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Replacement of Lead-Loaded Glovebox Gloves with an Attenuation Medium of non-RCRA-Hazardous Metals

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Abstract

Programmatic operations at the Los Alamos National Laboratory Plutonium Facility (TA-55) involve working with various amounts of plutonium and other highly toxic, alpha-emitting materials. The spread of radiological contamination on surfaces, airborne contamination, and excursions of contaminants into the operator's breathing zone are prevented through the use of a variety of gloveboxes (the glovebox, coupled with an adequate negative pressure gradient, provides primary confinement). Radiation shielding is commonly used to protect the glovebox worker from unintentional direct and secondary radiation exposure, while working with plutonium-238 and plutonium-239. In these environments, low-energy photons, i.e., those less than 250 keV, are encountered. Shielding glovebox gloves are traditionally composed of lead-based materials, but these are now considered hazardous waste. This has prompted the development of new, non-hazardous-shielding glovebox gloves. No studies, however, have investigated the effectiveness of these new glovebox gloves. We examined both leaded and non-hazardous-shielding glovebox gloves and compared their attenuation effectiveness over the energy range of interest at TA-55. All measurements are referenced to lead sheets, allowing direct comparisons to the common industry standard of 0.1 mm lead equivalent material. The attenuation properties of both types of glovebox gloves vary with energy, making it difficult for manufacturers to claim lead equivalency across the entire energy range used at TA-55. The positions of materials' photon energy absorption edges, which are particularly important to improved attenuation performance, depending upon the choice of radiation energy range, are discussed. This effort contributes to the Los Alamos National Laboratory Continuous Improvement Program by improving the efficiency, cost effectiveness, and formality of glovebox operations.

1. INTRODUCTION

Chemical and metallurgical operations involving plutonium and other nuclear materials in support of the U. S. Department of Energy's (DOE) nuclear weapons program account for most activities performed at the Los Alamos National Laboratory's Plutonium Facility (TA-55). To preclude uncontrolled release, gloveboxes are used to confine plutonium during laboratory work. Gloveboxes used for radioactive materials are maintained at a lower pressure than the surrounding atmosphere, so that microscopic leaks result in air intake rather than hazard outflow. A typical glovebox train is shown in Fig. 1.

Figure 1. Typical Glovebox Train

Many TA-55 operations generate large amounts of gamma radiation with the main sources of gamma rays coming from various isotopes of uranium and plutonium [1]. With freshly purified plutonium, most of the radiation comes from *soft* (17.2 keV) X-rays. More penetrating (59.6 keV) gammas are emitted by americium-241, which *grows in* as plutonium-241 decays. All grades of plutonium contain plutonium-241. In plutonium, that is more than 10 years old (since purification), these gammas are usually the source of most of the external radiation. Thus, the external radiation from plutonium increases during the 14-year half-life of plutonium-241. Plutonium-238 is used in the production of heat sources. Photon energies emitted from plutonium-238 that add to the energies of interest include the following: 43.5 keV, 74.7 keV, 99.9 keV, and 228.3 keV.

From a structural design standpoint, glovebox gloves are the weakest part of the glovebox system and more susceptible to failure from mechanical, chemical, radiological, and thermo stressors than the glovebox walls or windows. The low energy and moderately penetrating gamma and X-ray radiation from plutonium easily penetrate the rubber gloves in a glovebox, resulting in a radiation dose to the hands. Recognizing this and other vulnerabilities, the Glovebox Glove Integrity Program (GGIP) was developed at TA-55 to minimize and/or prevent glovebox glove events. Other accomplishments of this program have been previously reported [2]. A key element of the GGIP is the proper selection of glovebox gloves. Glovebox gloves with a layer of radiation shielding can be used to reduce the extremity dose.

