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Radioactive Material Inventory Optimization Using the Plutonium-239 Equivalent Curie Methodology

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ABSTRACT

The purpose of this paper is to discuss the Plutonium-239 equivalent curie (PEC) methodology used to optimize and limit the material-at-risk (MAR) in the recently established Irradiated Materials Characterization Laboratory (IMCL) within the Materials and Fuels Complex at the Idaho National Laboratory. IMCL is operated to provide a dynamic, re-configurable capability to analyze and characterize irradiated or non-irradiated radioactive material samples. The operations unique to IMCL include sample storage, preparation, and analysis using experiment-specific instrumentation such as, electron probe micro-analyzer (EPMA), focused ion beam (FIB), transmission electron microscope (TEM), and scanning electron microscope (SEM).

The methodology used to characterize the radioactive material inventory enables IMCL to operate in such a way that complements their capability and program goals without compromising the safety of the worker, co-located worker, or general public. The PEC concept offers a method in which all radionuclide inventory is normalized to a common radiotoxic hazard index that can be summed to a single value. The PEC concept is useful in the comparison of various material types for hazard categorization and for identifying worst-case and bounding inventory.

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Introduction

INL/MFC Background

Idaho National Laboratory (INL) is a government-owned reservation covering approximately 890 square miles in southeast Idaho, approximately 25 miles west of Idaho Falls, Idaho (Figure 1). INL was first established in 1949 as the National Reactor Testing Station (NTS) for construction and testing of various experimental and research reactor, reactor fuels, structural components, materials, and reactor safety programs.

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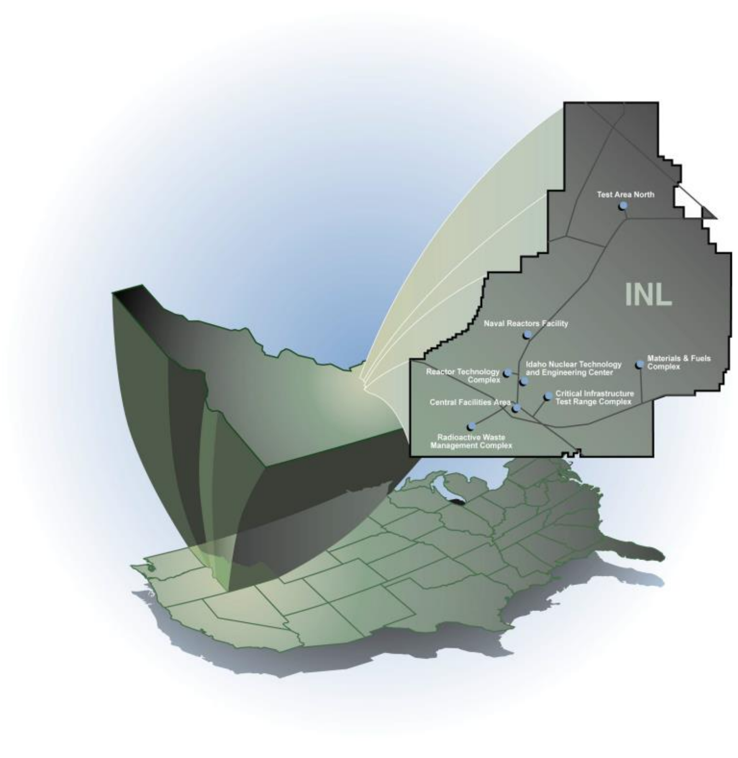

Figure 1. Location of the Idaho National Laboratory Site.

The Materials and Fuels Complex (MFC) is the easternmost facility located on the INL. MFC was formerly known as Argonne National Laboratory –West and operated by the University of Chicago, the MFC site covers an area of approximately 890 acres. Construction of the MFC site began in the mid-1950s with the Experimental Breeder Reactor-II (EBR-II) and support facilities, following the successful demonstration of the EBR-I reactor which is also located on the INL reserve. The EBR-II program, which is no longer in operation, was developed for research and development of liquid metal fast breeder reactor technology.

IMCL Facility Description

Irradiated Materials Characterization Laboratory (IMCL) is a single-story structure encompassing approximately 12,000 square feet of floor space, inclusive of both the operating wing (operating gallery, shipping bay) and support wing (mechanical room, office area). The IMCL floor plan is illustrated in Figure 2. IMCL is operated to provide a dynamic, re-configurable capability to analyze and characterize irradiated or non-irradiated radioactive material samples. The operations unique to IMCL include sample storage, preparation, and analysis using experiment-specific instrumentation such as, electron probe micro-analyzer (EPMA), focused ion beam (FIB), transmission electron microscope (TEM), and scanning electron microscope (SEM).

