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In my lecture today, I shall discuss the hazards of plutonium handling, the philosophy of plutonium handling, glove box construction and materials, some plutonium handling techniques, and briefly mention some of the plutonium work in progress in this country. I shall not attempt to discuss the more difficult problem of handling mixtures of α, β, and γ-active materials except to say that this will be a problem of increasing importance as more power reactors which produce plutonium are built and operated and as plutonium-burning reactors come into use. Some installations in this country and abroad are already considering this problem and some progress toward effective methods of handling materials of this type has been reported. I have drawn material for my talk from a number of sources including my limited experience and visits to plutonium facilities at Los Alamos and Argonne National Laboratory, published articles, the book "Glove Boxes and Shielded Cells", edited by G. N. Walton, and replies to a questionnaire by an AEC glove box committee.

The hazards of plutonium handling are of three basic types: toxicity, criticality and fire. Since the greater part of my talk will be related to the toxicity of plutonium and the real cause for concern in case of a fire in a plutonium facility is due to the possibility of uncontrolled release of plutonium, I shall speak briefly on criticality control before going on to plutonium handling problems that I know more about.

The criticality problem is emphasized by the fact that several fatal accidents at Los Alamos can be attributed to criticality inciddents and a recent review mentioned sixteen incidents in which the reactivity of a fissile system accidentally exceeded prompt critical. In addition to producing lethal doses of neutrons and gamma-rays, a criticality accident generates a large amount of heat in a very short time which may sometimes lead to an explosion.

Criticality control is achieved by two methods: the always safe mass and the always safe shape. The shape of a system largely determines the leakage of neutrons from a system. If one of three dimensions of a container is made sufficiently small as compared to the other two dimensions, then no matter how much fissionable material is put into the container, criticality cannot be achieved. Where large amounts of plutonium or other fissionable material must be processed, this is the only method which will guarantee that criticality will not be achieved and experience has shown that it is essential that there be no unsafe container available into which the fissionable material can be accidentally transferred. Plutonium metal is geometrically safe in a 1.4 in. diameter cylinder and in a 0.2 in. thick slab in a full water reflector. Aqueous solutions of plutonium are geometrically safe in a 4.5 in. diameter cylinder and in a 1.2 in. thick slab in a full water reflector. These figures include a safety factor greater than 1.3. Five hundred grams of plutonium in solution can be made critical with a full water reflector. Consequently, only about 250 grams are allowed to be processed at one time for "always safe mass" processing. This subject is much more complicated than might be suspected from my brief discussion, but perhaps this will be sufficient to indicate the nature of the approach to safe handling of plutonium or other fissionable materials from the standpoint of criticality control.

The toxicity of plutonium stems from the fact that it is an alpha emitter with a half-life of 24,400 years and that it is deposited predominately in the bones and liver. It is reported to be adsorbed from the gastro-intestinal tract only to the extent of about 0.003% while from 1 to 10% of the inhaled dose may be adsorbed, depending mainly upon particle size and solubility. A small amount may be absorbed through the skin and through contaminated cuts and puncture wounds, but lung absorption is potentially the most important route of entry into the body. Once in the body, Pu-239 is excreted extremely slowly, about 200 years being required to eliminate one-half of the body burden. Table I shows maximum permissible limits which have been established for body burdens of various plutonium isotopes and permissible concentrations of these isotopes in air.

The maximum permissible body burden of 0.04 μc of Pu-239 was established by comparison with Ra-226 for which a considerable amount of clinical information exists. This is equivalent to approximately 0.6 μgm or 6×10^{-7} gms of Pu. On the basis of animal experiments, it is estimated that introduction of 20 to 70 mg of Pu-239 into systemic circulation would result in a 50% chance of death within 30 days and that an individual surviving beyond the 30 day period would surely succumb eventually to chronic or delayed effects of such a dose. Smaller doses may have a long-delayed effect, such as bone cancer. Although there are some individuals who are known to have a body-burden in excess of the presently accepted MPL, no published case involving death or even serious body damage from exposure to plutonium has come to my attention. It should be mentioned that one of the difficulties involved in working with plutonium and other α-emitters is that there is no truly continuous method of monitoring for air-borne α-material. This type of monitoring is usually achieved by pulling air through filter material at a measured rate for a period varying from 5 minutes to 8 hours or more, and then counting the α's in the paper, either with automatically activated counters or with manually operated counters. It is necessary to live with a rather high background in this type of monitoring due to the radon and thoron content of air or to allow this activity to decay overnight. The allowable concentration of air-borne plutonium is 9 $d/m/M^3$ for an eight hour working day. Other types of α-monitoring routinely carried out usually involve determining surface contamination of gloves, floors, or other surfaces either by wiping with a filter disc and counting the filter or by use of a portable alpha meter. Permissible surface levels are in the range 1 to 20 $d/m/cm^2$.