Due to its high density, lead has been used for shielding against primary gamma rays. Previous studies have shown lead to be an excellent shield against this type of radiation [3]. Leaded glovebox gloves used for radiation shielding (hereafter referred to as *leaded gloves*) are a commercially available item produced by North Safety by Honeywell. In leaded gloves, lead oxide (Pb_3O_4) is integrated into the glovebox glove matrix during fabrication. Lead oxide is used because of its high mass absorption coefficient for low energy gamma rays, its high specific gravity, and its compatibility with Neoprene [4]. The leaded gloves are constructed of a layer of lead oxide dispersed in a Neoprene elastomer, bound between inner and outer layers of Hypalon. Hypalon with its exceptional chemical resistance to acid and alkali products, protect glovebox workers from chemical hazards inside the glovebox. There are several thousand leaded gloves at TA-55 that are potentially a mixed waste when discarded.

Glovebox gloves composed of non-hazardous shielding materials (hereafter referred to as *unleaded gloves*) have been relatively recently developed and are commercially available from Piercan U.S.A. These unleaded gloves are composed of three different and inseparable layers: polyurethane, a shielding layer, and Hypalon. Polyurethane provides superior protection against mechanical risk including tears, puncture, cuts, and wear. While the exact formulation is proprietary, the components of the shielding layer consist of bismuth, tungsten, and lanthanum.

The TA-55 specification for shielding glovebox gloves requires that leaded and unleaded gloves have nominal attenuation properties to soft gamma radiation equal that of 0.10-mm-thick lead metal foil. Uniformity must be such that there will be no significant areas (1.25 square cm) containing less than 0.08 mm or more than 0.15 mm lead equivalence. If this requirement cannot be met, measurement of dose attenuation for weakly penetrating photon radiation (<70 keV) must be performed. The K absorption edge of the shielding material is a parameter that is also worth consideration, as the element absorbs strongly in the energy levels immediately above this value. The K absorption edge for lead, bismuth, tungsten, and lanthanum are compiled in Table 1.

Table 1. Shielding Material Properties

Element	Atomic No.	Density (g/cm^3)	K Absorption Edge (keV)
Lead	82	11.36	88.004
Bismuth	83	9.75	90.525
Tungsten	74	11.36	69.525
Lanthanum	57	6.16	38.930

Another TA-55 glovebox glove specification is the following: in order for shielding glovebox gloves to be considered non-hazardous, no constituents listed in the EPA standard, 40 CFR 261.24, *Toxicity Characteristics*, may be in the formulation. In a previous report, using Toxicity Characteristic Leach Procedure (TCLP) Method 1311, leaded gloves were determined to have a concentration range of 26–38 ppm; well above the regulatory level of 5 mg/L [5].

The three major risks from glovebox operations come from glovebox ergonomic injuries, internally deposited radionuclides, and the dose from externally penetrating radiation [6]. When a measure is proposed to improve the hazard control system of glovebox operations, these three risk factors must be considered and weighted. Product specifications, such as thickness, tensile strength, and ultimate elongation, have a direct correlation with these risks. In general, thicker glovebox gloves of the same material provide better protection against punctures, cuts, sharps, and abrasive hazards with an increased likelihood of glovebox ergonomic injuries and external dose. Thinner and/or softer material glovebox gloves are preferred for tasks that require more dexterity. Tensile strength and ultimate elongation values are independent of thickness. In general, the higher the tensile strength and ultimate elongation values, the more resistant the glovebox glove is to physical hazards. The EN 388 mechanical ratings for puncture take into account the thickness and different material properties of the glovebox glove. To summarize, the thicker the glovebox glove of the same material, the lower the likelihood of a glovebox glove opening and the higher the likelihood of glovebox ergonomic injuries and external dose [7]. The higher the tensile strength and ultimate elongation values, the lower the likelihood of glovebox glove openings. The higher the tensile strength and the lower the ultimate elongation values, the higher the likelihood of glovebox ergonomic injuries and external dose. In many cases, the higher the EN 388 rating, the more the glovebox gloves are resistant to puncture. In the following report, the features of leaded and unleaded shielding glovebox gloves are compared, arguments recommending the latter are presented, and pollution prevention benefits discussed.