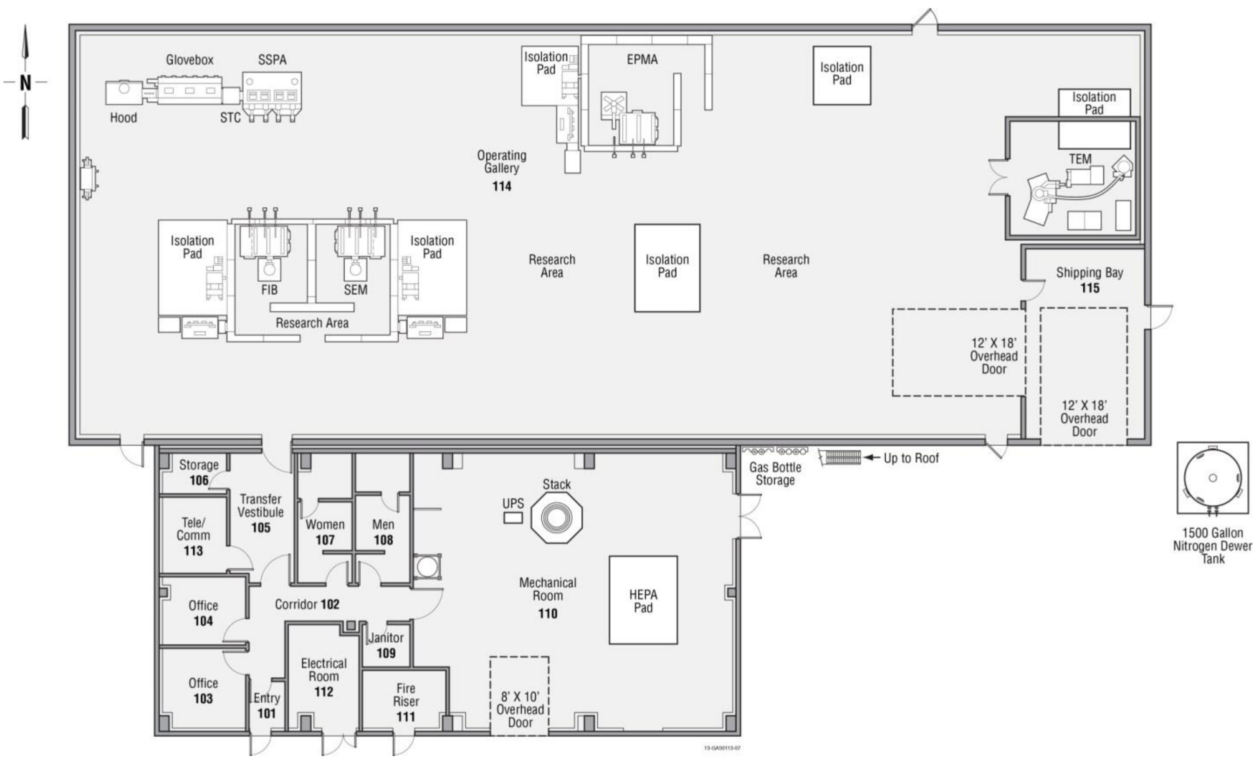

Figure 2. The IMCL floor plan.

IMCL Processes

Sample material is received at IMCL in a shielded container (SC) or is hand carried in a bag or small container. Assuming the material is in an SC, the material can be transferred to the FIB, SEM, or EPMA confinement and shielding via a transfer port. The sample is unloaded remotely using manipulators and transferred into the instrument for analysis. If the sample requires preparation, the SC is connected to the east side of shielded sample preparation area (SSPA) where it is cut and mounted in epoxy in a met mount for polishing. The met mount is transferred to the west side of the SSPA where it is examined using an optical microscope to ensure the quality is adequate prior to moving to the instrument confinement and shielding. Radiation rates of the sample are taken here. Based on radiation levels, the sample may be transferred from the shielded transfer cell (STC) to the glovebox for additional preparation or decontamination.

After the sample has been prepared, either in the SSPA or by another lab, the sample may go to the EMPA or SEM shielding and confinement to be analyzed. The sample is coated for the EPMA, which employs a non-destructive chemical analysis process. The amount of material that can be transferred by SC or into a confinement is 10 PEC. To prepare the sample for the TEM, it is sent to the FIB shielding and confinement where a typical sample size of 10 × 10 × 50 µm is cut. The prepared sample can usually be transferred in bags or small containers.