The generally accepted philosophy of plutonium handling should be obvious from the discussion of plutonium toxicity. It is, briefly, to take every precaution possible to avoid <u>any</u> exposure of operating personnel to plutonium. There are, no doubt, some rugged individualists around the country who handle plutonium successfully under less than ideal conditions, but it seems likely that increasingly firm applications of administrative controls will eventually eliminate this type of handling except possibly in emergency situations. The more nearly normal practice is to maintain complete enclosure of plutonium at all times except possibly for small quantities of plutonium in solution, which may be handled in open front hoods having a high air intake rate, approximately 150 lfm, to minimize the possibility of air-borne droplets of plutonium solution being carried into the laboratory. Company-issue clothing is always worn by people using this type of installation and the operator's hands are usually protected by disposable surgeon's rubber gloves. Many operations, particularly in analytical laboratories, can be handled much more easily in open front hoods than in glove boxes, and there seems to be no question that significant amounts of plutonium in solution can be handled in hoods by careful, competent personnel. So far as I have been able to determine, no hard and fast rules have been set up governing the amounts of plutonium that will be permitted to be handled in open front hoods. A British worker has suggested that the limit be set at 1 to 10 millicuries of plutonium in solution (approximately 15 to 150 milligrams).

Very few workers advocate handling solid plutonium in any manner other than by the total enclosure method but the method of choice varies widely with the type of equipment employed for plutonium operations and the installation where the work is done. As one writer on this subject has well said, the planning of glove box facilities is usually influenced by the planner's experiences and personal preferences. Consequently, a great deal of individuality and ingenuity has been evidenced in this field and even where commercially manufactured boxes have been employed, they were seldom used without modifications.

Some qualities considered desirable for Pu glove boxes include tightness, fire resistance, convenience of operation and decontamination, high degree of visibility, provision for safe entry and for removal of contaminated materials, and moderate cost. Some of these rquirements are, to some extent, mutually exclusive.

A wide variety of materials has been used for the construction of glove boxes, some of which were chosen for convenience in fabrication rather than for fire resistance. This fact, coupled with the plutonium fire at Rocky Flats in 1957, has resulted in a re-examination of facilities for handling plutonium at AEC installations which is still being pursued by a special glove box committee. The effects of this re-examination have, however, already been shown by stiffer controls on the use of flammable materials in glove box construction and in demands for secondary containment facilities.

Plywood has been widely used in the construction of glove boxes because of its low cost and ease of fabrication and Lucite has been a very popular window material for similar reasons. It seems likely that fewer plutonium glove boxes will be constructed of materials of this type in the future, and consequently, I shall confine my remarks to materials which are presently considered acceptable for glove box construction. Laminated glass is the preferred window material because it is more resistant to heat than existing plastics, but it is far from being an ideal construction material. It is more difficult to cut glove ports in glass panels than in plastics and great care must be exercised in attaching glass panels to glove boxes to avoid cracking the panels. Plastic window materials are available which are more fire-resistant than Lucite but some workers in this field have expressed a desire for further improvement in this area. The perfect window material has not yet been fabricated.

Stainless steel is one of the more common materials for construction because of its resistance to most corrosive atmospheres, ease of fabrication, ease of decontamination, and good structural properties. However, when hydrochloric acid must be used in the glove box it is necessary to provide a protective coating on the stainless steel surfaces and some people in this field feel that in such cases one may as well use a less expensive construction material, such as mild steel. At the Argonne National Laboratory, numerous glove boxes have been constructed by the modular approach in which a few standard steel shapes were welded together or aluminum shapes were bolted together to form glove boxes of varying sizes. Windows used in the welded boxes were attached by use of the same type of rubber molding that is used for automobile windshields. As mentioned previously, the steel surfaces are protected from corrosive atmospheres by suitable plastic coatings of paint. A less commonly used glove construction material is extruded aluminum which has good corrosion resistance and structural properties but is rather expensive. A newer and quite promising type of box construction which is still under development at Argonne is based on woven fiber glass impregnated with plastic. This type of construction apparently has no advantage, cost-wise, over stainless steel, but is very attractive for applications requiring resistance to corrosion by HCl.

Full-length gloves are commonly made of neoprene but one ORNL installation presently uses a combination plastic sleeve and ordinary rubber gloves connected by means of embroidery hoops. Glove ports, which may be either an integral part of the glove box or attached later to the front panels, depending on the type of construction, are usually made of metal or plastic. They are fabricated in such a manner that gloves can be changed without opening the box to the laboratory. The old glove or a part of the sleeve ends up inside the glove box and is discarded with the other contaminated waste. Glove ports vary in diameter from 5 in. on the "Berkely Boxes" to 10.5 in. on some ORNL glove boxes but many of the boxes now being built have glove ports 8 to 8.5 in. in diameter, which seems like a reasonable compromise between the extremes mentioned. One of the facts of life which must be kept in mind in planning glove boxes and glove box work is the average length of the human arm. For small installations this is usually accomplished by making the box small enough so that any part of the interior of the box can be reached from a single pair of gloves. In larger installations, several pairs of strategically located gloves may be required. "Free-standing" glove boxes which can be approached from all sides provide advantages for many types of operations and seem to be gaining in popularity both in this country and abroad.