2. EXPERIMENTAL DESIGN

2.1. Materials

Planchets (9 cm X 9 cm) were cut from five pairs of 0.9 mm Polyurethane/Shielding Polymer/ Hypalon glovebox gloves (8USY4DA, $1100 a pair), as received from Piercan U.S.A. (San Marcos, CA) and five pairs of 0.8 mm Hypalon/lead oxide-Neoprene/Hypalon glovebox gloves (8YLY, $270 a pair), as received from North Safety Products (Cranston, RI).

2.2. Rheological Instrumentation

The tensile properties of the samples were determined on a Test Resource 1000R System, a computerized mechanical testing system. The testing regime for the glovebox glove sample was from American Society for Testing and Materials (ASTM) D 1708-02a. Samples were strained at a rate of 50 cm/min at room temperature with an initial jaw separation of 2.5 cm. A 100 N transducer was used for all measurements. Five samples from three glovebox gloves were tested and the data were averaged.

2.3. Lead Equivalency

Lead equivalency was determined by comparative measurement of the X-ray attenuation properties of a lead foil step wedge with the subject shielding glove on X-ray film. A densitometer reading of the X-ray film determined the equivalence of lead thickness as compared to the step wedge based on the measured shades of gray. The thickness measurement was converted to lead equivalency using a ratio determined by a test conducted a minimum of twice annually. The test consisted of placing glovebox glove samples and a step wedge on an X-ray film. The film was then exposed to an X-ray tube source with an energy level of 70 kV. The film was developed and the images were evaluated on a densitometer. Using the film density readings, the ratio between the thickness of the glovebox glove's lead layer and the lead equivalency was calculated. Refer to ASTM F640 *Standard Test Methods for Determining Radiopacity for Medical Use,* for guidelines for conducting lead equivalency measurements.

3. RESULTS

3.1. Physical and Mechanical Properties

The physical and mechanical properties of the 0.8 mm Hypalon/lead oxide-Neoprene/Hypalon glovebox gloves (8YLY) from North Safety and 0.9 mm Polyurethane/Shielding Polymer/ Hypalon glovebox gloves (8USY4DA) from Piercan U.S.A. are compiled in Table 2.

Table 2. Glovebox Glove Physical and Mechanical Specifications

Specification	8YLY	8USY4DA
Company	North	Piercan U.S.A
Shielding Material	Lead	Bismuth/Tungsten/Lanthanum
Lead Equivalence (mm)	0.1	
Thickness (mil.)	30	35
Tensile Strength (psi)	1200	1500
Ultimate Elongation (%)	275	500
Puncture (newton)*	>20	>40

***EN 388 puncture rating for each glove.**

In the leaded gloves, lead oxide that is embedded in the Neoprene is sandwiched between two Hypalon glove matrices. The glovebox glove from Piercan U.S.A. is composed of a proprietary polymer shielding layer composed of bismuth, tungsten, and lanthanum, specially formulated with Neoprene to provide shielding properties equal to or better than lead without the health or environmental hazards of lead. According to its product specifications, the leaded gloves should have nominal shielding power to soft gamma radiation equal that of 0.10-mm-thick lead metal foil. Leaded gloves are 0.1 mm thinner than the non-leaded shielding glovebox gloves. While both leaded and unleaded gloves meet the tensile strength and elongation requirements, unleaded gloves have significantly higher tensile strength, elongation and puncture resistance values.

3.2. Stress and Strain Properties

A plot of stress versus strain as an example of each material's response when loaded to failure is shown in Fig. 2.