Plutonium-Equivalent Curie Methodology

Background

The PEC methodology was developed to be utilized in the Waste Isolation Pilot Plant – Waste Acceptance Criteria (WIPP-WAC). The concept of ^{239}Pu equivalent curie was intended to eliminate the dependency of radiological analyses on specific knowledge of the radionuclide composition of a transuranic (TRU) waste stream by normalizing all radionuclides to a common radiologic hazard index.[1] ^{239}Pu was selected as the common radiotoxic hazard index because it yields the bounding inhalation dose component and is a common constituent of waste streams encountered in the defense complex. The PEC methodology allows for the comparison of various radioactive material distributions for the purpose of hazard categorization and for identifying a facility’s worse-case MAR inventory.

PEC is defined mathematically as follows:

Eq. 1

$$\boldsymbol{PEC = \sum_{i=1}^{k} \frac{A_i}{WF_i}}$$

Eq. 2

$$\boldsymbol{WF_i = \frac{e_0}{e_i}}$$

where,

$$k = number\ of\ radionuclides$$

$$A_i = activity\ of\ radionuclide\ i\ (Ci)$$

$$WF = weighting\ factor\ (unitless)$$

$$e_0 = {}^{239}Pu\ dose\ coefficient$$

$$e_i = radionuclide\ i\ dose\ coefficient\ \left(\frac{Sv}{Bq}\right)$$

The WIPP-WAC has deemed this methodology appropriate for the WIPP facilities on the basis that the radiological dose consequences of an airborne release of a quantity of TRU radioactive material with a known radionuclide distribution will be essentially identical to that of a release of that material expressed in terms of a quantity of ^{239}Pu.[1]

Justification

The MAR in IMCL is equated to ^{239}Pu for the purposes of developing material quantity limits. These limits were established based upon bounding accident scenarios involving the inhalation of ^{239}Pu. The inhalation pathway is considered to be the primary route for the intake of radionuclides by the IMCL facility worker. The deposition and retention of inhaled radionuclides in the respiratory tract constitute a unique radiation protection problem.[2] Alpha-emitting radionuclides deposited in the respiratory tract are either "insoluble" (not readily translocated to other tissues) or "relatively soluble" (more readily translocated to other tissues) which may be more readily excreted from the body.[2] ^{239}Pu is relatively insoluble and retained in the lungs or translocated to bone surfaces and the liver. The biological half-life for plutonium deposited in the skeleton is approximately 100 years and 40 years for deposition in the liver.[3] ^{239}Pu is an alpha emitter with a mean energy of 5.15 MeV.[4] However, plutonium is considered radiotoxic not because of its energy emission but because of its tenacious retention by the skeleton and liver. This reasoning may provide the basis for the selection of ^{239}Pu as the common radiotoxic hazard index and can be utilized to bound the MAR in IMCL.

Applicability to IMCL

The small-scale research activities performed in IMCL involve irradiated and non-irradiated nuclear fuel samples in metallic or ceramic (oxide, nitride, carbide) form. These samples will typically contain activation products (^{60}Co, ^{63}Ni) and trans-uranium species (^{239}Pu, ^{241}Am). The PEC methodology was selected for use at IMCL since ^{239}Pu is a common component in the of legacy materials at MFC and provides a bounding inhalation dose component for the majority of materials to be encountered in the facility.[9] This methodology enables IMCL to operate in such a way that complements their capability and program goals without compromising the safety of the worker, co-located worker, or general public.

Example

The PEC calculation involves the ratio of ICRP-68, "Dose Coefficients for Intakes of Radionuclides by Workers,"[5] and ICRP-72, "Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 5 Compilation of Ingestion and Inhalation Coefficients,"[6] inhalation effective dose coefficients (e) to yield the weighting factor (WF) shown in Eq.2. The weighting factors (WF-68 and WF-72) are first calculated based upon either ICRP-68 or ICRP-72 (e) values. The values selected are dependent on the specifics of the accident scenarios to be analyzed. In cases where the worker doses are limiting (i.e., approaches evaluation guidelines), the WFs are based on ICRP-68 (e) values. However, in cases where the dose to the public instead of to the worker is most limiting, the WFs should be based on ICRP-72 (e) values. At INL, due to distance to the public for most of the facilities, the worker doses will more than likely be limiting; therefore, the WF-based ICRP-68 (e) values would be most appropriate.

The ICRP-68 dose coefficients are derived from the Human Respiratory Tract Model, defined in ICRP-66, and the organ/tissue weighting factors cited in ICRP-60 for uses of radiological protection calculations for the inhalation of radionuclides by the worker.[7] Similarly, ICRP-72 dose coefficients are used in radiological protection calculations for the inhalation of radionuclides by a member of the public. For workers, the dose coefficients used are based upon a 5 μm activity median aerodynamic diameter (AMAD) particle size and a pulmonary clearance class that will yield the highest 50-year committed effective dose. The pulmonary classes and particle sizes are specified in ICRP-68 and ICRP-72. Table 1 displays an example calculation of the weighting factors using Eq.2.

