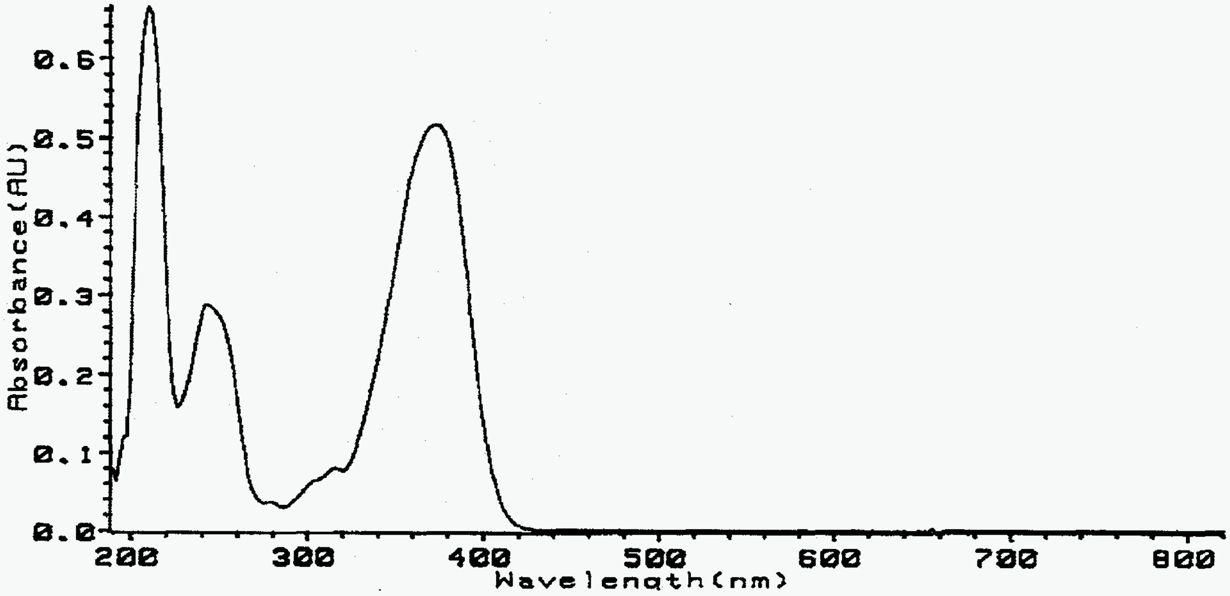
Appendix A
Absorbance Spectra for Candidate Fluorescence Compounds

The following absorbance spectra were obtained using a Hewlett Packard HP8452A diode array spectrophotometer. The candidate fluorescence compounds were dissolved in Fisher electronic grade isopropanol (2- propanol) 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.

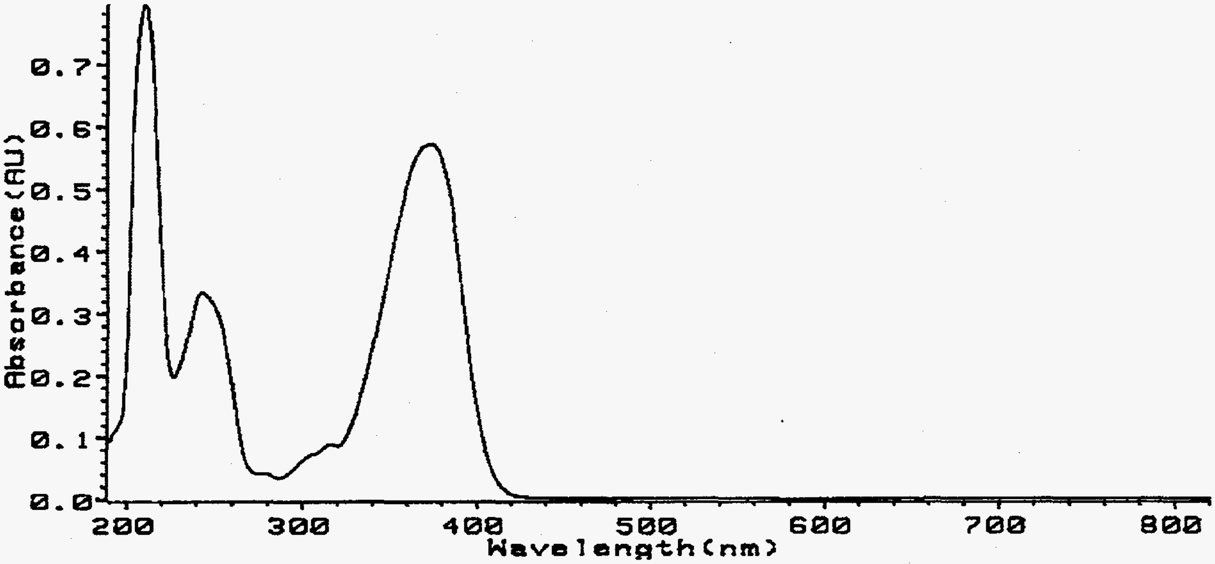
The absorbance spectrum for Day-Glo Color Corporation's UV Blue D-282:



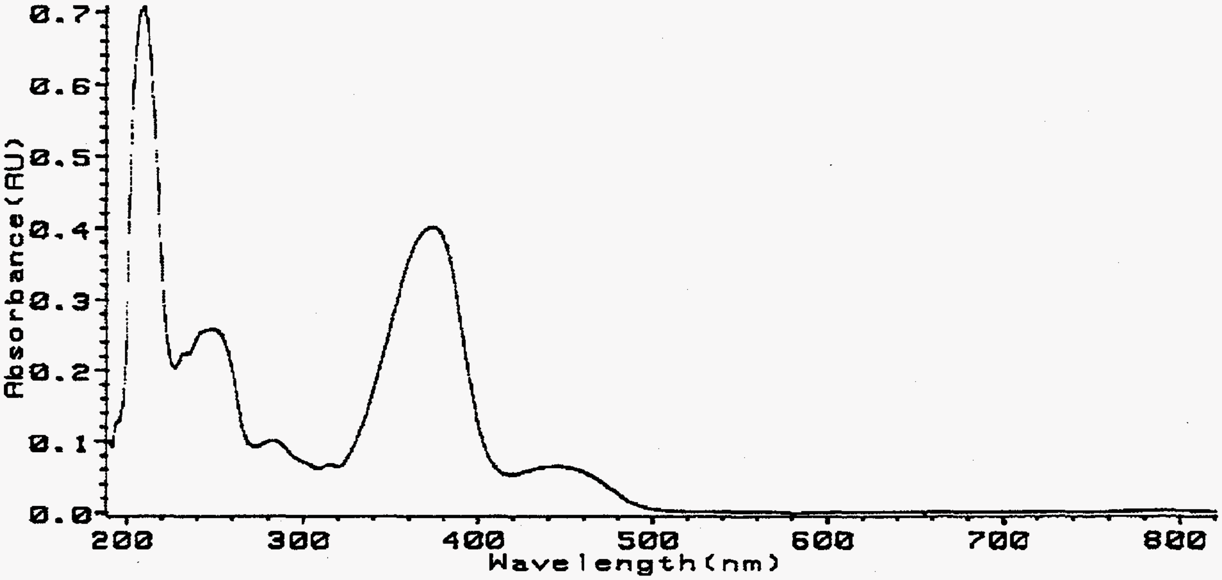
The absorbance spectrum for Day-Glo Color Corporation's Columbia Blue D-298:



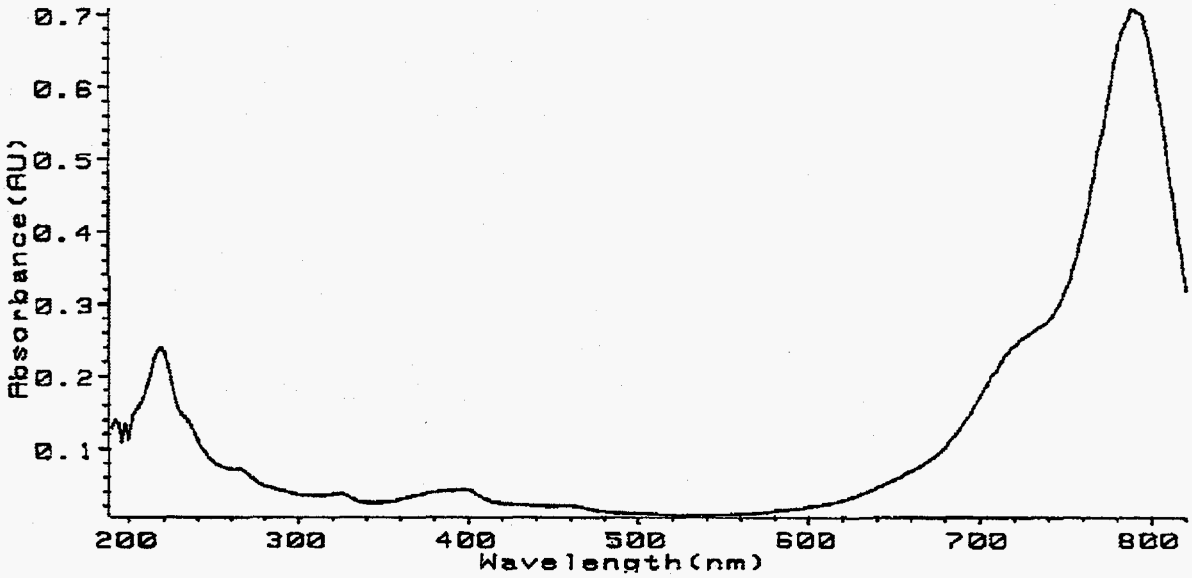
The absorbance spectrum for Sherwin Incorporated DUBL-CHEK W2-D3F:



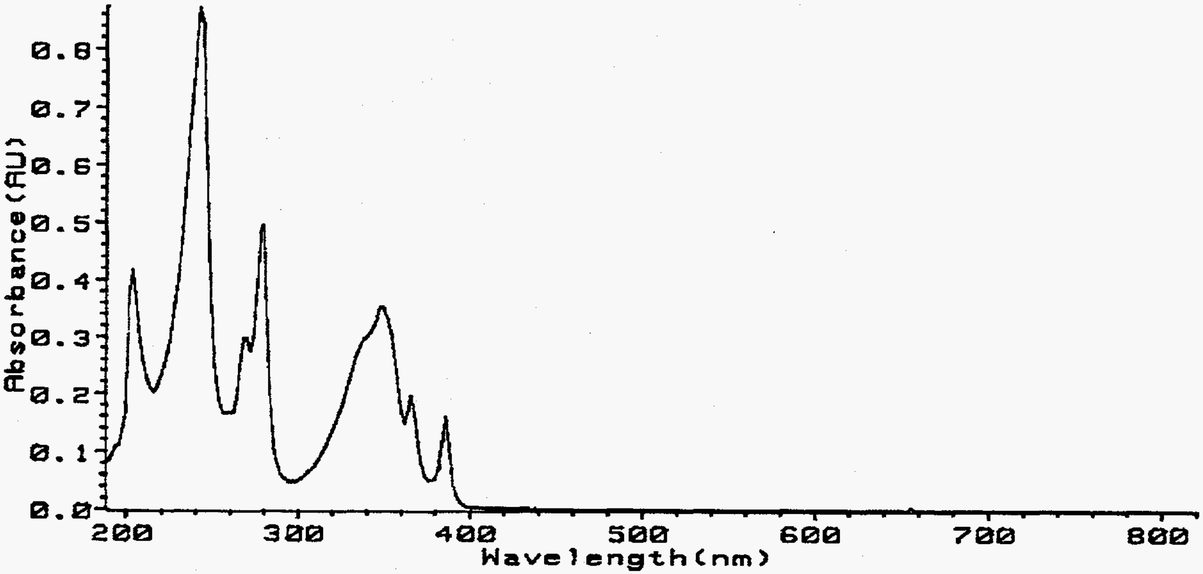
The absorbance spectrum for Sherwin Incorporated DUBL-CHEK HM-704:



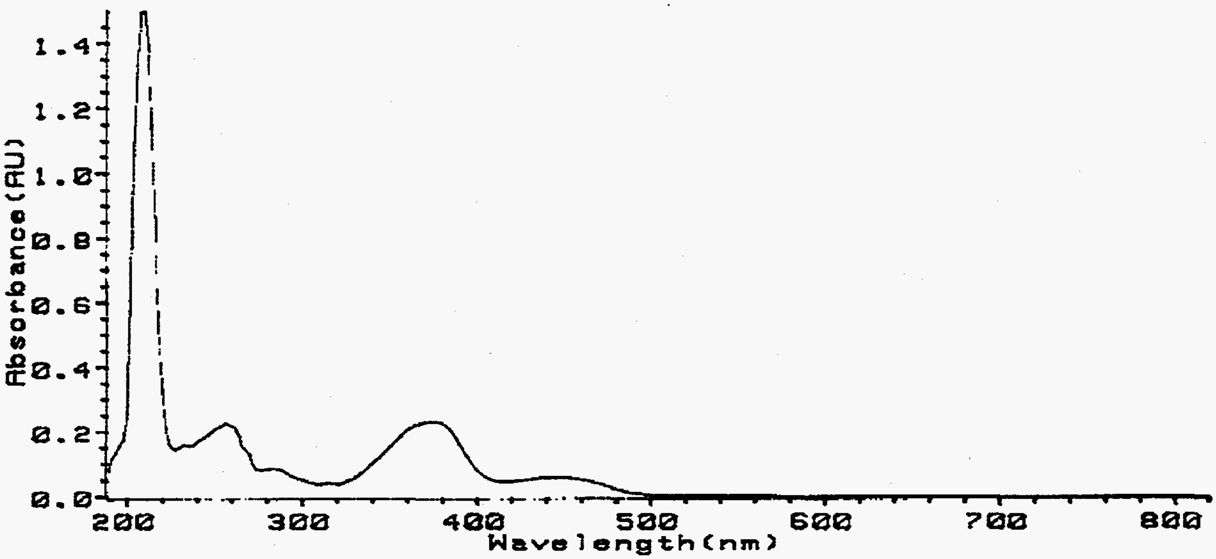
The absorbance spectrum for Exciton Incorporated IR-125:



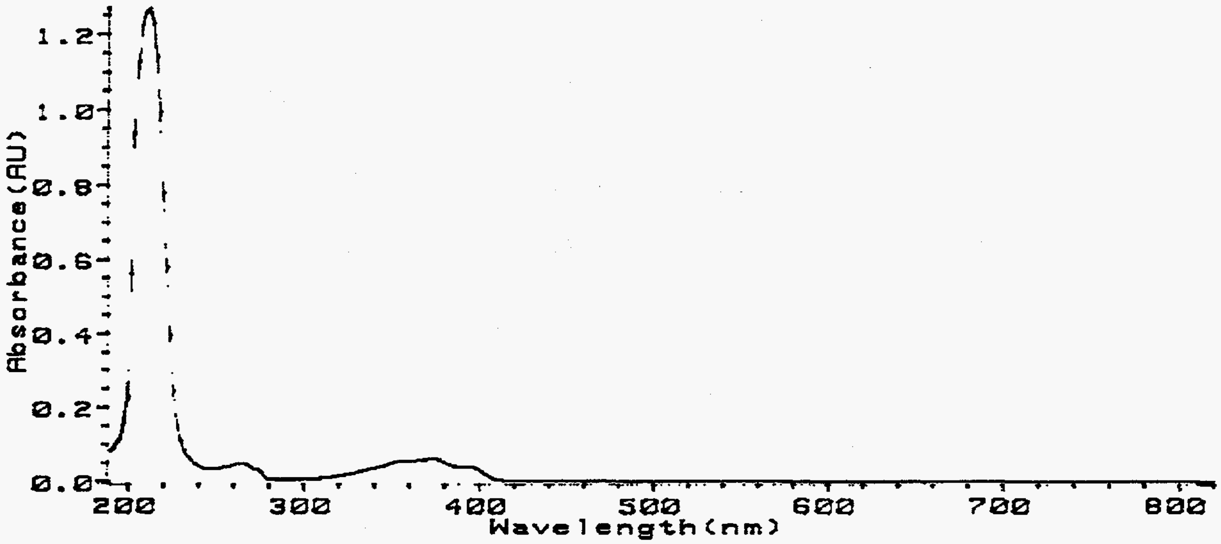
The absorbance spectrum for Molecular Probe Incorporated 1-hydroxypyrene:



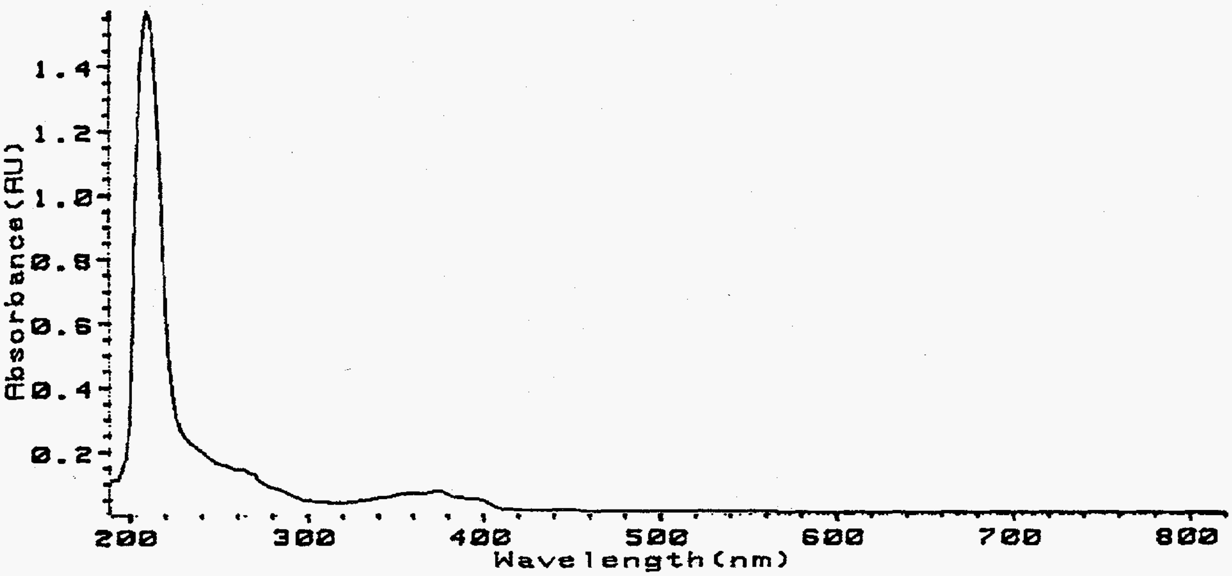
The absorbance spectrum for Magnflux Incorporated SpotCheck ZL-27A:



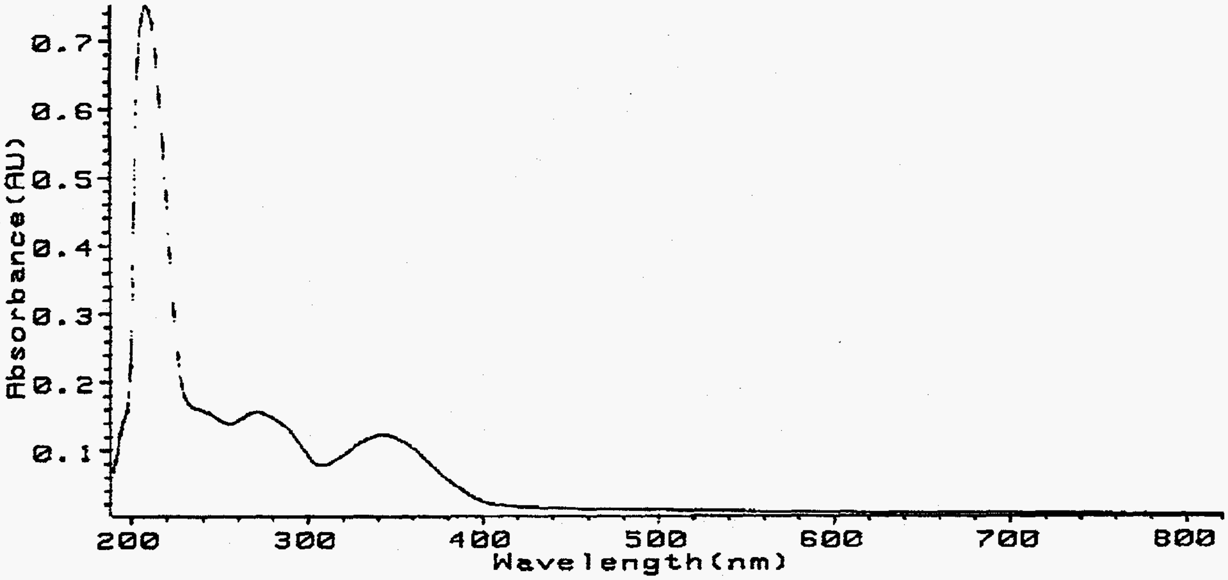
The absorbance spectrum for Shannon Luminous Materials Incorporated I-2120:



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



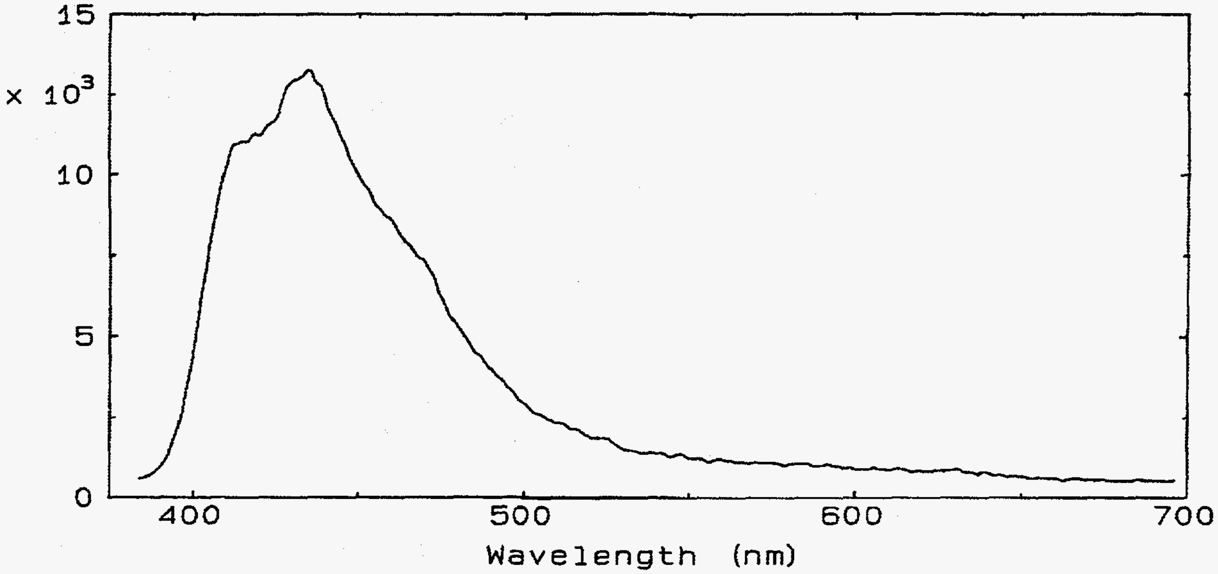
The absorbance spectrum for Spectronics Corporation PMI-B-WB:



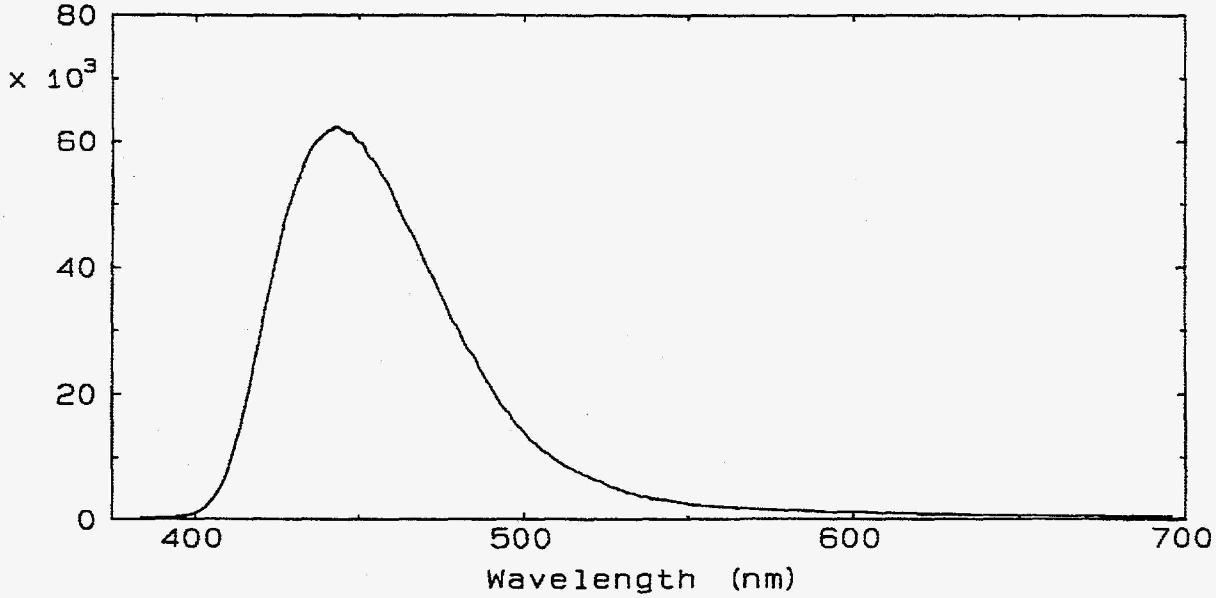
Appendix B
Fluorescence Spectra for Candidate *Fluorescence Compounds*

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.

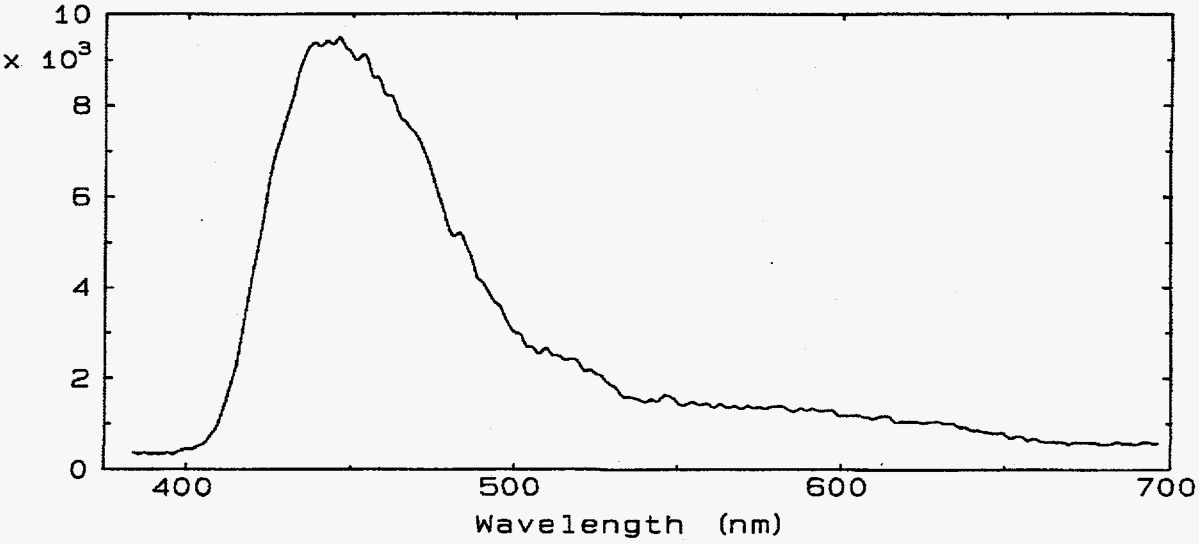
The fluorescence spectrum for Day-Glo Color Corporation's UV Blue D-282:



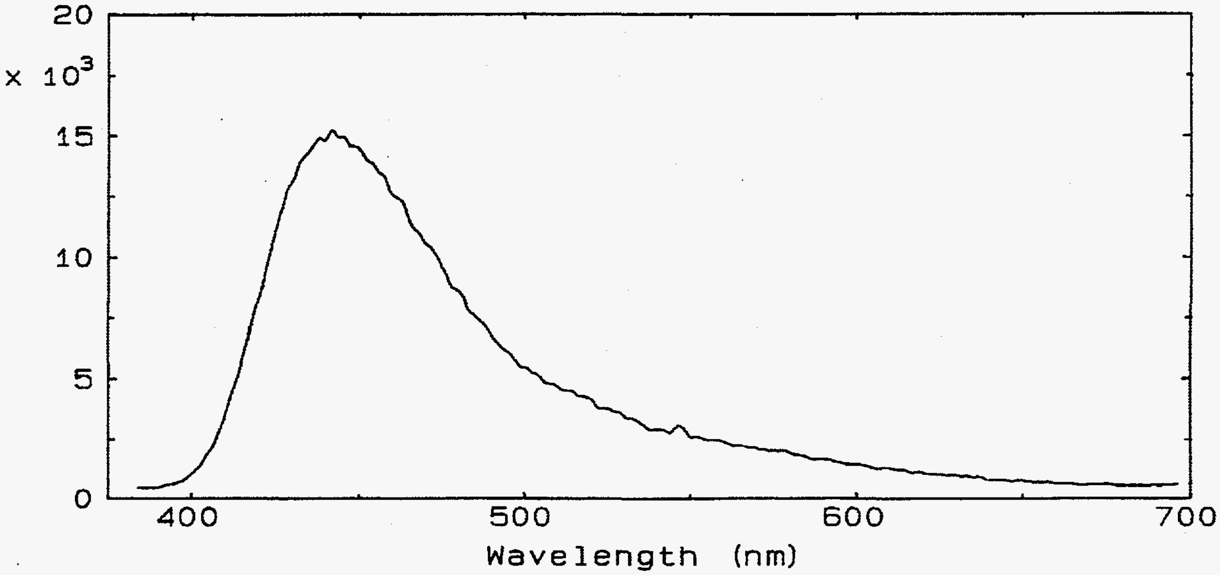
The fluorescence spectrum for Day-Glo Color Corporation's Columbia Blue D-298:



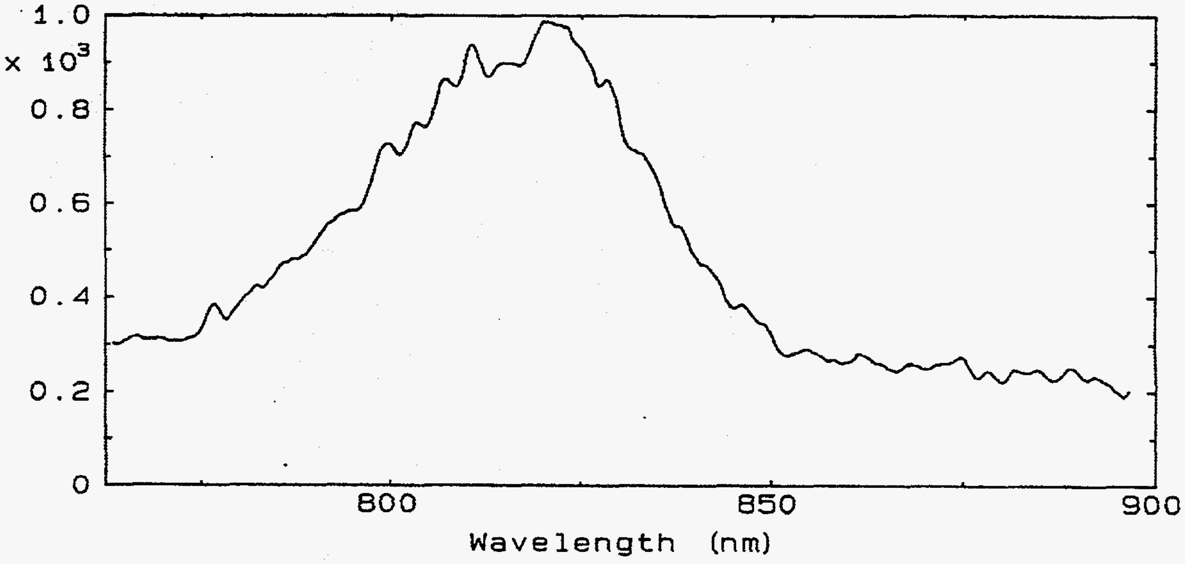
The fluorescence spectrum for Sherwin Incorporated DUBL-CHEK W2-D3F:



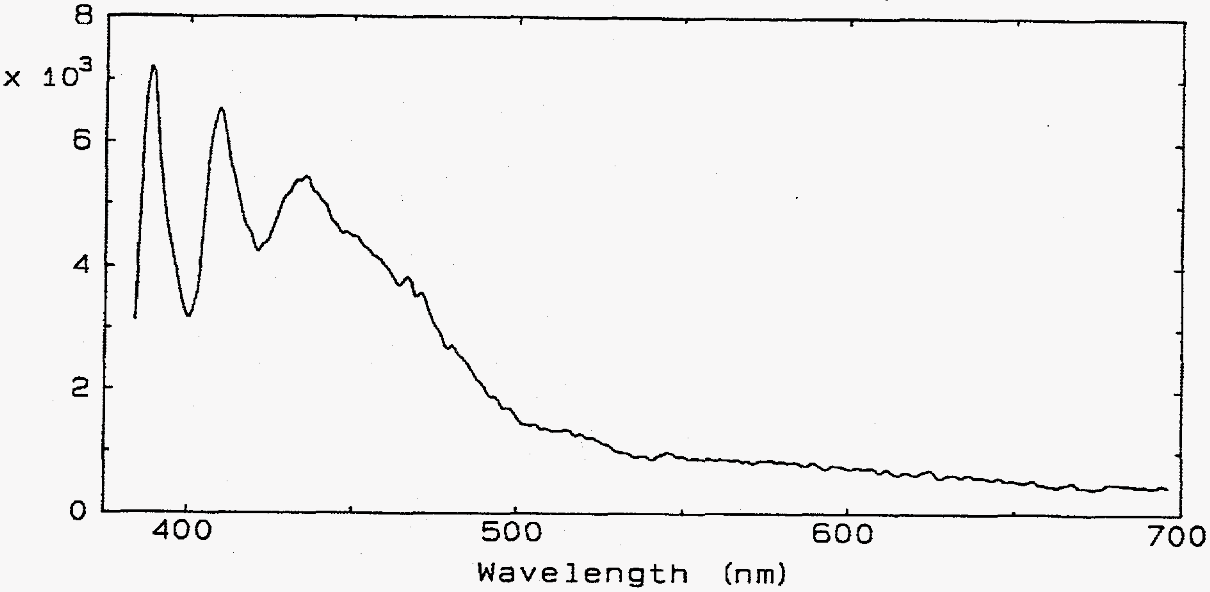
The fluorescence spectrum for Sherwin Incorporated DUBL-CHEK HM-704:



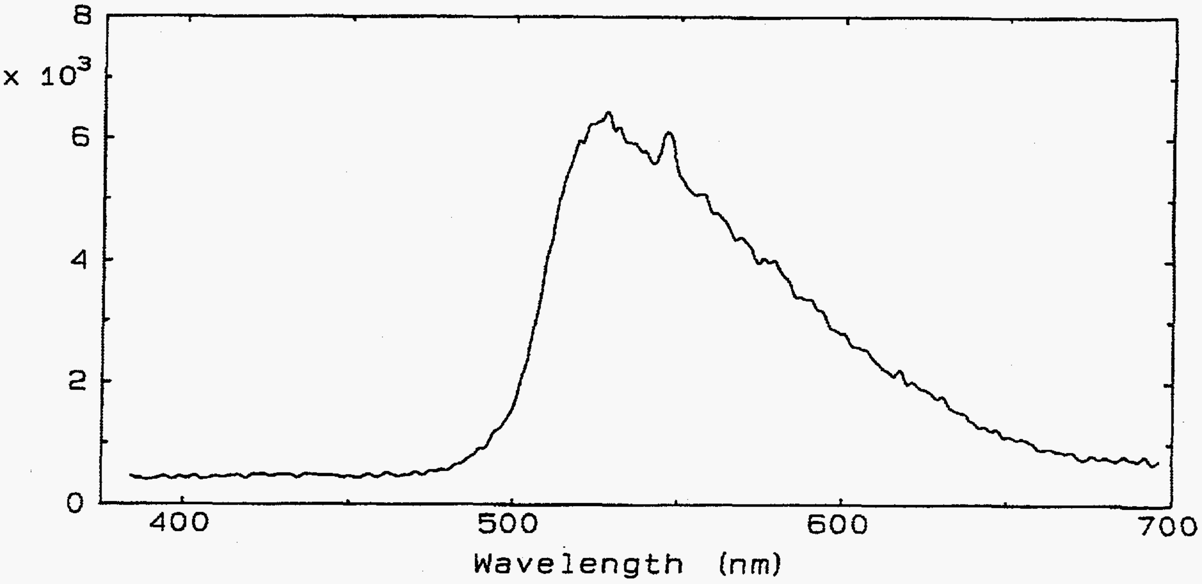
The fluorescence spectrum for Exciton Incorporated IR-125:



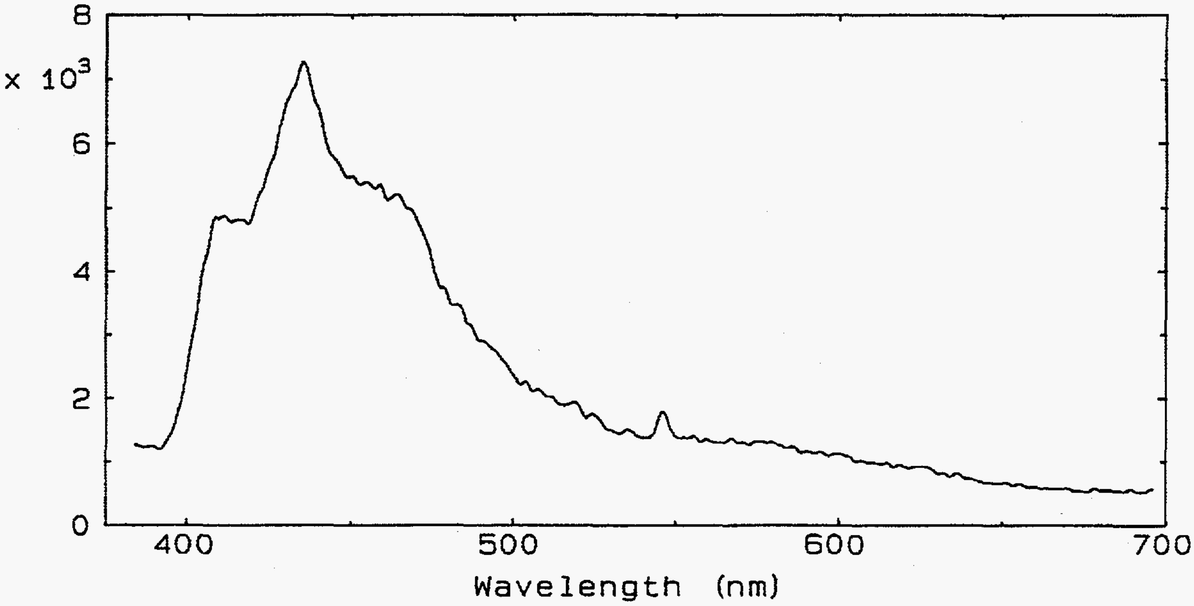
The fluorescence spectrum for Molecular Probe Incorporated 1-hydroxypyrene:



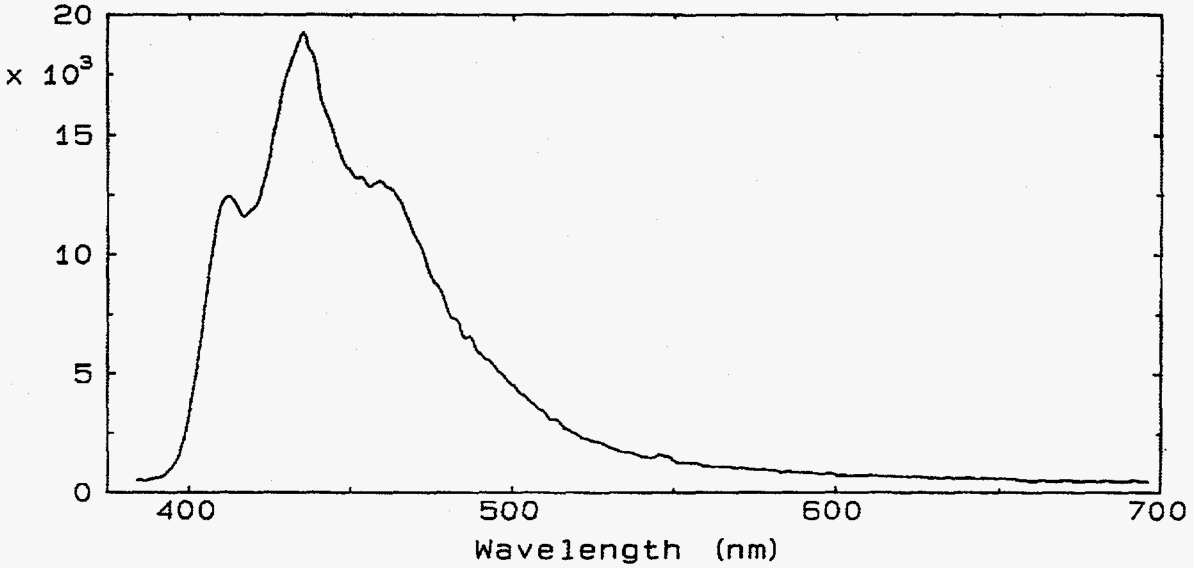
The fluorescence spectrum for Magnaflux Incorporated SpotCheck ZL-27A



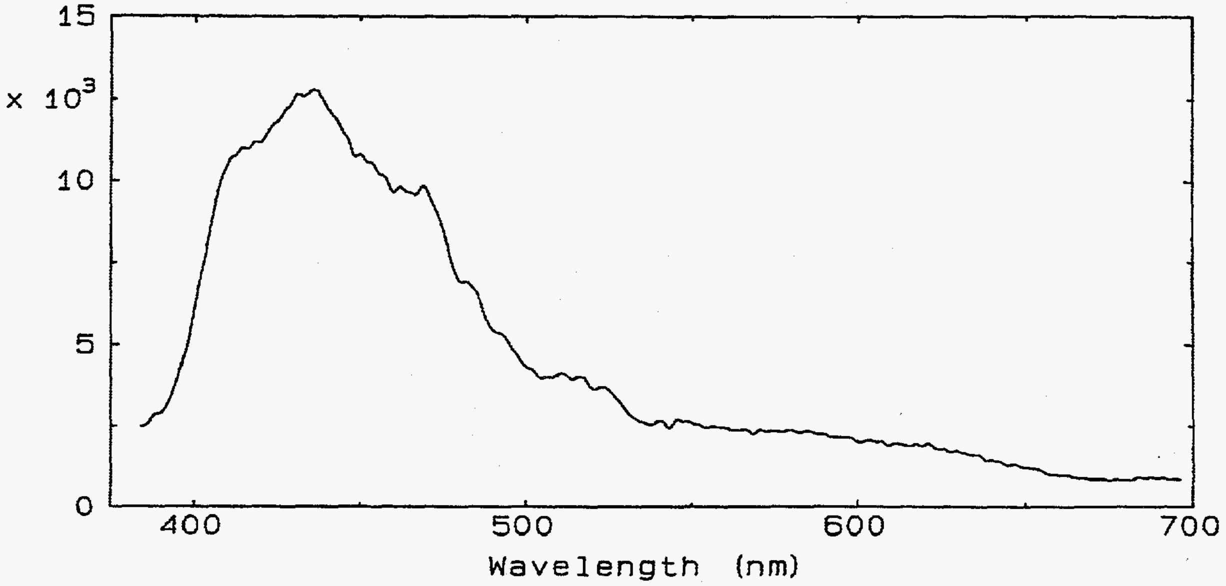
The fluorescence spectrum for Shannon Luminous Materials Incorporated I-2120:



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		Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
			Energy(keV)	Nuclide			
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
Ba-133	276.29	0.07	277.65 277.63	Hf-178n 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
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
Bk-247	265.00	0.30			1380.0	31.440	17.571
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
Cf-251	285.00	0.01			898.04	1.5054	.85968
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	276.29 277.35	Ba-133 Hf-178n	29.100	684.35	387.91
Co-60	1173.23	1.00			5.2700	39478.	30182.
Co-60	1332.51	1.00			5.2700	36081.	28003.
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

Ho-

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			
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
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	276.29 277.63	Ba-133 Cm-243	31.000	125.37	71.044
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
Nb-94	702.50	1.00			20300.	9.3994	6.5821
Nb-94	871.10	1.00	869.23 873.20	Eu-150n Eu-154	20300.	8.0738	5.8931
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
Po- 209	260.50	0.002			102.00	3.9010	2.1681
Po- 209	896.60	0.003			102.00	1.8411	1.3507
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		Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
			Energy(keV)	Nuclide			
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	24.100	457.63	310.18
			602.71	Sb-124			
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	49.510	139.82	98.546
			725.7	Pm-148m			

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			
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		Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
			Energy(keV)	Nuclide			
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
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
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
Te-129m	695.98	0.03			33.600	120.80	84.429
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		Half Life (y)	CPM/100 ng Air	CPM/100ng 1/4 inch Fe
			Energy(keV)	Nuclide			
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	392.49 392.64	Tb-160 Ag-105	83.400	3464.1	2137.4
Zr-95	724.18	0.44	722.78 725.21 725.70	Sb-124 In-114m Pm-148m	64.020	1270.0	894.85
Zr-95	756.72	0.55			64.020	1539.0	1093.9