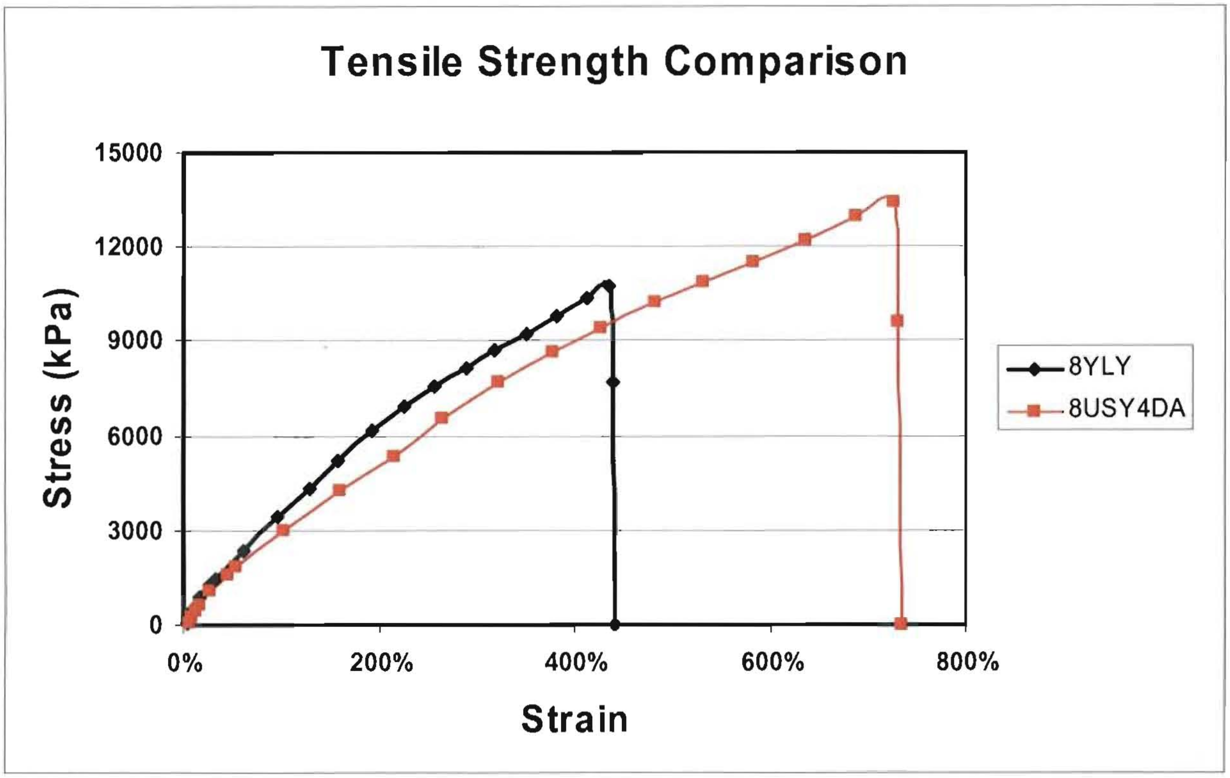

Figure 2. Stress versus Strain Comparison

Tensile strength is defined as the maximum load applied in breaking a tensile test piece divided by the original cross-sectional area of the test piece (also termed maximum stress and ultimate tensile stress). Ultimate elongation is the elongation at time of rupture (also termed maximum strain). The specification for the tensile test and ultimate elongation are the minimum acceptable values. The strain is the amount of elongation divided by the original gauge length of the material. The data provides information about the strength of each material and its response to load. The most important material property to consider is the toughness, for which there is no current specification. Toughness is the integral under the engineering stress versus strain curve, which is the total integrated force required to take the sample from its initial unstressed state to failure. The value for toughness is more important than the value for maximum stress, because maximum stress gives no indication about the energy required to reach that maximum value.

3.3. Lead Equivalent Thickness

Lead Equivalent Thickness results are shown in Table 3.