Table 1. Calculation of weighting-factors (WF-68).

Radionuclide	ICRP-68 e_{50}, (Sv/Bq)	Inhalation Class	WF-68
Pu-239	3.2E-05	M	1.0E+00
Cs-137	6.7E-09	F	4.8E+03
Am-241	2.7E-05	M	1.2E+00
Co-60	1.7E-08	S	1.9E+03

The facility MAR, in units of curies, is divided by the WFs as shown in Eq. **1** to obtain the radionuclides ^{239}Pu equivalency in curies, Table 2.

Table 2. Calculation of the MAR associated with 1 PEC.

Radionuclide	MAR, (Ci)	WF-68	PEC-68, (Ci)
Pu-239	1.0E+00	1.0E+00	1.0E+00
Cs-137	4.8E+03	4.8E+03	1.0E+00
Am-241	1.2E+00	1.2E+00	1.0E+00
Co-60	1.9E+03	1.9E+03	1.0E+00

The data in Table 2 illustrates that it would take 1.2 Ci of ^{241}Am or 4,800 Ci of ^{137}Cs to yield an effective dose equivalent to the inhalation of 1 Ci of ^{239}Pu. MAR limits can be established based upon these PEC values. However, the risks associated with direct radiation exposure and other potential pathways of intake must be evaluated and controls put in place to mitigate these risks for non-TRU radionuclides.

IMCL Accident Analysis

The chosen accident release scenarios for IMCL could involve radioactive material handled outside of closed confinement released due to drop or fire events, radioactive material in closed confinement (assumed to be contained within a single confinement box or a shipping cask/shielded/container) released due to drop or fire events, radioactive material inside of a sample analysis station released due to a seismic event, or the total building inventory released due to a combined natural phenomena hazard (NPH) (any or all) event.[8]

Inhalation Scenarios

The inhalation pathway was determined to be the most limiting pathway which in turn would yield the highest dose consequence to the facility worker, collocated worker, and public. The uptake of radionuclides via inhalation is the primary pathway for the accidents analyzed at IMCL. The drop and combustible fire scenarios are assumed to involve radioactive material outside of closed confinement at a maximum of 1 PEC. The vehicle fuel fire and cask/shielded container breach are assumed to involve radioactive material within closed confinement at a maximum of 10 PEC. The seismic event is assumed to involve radioactive material within a sample analysis station at a maximum of 10 PEC. The combined NPH (any or all) event is assumed to involve the entire facility inventory of 300 PEC. The analysis concluded that a radioactive material form control be put into place in IMCL to mitigate the resulting doses received from these postulated accidents. The 10 and 1 PEC radioactive material handling safety analysis commitment (SAC) was developed to limit the radioactive material release to workers inside IMCL under normal and accident handling conditions. Also, the facility radioactive material limit (SAC) of 300 PEC was developed to limit the amount of radioactive material that could potentially be exposed to forces associated with the NPH-induced event, such as building damage or collapse.

Radioactive material releases would be detected through the use of continuous air monitoring (CAMs), Radiation Area Monitors (RAMs), and other radiation detection equipment.

Direct Radiation Exposure

External exposure hazards are not evaluated using the PEC methodology.[9] However, due to the presence of high energy gamma emitting radionuclides (^{60}Co) in the samples, the consequences of external radiation exposures have been evaluated in the accident analyses. A shielding configuration control is used to mitigate the radiation exposure to facility workers when the direct penetrating radiation rate is ≥18 rem/hr at 30 cm. The single sample source term that measures ~18 rem/hr at 30 cm in the absence of shielding is 3 Ci of 1 MeV photons (comparable to ~ 1.3 Ci of Co-60).

Summary

The PEC concept offers a method to normalize all radionuclides in a specific radioactive material to a common radiotoxic hazard index that can be summed to a single value. The use of the PEC methodology eliminates the dependency for radiological analyses sample material for the creation of the bounding accident scenarios for IMCL. Therefore, the use of the PEC will allow for the comparison of various radioactive material distributions for the purpose of hazard categorization and identifying a worst-case inventory related to inhalation risk in facility accidents. Further, as ^{239}Pu is the bounding inhalation dose component for the majority of materials encountered at MFC, the PEC methodology may also be utilized for the preliminary estimation of dose consequences for specific accident scenarios. However, further analysis involving the Radiation Protection Program (RPP) is necessary for non-TRU radionuclides present in the facility as the PEC methodology only encompasses the inhalation dose consequence and does not account for direct radiation hazards or other pathways of intake.

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