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CONTROL OF PERSONNEL EXPOSURES TO EXTERNAL
RADIATIONS IN A PLUTONIUM CHEMICAL PLANT

by

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ABSTRACT

This paper describes a practical approach to the control of personnel exposures to external radiations in a chemical plant handling kilogram quantities of plutonium in glove boxes. Calculated dose rates from various processing operations are compared to measured dose rates. Shielding materials for x-rays, gamma rays, and fast neutrons and their economic and operational impact are discussed. Personnel training, rotation, and work habits which must be considered in maintaining exposures below the accepted guide levels are included.

INTRODUCTION

The intent of this paper is to describe the application of a practical approach to the control of exposure to external radiations during the chemical processing of kilogram quantities of plutonium in glove boxes. I will, therefore, limit the discussion of technical considerations to that which is necessary for the sake of understanding.

When one considers the decay properties of the plutonium isotopes 238, 239, 240, 241, and 242⁽¹⁾, it will be noted that all are alpha emitters except 241 which is primarily a beta emitter. These properties, numerous animal experiments, and limited observation of human internal exposure have resulted in the acceptance of glove boxes as a means of limiting the internal exposure potential while handling plutonium.

Further study of the decay properties⁽²⁾ will reveal that when these isotopes of plutonium decay, a small part of their energy is given off as x and gamma rays. Decay products, such as americium 241 and uranium 237, contribute significant amounts of additional x and gamma rays. Spontaneous fission results in neutrons and additional gamma rays and when the plutonium is mixed or compounded with light elements, additional neutrons are produced by alpha-neutron reactions.

The amount of x-rays, gamma rays, and neutrons from a mixture of plutonium isotopes is dependent upon the isotopic concentration and

time since chemical purification. At Rocky Flats the majority of the plutonium mixtures handled have an isotopic concentration of less than 0.1 percent ²³⁸, about 95 percent ²³⁹, about 6 percent ²⁴⁰, less than 1.0 percent ²⁴¹ and less than 0.1 percent ²⁴². The time since chemical purification ranges from a few days to more than ten years.

The discussion of personnel dosimetry techniques and radiation detection equipment used at Rocky Flats would constitute complete papers in themselves. I will merely mention that we are including thermoluminescence in our planning for the future, but we are presently using DuPont 558 beta-gamma film worn on the body and at the wrist and Kodak Type A neutron monitoring film worn on the body. All radiation surveys for x and gamma rays are performed with an ionization chamber, such as the Victoreen 440 or 440 RF, and all neutron surveys are performed with a Bonner Sphere type⁽³⁾ survey instrument.

The Rocky Flats plutonium chemistry operations, which are performed in glove boxes and to which this discussion is directed, are batch dissolution of plutonium metal, oxide and tetrafluoride, continuous peroxide precipitation, americium separation and purification, continuous calcining, continuous fluorination, and batch reduction to metal. These operations involve hundreds of kilograms of plutonium and up to 100 grams of americium each month.

X and Gamma Rays

H. A. Moulthrop⁽⁴⁾ has reported the results of a study of the energy spectrums of the x and gamma rays from special plutonium samples in various physical forms and the effectiveness of various shielding materials, such as lead, lead glass, steel, safety glass, and Plexiglas.

Rocky Flats experience indicates that significant exposure to x and gamma rays can result from the handling of a few hundred grams of plutonium. We have found that the dose rates at the work stations can be reduced to less than 1.0 mr/hr through the use of 1/8 inch thick lead sheets over the metal portions of the glove boxes, 1/4 inch thick lead glass (equivalent to 1/16 inch of lead) over the windows, and hinged covers of 1/8 inch thick lead over the glove and bag ports. Hand exposures are controlled through the use of leaded dry box gloves. The majority of these gloves contain the equivalent of 0.16 mm of lead with a maximum of 0.36 mm of lead used at the operations which concentrate the americium 241.

Our experience indicates that this shielding has very little, if any, affect on the operator's efficiency. The cost of the glove box is increased by a factor of about 1.8 when this shielding is added.

Neutrons

When one considers neutron shielding, the energy of the neutrons must first be determined. The neutrons from spontaneous fission have a

continuous energy spectrum with a maximum intensity near 1 Mev. The neutron energy and abundance from alpha-neutron reactions is dependent upon the alpha energy and the target nuclei. Rocky Flats experience indicates that for the types of operations under discussion the predominant sources of neutrons are spontaneous fission and alpha-neutron reactions in oxygen and fluorine. For practical purposes, all of these neutrons can be considered as having 1 Mev of energy.

F. J. Allen and A. T. Futterer⁽⁵⁾ have reported the dose transmission factors for various neutron energies in polyethylene, water, concrete, and Nevada Test Site soil. This data and studies at Rocky Flats indicate that for the neutrons of concern polyethylene has a half value of 1.2 inches and a tenth value of 3.3 inches, water has a half value of 1.25 inches and a tenth value of 3.5 inches, Plexiglas* or Benelex** have a half value of 1.8 inches and a tenth value of 4.9 inches, and concrete has a half value of 2.1 inches and a tenth value of 5.7 inches.

Calculations will show that the neutron dose rate will exceed 1.0 mrem/hr at the work station if the amount of plutonium exceeds about 4 kilograms as metal, about 2 kilograms as oxide, or about 20 grams as a fluoride.

*Manufactured by Rohm and Haas, Philadelphia, Pennsylvania, and American Cyanamide, Wallingford, Connecticut

**Manufactured by Masonite Corporation, Chicago, Illinois

Time will not permit a detailed discussion of the chemistry operations, but it should be noted that the health physicist must be aware of the detailed design of the processing equipment and watch for potential problems. For example, the design of the Rocky Flats continuous calciner involved an almost horizontal tube about 8 feet long with a loading hopper on one end and a receiving hopper on the other. It was intended that each hopper would contain up to 2 kilograms of plutonium and that the tube would contain up to 5 kilograms of plutonium all as oxide. A line source calculation indicated a neutron dose rate of about 1.8 mrem/hr would result at a point 2 feet from the center of the tube. Neutron surveys at this point indicated up to 6 mrem/hr. Investigation of this difference revealed that plutonium oxide had spilled from the hoppers and a significant number of kilograms of plutonium had collected in the drive mechanism which turns the tube.

As a word of caution, criticality experts should be consulted and made aware of any plans to install neutron shielding. These shielding materials are also very effective neutron reflectors.

At Rocky Flats we have found that the practical limit for the thickness of neutron shielding on a glove box is a maximum of 5 inches. The first 2 inches do not have a significant effect on the operator's reach and vision, but at 5 inches the operator is seriously restricted. The

cost of the glove box is increased by a factor of about 2.2 when 2 inches of shielding is added and a factor of about 2.8 by the addition of 5 inches of shielding.

Personnel Training and Rotation

The experienced health physicist will be quick to point out that shielding alone is not a complete control. The shielding merely makes it possible for the trained operator to use the shielding in a manner which maintains exposures below the accepted guide levels. At Rocky Flats we have found it necessary to maintain a continuous training program. The training program stresses the need for good housekeeping practices inside of the glove boxes, good storage practices outside of the glove boxes, and the use of shielding, time and distance to limit exposures.

Supervision has found it necessary to administer a rotation program such that the workers are frequently moved between operations of high and low exposure potential. This program requires continuous and conscientious attention of the workers and their supervision.

CONCLUSION

The Rocky Flats experience indicates that kilogram quantities of plutonium mixtures typical to Rocky Flats can be chemically processed in glove boxes while maintaining external exposure within the accepted guide levels providing the proper shielding, personnel training and personnel rotation programs are utilized.

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