Table 3. Lead Equivalent Thickness Results

Glovebox Glove	Lead Equivalent Thickness (mm Lead)	
	Average	Standard Deviation
8YLY	0.10	0.023
8USY4DA	0.06	0.008

The attenuation equivalent of the glovebox gloves is expressed in mm lead; it corresponds to the thickness (in mm) of the reference material (lead) with the same attenuation factor as the equipment submitted to test, for the same quality of radiation (energy and spectrum). The tests have been conducted with an X-ray tube voltage and filtration of 70 keV and 0.10 mm copper, respectively. Taking into account that shield material differences can create different radiological phenomena like bremstrhaum effects in the material that can create low energy film exposures, slewing the results. Considering the fact that the intent of the shielding glove is to mitigate skin dose in the realm of 0.07mm depth, very low energy photons i.e. bremstrhaum are not critical to the skin dose at 0.07mm depth. Therefore direct attenuation by photon energy measurements can provide more usable data.

3.4. Attenuation Properties by Photon Peak Energy

A plot of percent attenuation versus photon energies is shown in Fig. 3.

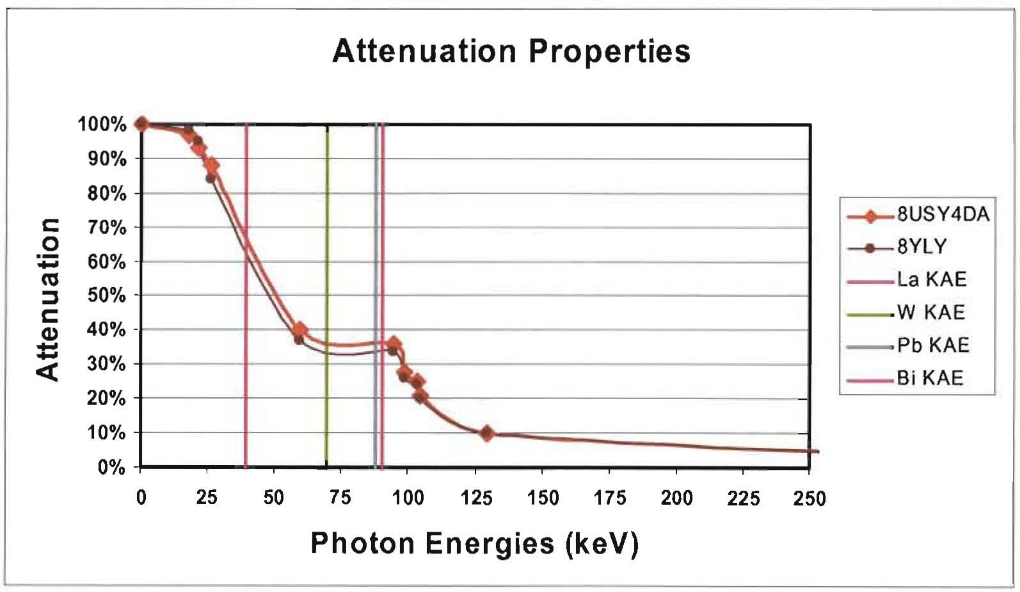

Figure 3. Attenuation Properties Comparison

Figure 3 depicts the theoretical calculated attenuation properties for the 0.1mm lead equivalent leaded (8YLY) and unleaded (8USY4DA) gloves.

4. DISCUSSION

As discussed above, thinner glovebox gloves are preferred for tasks that require more dexterity, as shown in Table 2. Thus, the leaded gloves should be preferred over the unleaded gloves. Nevertheless, the unleaded gloves are a better option from an ergonomic perspective because they have a significantly higher elongation value. These unleaded gloves allow for more flexibility and less strain on the upper extremity and the back. This correlates with a decrease in injury, particularly injuries resulting from overuse. In addition, unleaded gloves would also be useful for situations in which the use of protective gloves over glovebox gloves is necessary [8]. Loss of dexterity that results when the protective gloves are used is lessened with the use of the more flexible unleaded glove. Unleaded gloves also have a significantly higher tensile strength value. The higher tensile strength and elongation values, as compared to the leaded glove, results in overall greater safety from mechanical hazards, as shown in Fig 2. This is particularly true and important for operations around equipment or machinery that could cause injury or penetration of the glovebox gloves (e.g., around rotating parts, sharps, or operations that require fine motor control).