Appendix E
Gamma Ray Spectra Collected with Radioisotope Detection Systems

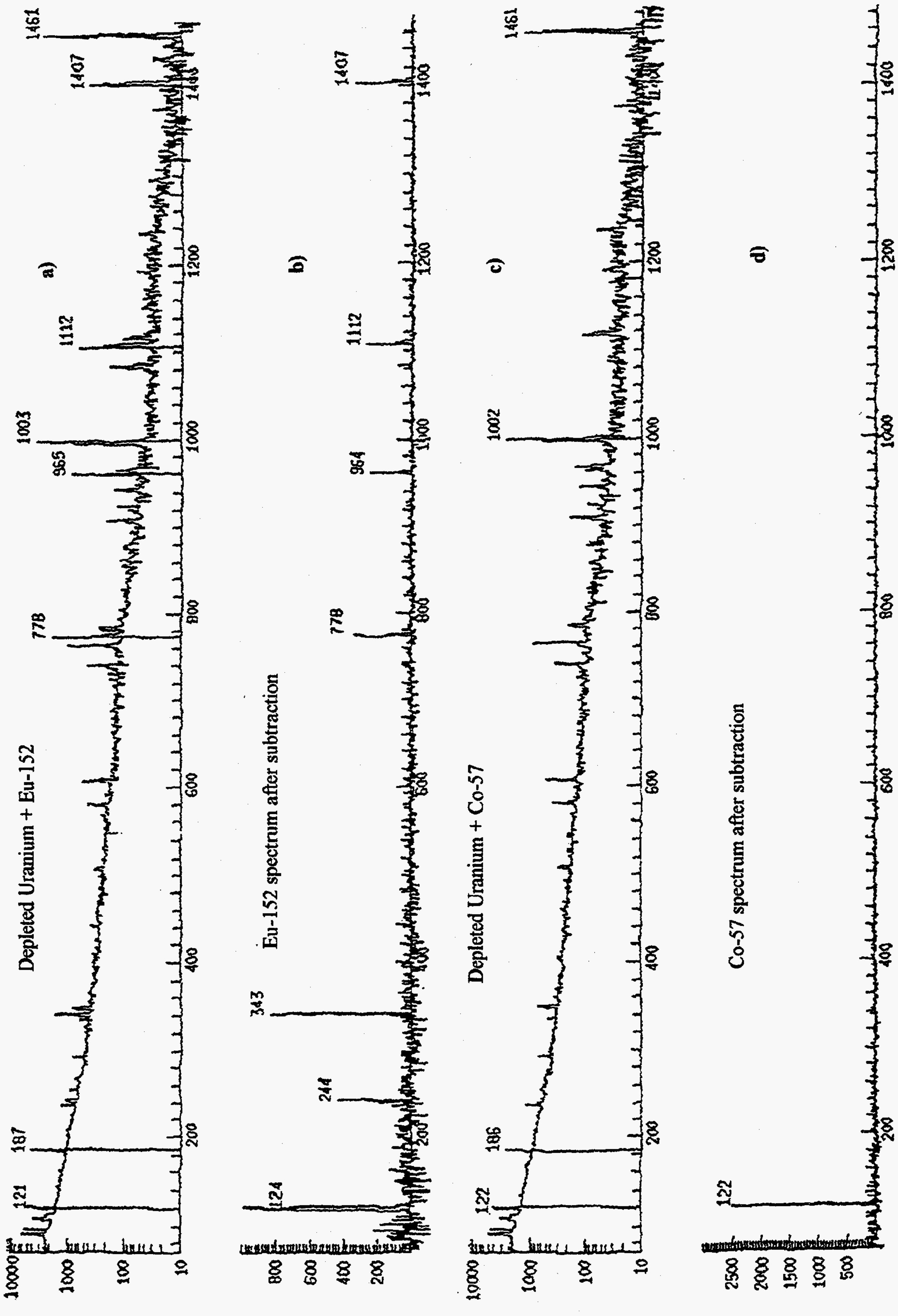


Figure E-1. Depleted Uranium spectrum taken with Eu-152 and Co-57 sources, using a Ge detector.

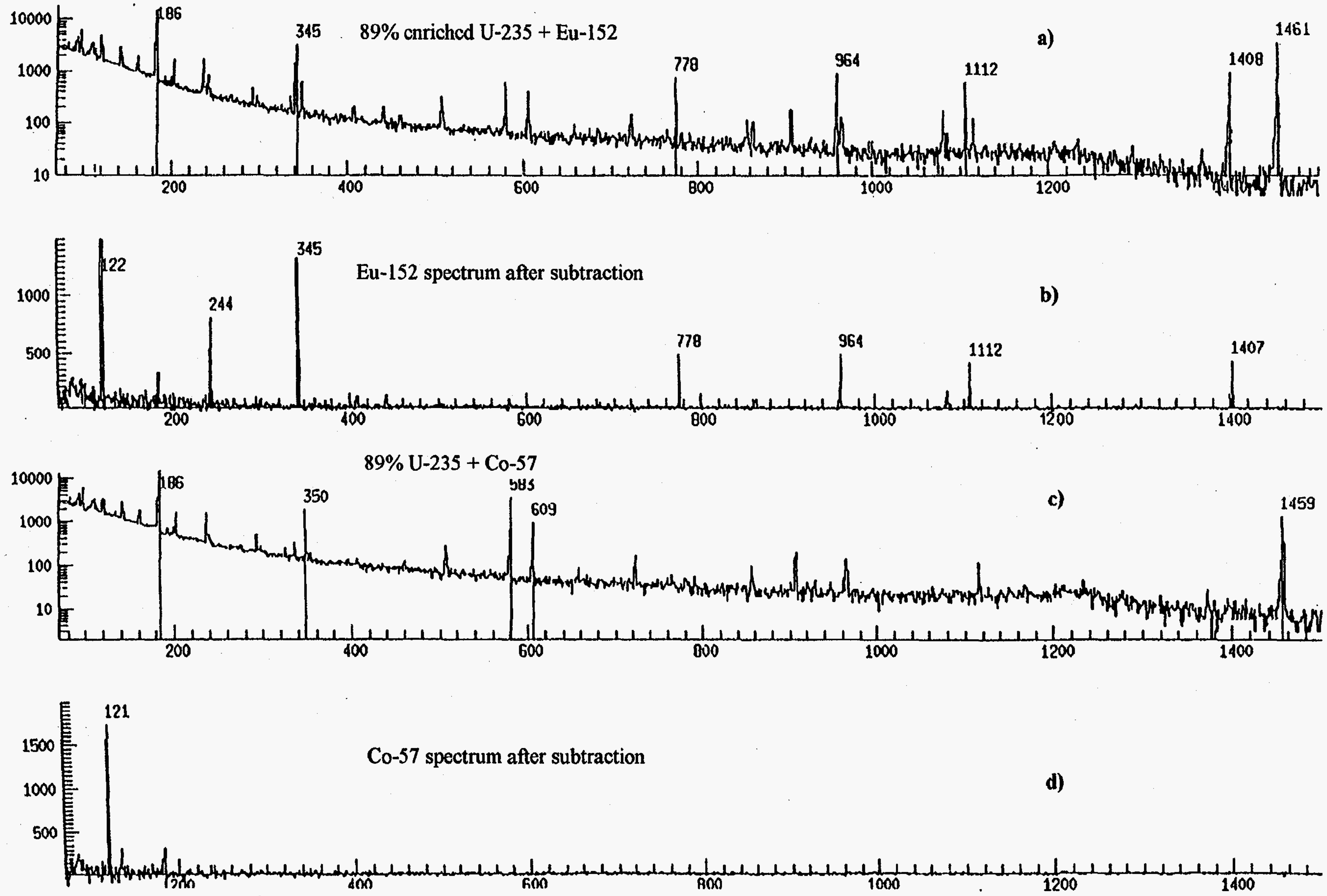


Figure E-2. 89% enriched Uranium spectra taken with Eu-152 and Co-57 sources, using a Ge detector