Based on the lead equivalent thickness results, it would appear that the unleaded glove is a significantly less effective attenuation medium, as shown in Table 3. The complementary study carried out in different test conditions gives a better estimate of the attenuation properties of the unleaded gloves for the photon energies of interest for skin dose, as shown in Fig. 3. These conditions were representative of the risk encountered by the glovebox worker. Since shielding glovebox gloves are mainly used in plutonium-238 operations, the selected shielding materials must be effective for low photon energies found in the spectrum of the plutonium-238, especially the prominent 43.5 and 99.9 keV photons. Choosing bismuth as the primary component of the non-hazardous shielding glovebox glove was prudent because of bismuth's k-absorption edge at 90 keV, high atomic number and low density. Since the density of lead is higher than bismuth, a thicker bismuth layer can be used without exceeding the weight of the lead. The lower density of bismuth allows a slight improvement over lead. The selection of lanthanum was based on its K-absorption and low density. Lanthanum absorbs strongly the photons in the energy range of the 43.5 keV photon. This photon is a major source of exposure arising from the decay of plutonium-238. Tungsten is effective, but with a density higher than lead, heavier shielding is required. The position of the tungsten K absorption edge at 69.5 keV provides superior attenuation compared to lead in the range of the 74.7 keV photon.

In order to get a better estimate of the attenuation properties of the glovebox gloves, a complementary study should be carried out in different test conditions than those reported in this paper; with more focused photon energy attenuation measurements using specific sources and spectroscopy, rather than the gross large scale X-ray tube/film method. Future measurement of dose attenuation for weakly penetrating photon radiation (<70 keV) should be performed with the use of Hp (0.07)-specific thermoluminescent dosimeter (TLD) chips and a sufficient activity americium-241 source to provide an accumulated dose of approximately 300 mrem in < 24 hours.

Lead measurements and TLD choice could be performed per ICRU 47 requirements [9], and sufficient shielding used around the test area to avoid any interference dose from external sources. Using TLD manufacturers handling instructions, four lithium borate ($Li_2B_4O_7$) or lithium fluoride (LiF), TLD chips could be placed inside a shielded and unshielded (control) glovebox glove, The control glovebox glove should be made of the same material as the outer layers of the shielding glovebox glove to be tested. To ensure consistent geometry of TLD placement, a glovebox glove insert with fingers would be used. The TLDs would be positioned on the insert prior to placement in glovebox glove. X-ray testing of the glovebox gloves would be a part of the evaluation. One TLD chip would be placed at the middle of pinky finger, one TLD chip at middle of index finger, one chip at center of the palm, and one chip at 38 cm up the sleeve from the middle finger tip. In addition, a test would be performed to ensure that there is no back-scatter when performing the Dose Attenuation Factor (DAF) Calculation. A phantom (tissue equivalence) would be considered when performing this test. Glovebox gloves would be laid flat on platform 15 cm apart with the TLDs inside, facing upward. Measures would be taken to ensure equal source to glovebox glove geometry and equal source exposure for each glovebox glove. The source would be exposed to glovebox gloves and left for ample time to provide ~300 mrem of unshielded accumulated dose to the exterior of the glovebox gloves, then the source would be shielded and the TLD chips removed from glovebox gloves, placing them in marked envelopes with positions and glovebox glove name on envelopes. Background TLD would be included in the calculation. The TLDs would then be sent for analysis and the results recorded. The following formula would be used to calculate the DAF: DAF = unshielded dose/shielded dose. The average value of the four TLD chips would be used in the calculation. Source type and activity would be recorded with the DAF. Attenuation uniformity between the four chips should not vary by more than -30% from rated DAF. Last, the attenuation between the four chips should not vary more than 20%. The DOE Standard is 10%, but at LANL a 20% variation is allowed for instruments and calculations.

Leaded gloves are hazardous waste when discarded, and mixed-waste is created when this material becomes contaminated with radioactive material. It is known that mixed-waste is more expensive to dispose of than radioactive waste. This adds up to a significant annual expense, especially considering the amount of leaded gloves generated each year in all the DOE facilities. Los Alamos National Laboratory (LANL) annually generates about 2,500 kg of mixed waste in the form of leaded gloves.

Mix TRU waste costs $111K per cubic meter. LLW costs $17K per cubic meter. The cost for a pair of leaded and unleaded gloves is $288 and $1100, respectively. Short-term use leaded gloves is more financially attractive than their non-hazardous substitute. With lead being the fourth most abundant metal, this is not unexpected. Leaded gloves are disposed of as mixed transuranic (TRU) waste, while most unleaded gloves could be disposed as Low Level waste (LLW). This brings up several important issues that need to be addressed. Unleaded gloves cost 4 times as much as leaded gloves to buy. Most leaded gloves cost 7 times as much as unleaded ones to dispose of. No additional hazards are introduced by replacing lead with a tungsten, bismuth, and lanthanum formulation. This along with intangible costs of occurrence reports and audit findings would make replacing lead, with a non-hazardous substitute, a prudent decision in the long-term. This leaves hazards that adversely impact the environment. Line managers will find that the elimination of liability associated with using a Resource Conservation and Recovery Act (RCRA) material more appealing than that they are meeting the performance requirements of the institution.

A primary objective of an organization should be to maintain a safe and healthy workplace for personnel and to protect the public and the environment. The GGIP contributes to this objective by providing process improvements, so that workers can work more safely, easily, and effectively in a glovebox. Excellent performance in the GGIP is evident when radiation exposures are maintained well below regulatory limits, contamination and ergonomic injuries is minimal, and uncontrolled releases are prevented. Finally, it is the expectation of TA-55 management that glovebox ergonomic injuries, excursions of contaminants into the operator's breathing zone, and excess exposure to radiological sources associated with glovebox operations approach zero.

In summary, when radiation passes into an absorbing medium such as a body, some of the energy is transferred to that medium. Radiation passing through body tissues may produce biological damage, so glovebox workers routinely protect their arms (extremities) by using glovebox gloves with shielding to protect themselves from direct and secondary exposure to radiation. Shielding glovebox gloves are traditionally composed of lead-based materials, but these are now considered hazardous waste. This has prompted the development of new, non-hazardous shielding glovebox gloves. For a cost of 4 times that of leaded gloves, unleaded gloves manufactured by Piercan U.S.A. provide more effective shielding. Since no compromise in programmatic efforts is indicated and the quality of protection is maintained, these unleaded gloves are recommended over their leaded counterparts. The nonhazardous substitutes are higher in cost, but this is offset by eliminating the costs associated with onsite waste handling of RCRA items. In the end, replacing lead with non-hazardous substitutes eliminates waste generation and future liability. A primary objective of the GGIP is to minimize hazards associated with glovebox operations whenever possible. As with all other elements of business, there are costs associated with implementing a hazard evaluation and elimination program. While eliminating the hazards associated with materials may not be immediately apparent, simple life-cycle principles show that it is a wise investment in the end.

5. CONCLUSIONS

When practicable, complete elimination of lead in the workplace, especially at nuclear facilities, is desired. Overall, non-hazardous shielding glovebox gloves made from bismuth, tungsten, and lanthanum are more effective shielding materials and eliminate the toxic and environmental hazards associated with lead without adding hazards. Onsite waste handling cost associated with leaded glovebox gloves and long-term overhead costs justify these more expensive commercially available non-hazardous substitutes.

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