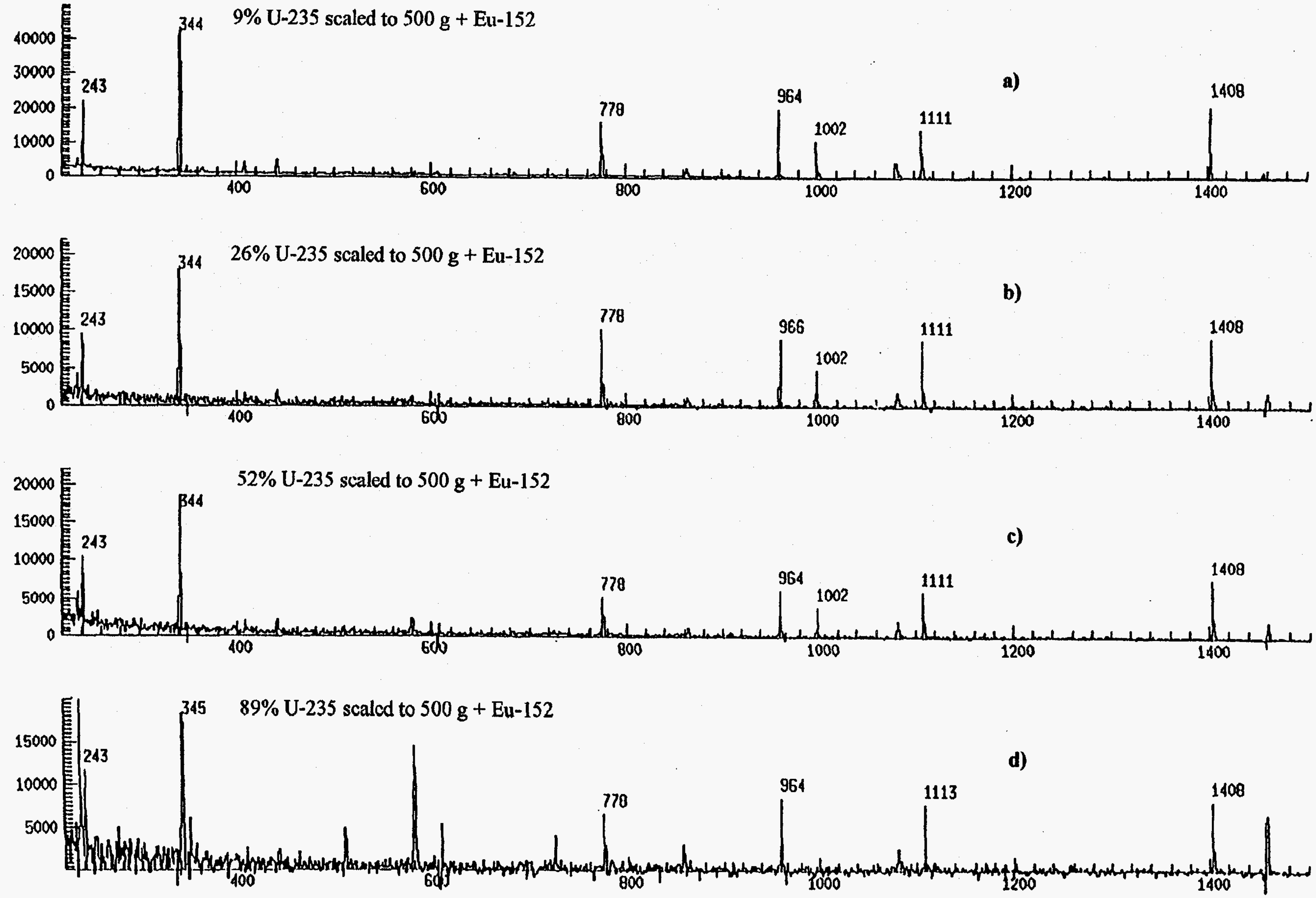
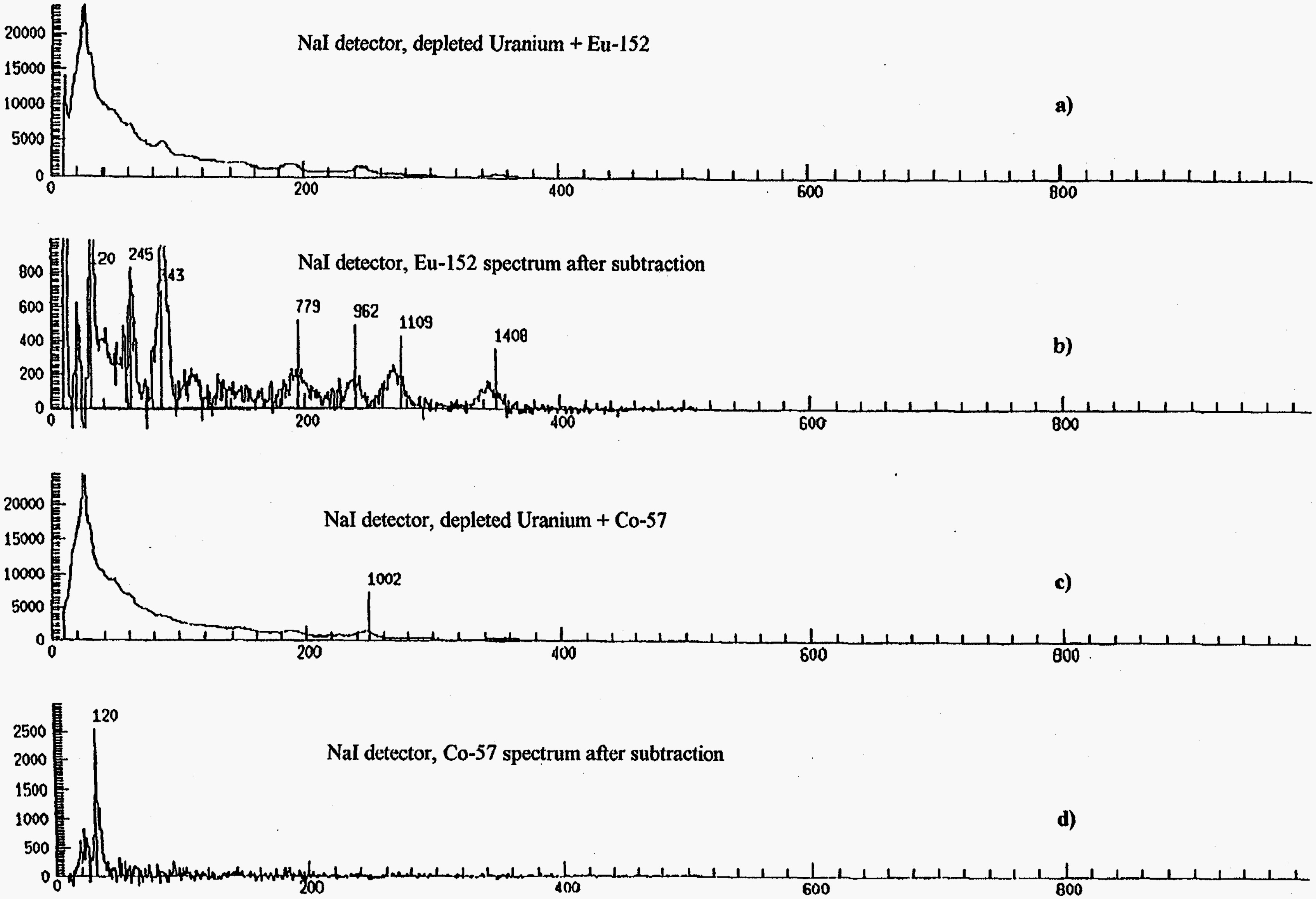


Figure E-3. Eu-152 spectra taken with different enriched Uranium samples that were scaled to 500 g.


NaI detector, depleted Uranium + Eu-152

NaI detector, Eu-152 spectrum after subtraction

NaI detector, depleted Uranium + Co-57

NaI detector, Co-57 spectrum after subtraction

Figure E-4. Depleted Uranium spectra taken with Eu-152 and Co-57 sources, using a NaI detector.

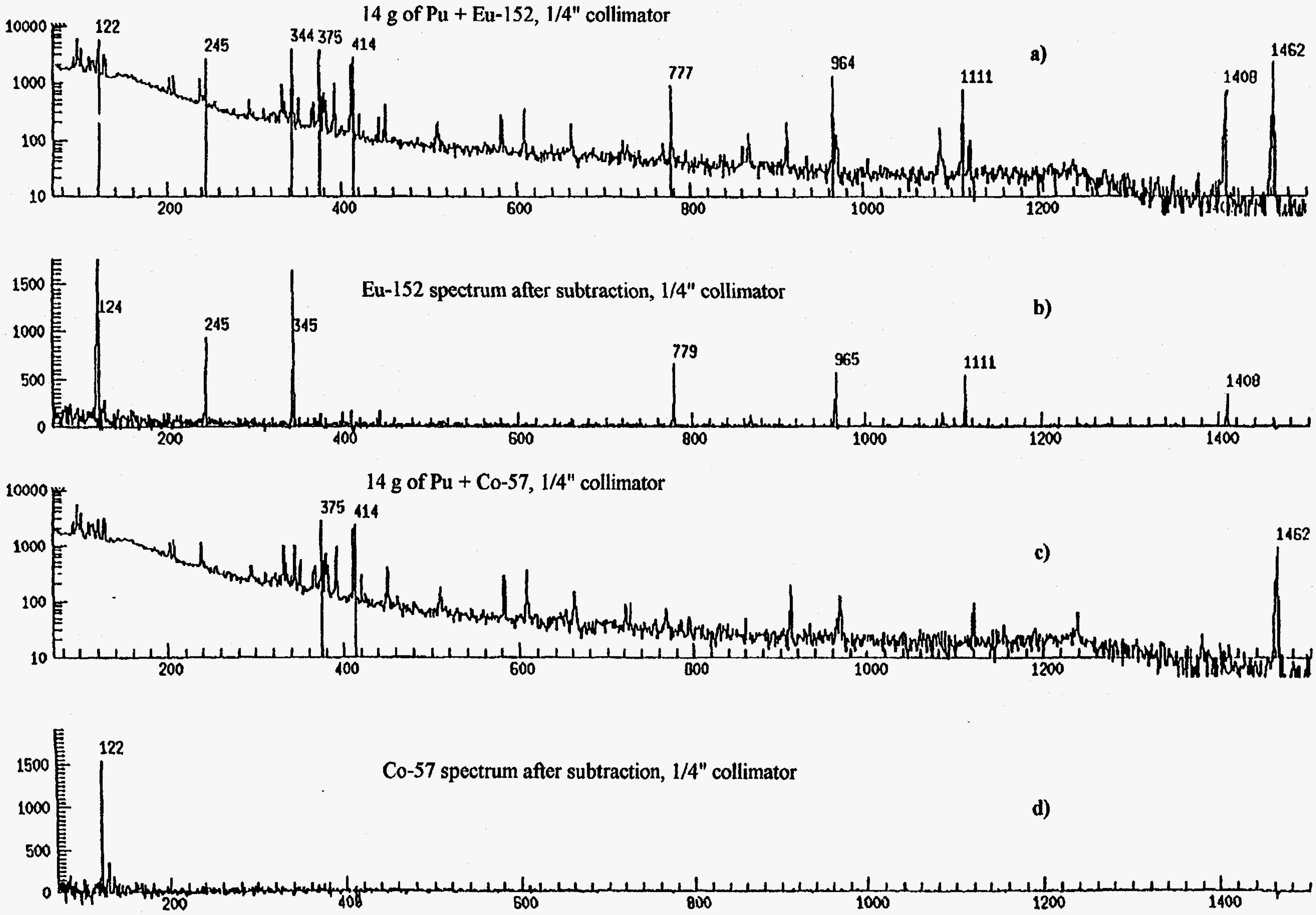


Figure E-5. Spectra obtained with Pu, using Eu-152 and Co-57 sources. A 1/4" collimator was placed between the sources and the Ge detector.

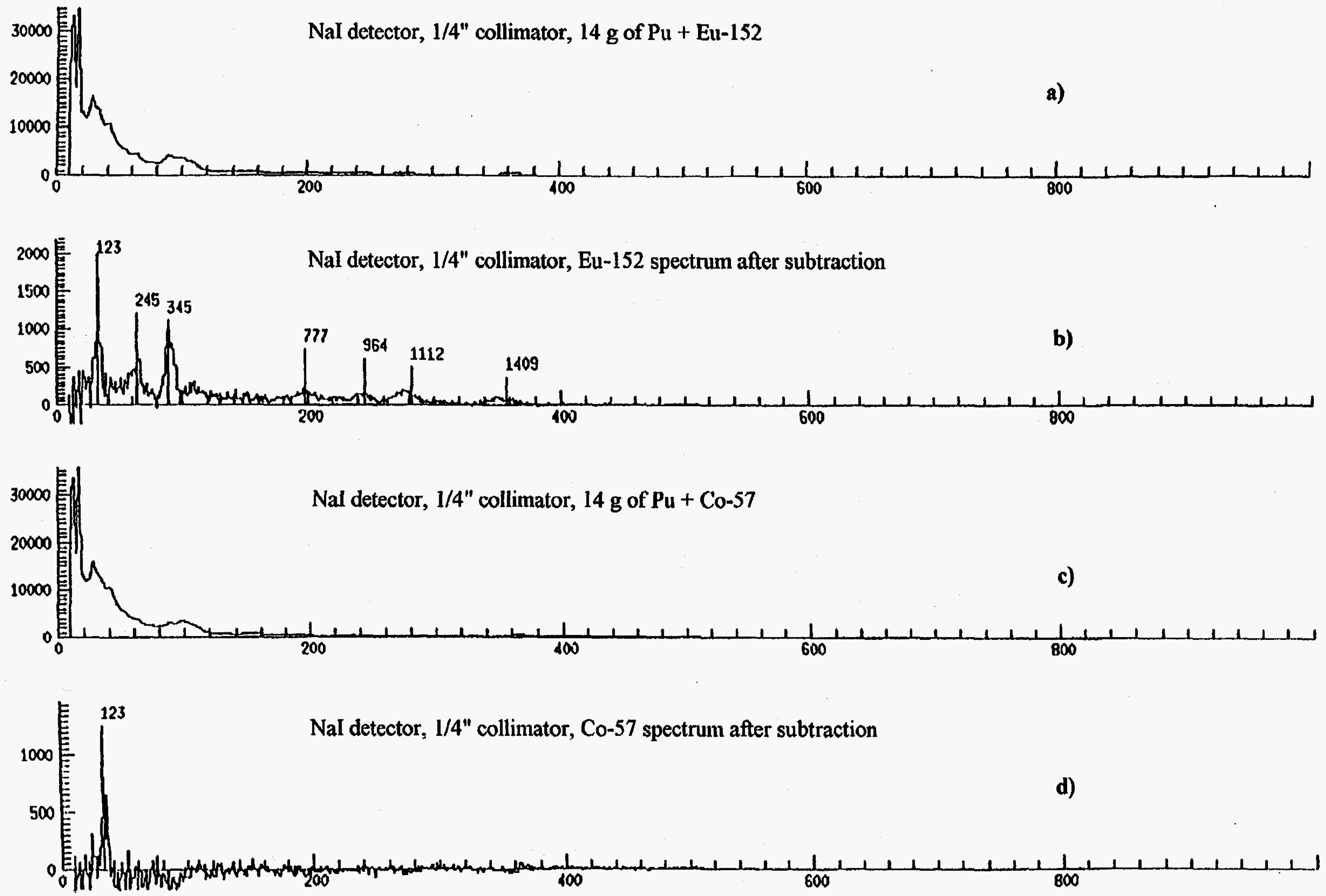
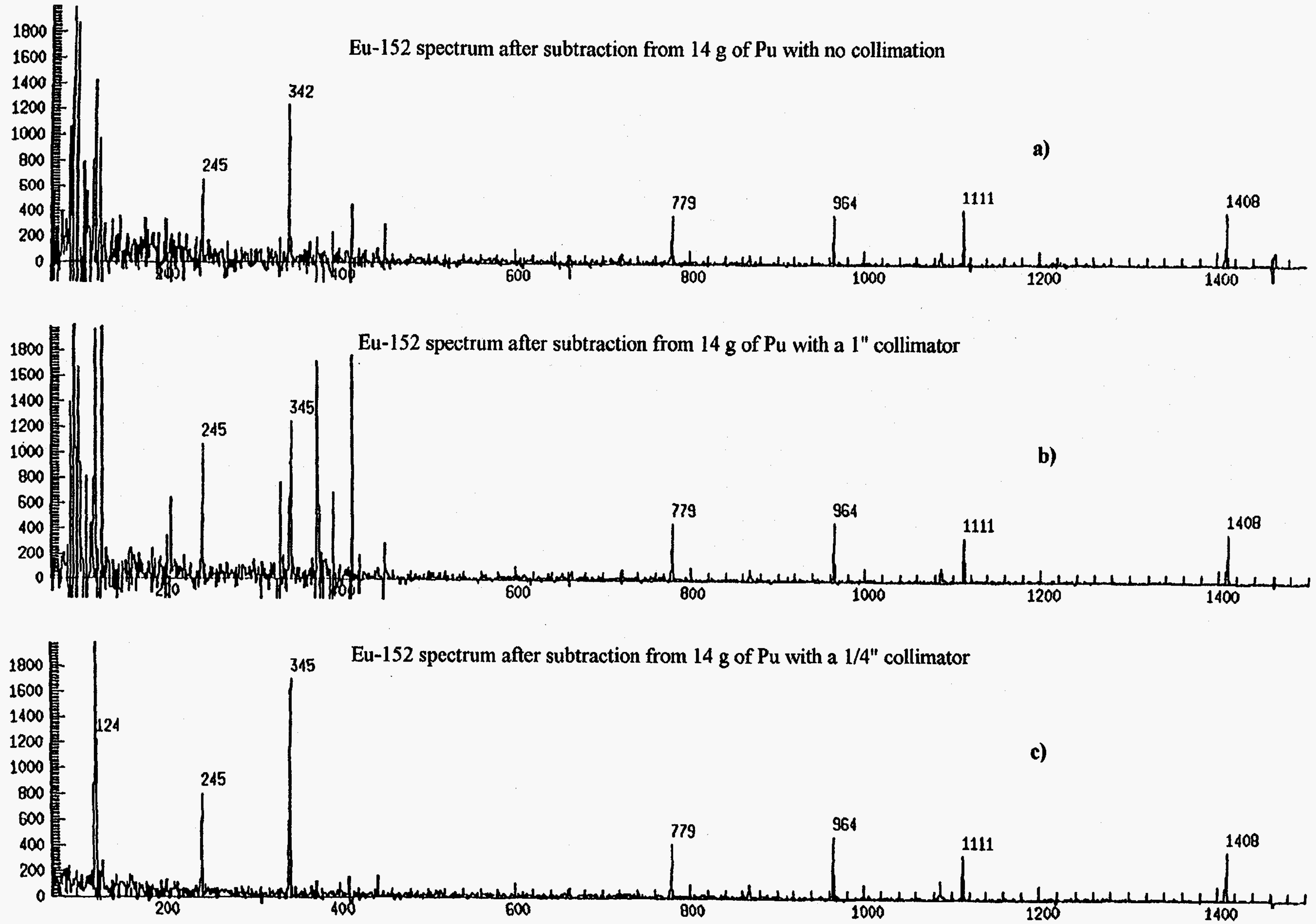


Figure E-6. Spectra obtained with 14 g of Pu using Eu-152 and Co-57 sources. A 1/4" collimator was placed between the sources and the NaI detector.



Eu-152 spectrum after subtraction from 14 g of Pu with no collimation

Eu-152 spectrum after subtraction from 14 g of Pu with a 1" collimator

Eu-152 spectrum after subtraction from 14 g of Pu with a 1/4" collimator

Figure E-7. The effects of no collimation, a 1" collimator, and a 1/4" collimator on subtracted Eu-152 spectra.