Glove port covers, or interior closure plugs, make a definite improvement in the fire resistance of glove boxes when the gloves are not in use. Glove boxes for Pu work are nearly always operated at a pressure of 0.5 to 1 in. below that of the laboratory, and the air entering the glove box and leaving it must be filtered by high efficiency fire resistant filters.

I now wish to turn to the subject of glove box assemblies. Although techniques are available for safely transferring plutonium and plutonium contaminated materials into and out of glove boxes, which I shall discuss a little later, this is, in general, a time consuming operation. Whenever operations must be performed with plutonium materials which cannot all be performed in one box, it is common practice to connect several glove boxes together through connecting chambers generally referred to as interlocks. The boxes on both sides of the interlock are provided with doors so that the glove boxes need not be opened to each other during transfers. Such assemblies vary from very simple ones containing two or more interconnected boxes to very elaborate installations such as the new Fuel Fabrication Facility at the Argonne National Laboratory, in which all types of operations required for fabrication of plutonium fuel elements can be performed.

As was mentioned earlier in my talk, methods for transferring plutonium and plutonium contaminated equipment and materials into and out of glove boxes are very important. Locks of several different types are sometimes used for transferring materials into a glove box, but they find very limited application for removing materials from a glove box. It is necessary to assume that any equipment or material which has been exposed to a glove box atmosphere containing plutonium is contaminated. The most widely used method for removing such materials and plutonium samples from glove boxes is the plastic bag technique. In the previously mentioned ORNL installation, transfers are generally made through the glove ports, but it is more common to use special ports similar to glove ports for this purpose. After the contaminated material is transferred into a plastic sleeve glove or a plastic bag of suitable size, the bag is twisted and taped for several inches. A cut is then made through the center of the taped section and both ends of the cut are immediately covered with more tape. Some installations prefer to effect the sealing by means of a portable heat sealing device. A cut is made through the middle of a broad seam "sewed" in the bag at the appropriate distance above the contaminated material. Either method gives good results when properly handled but neither should be regarded as fool-proof. Disposal of contaminated material is made in a controlled area.

Just about every type of chemical and metallurgical operation that one can think of has been performed with plutonium in a glove box and in the time remaining I shall only briefly mention a few. My own work involved heating mixtures containing PuF_3 mixed with various other fluorides in a stainless steel glove box and determining the solubility of Pu in these mixtures by a filtration method. Other research performed in this box included thermal analysis studies of PuF_3 systems. In order to attain the high temperature needed for these studies, it was necessary to supplement the heat supplied by the outside furnace with heaters inside the glove box.

One of the more important operations carried out routinely with plutonium at several installations is the production of plutonium metal. A bomb technique for producing high purity plutonium metal in which PuF_4 was reduced with calcium metal with iodine as a booster in a steel bomb lined with CaO, was reported recently by a worker at Los Alamos and this method, or variations of it, seems to be the preferred method for making Pu metal at present.

Returning to the ORNL installation with which I am most familiar, operations connected with recycle of Pu for the calutron separation of Pu isotopes include evaporating dilute Pu solutions, extraction, precipitation of Pu peroxide, conversion of the precipitated oxide to PuO_2 and of the oxide to $PuCl_3$ feed material for re-introduction into the calutron.

Mound Laboratory has been engaged in a program for determining density, viscosity, and phase relations for Pu-alloy systems in support of the Los Alamos effort to develop a fast neutron reactor fueled with a Pu-alloy. A program of fundamental research in the CMB division at Los Alamos has furnished phase diagrams of $PuCl_3$ systems, thermodynamic data on $PuCl_3$, and reprocessing methods for use with Pu-alloy fuel materials, also in support of the fast reactor concept.

Metallurgical research with plutonium and plutonium alloys has been pursued at both Los Alamos and Argonne. The chemical engineering division at Argonne is investigating the volatility process for separating Pu and U and carrying out ignition experiments with plutonium. These researches, I am sure, make up only a fraction of the total effort in the field of plutonium research but they will serve to illustrate the variety of work in progress.

I have mentioned several times in my talk the emphasis that is being placed upon the use of fire resistant materials in glove box construction because the prevention of fires in plutonium facilities is generally regarded as the best method of avoiding release of material from this type of accident. Consequently, the use of flammable materials, such as solvents, should be minimized or eliminated wherever possible in glove box work with plutonium. Plutonium metal and some plutonium alloys are pyrophoric and they require special facilities such as dry air or inert atmosphere glove boxes for safe handling. Materials which will minimize availability of air, such as graphite powder, MgO or dry sand are generally recommended for fighting plutonium fires.

In conclusion, I would like to leave with you the idea that plutonium in any amount should be treated with a great deal of respect, that solid plutonium-containing materials should be handled in such a manner that they are never exposed to the laboratory air, that plutonium work should only be performed in well-planned and well-constructed facilities having adequate provisions for monitoring for escape of Pu, and that eternal vigilence is the price of safety in plutonium work.

Table I. Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Pu in Air for Occupational Exposure

Isotope	Critical Organ	Maximum Body Burden (μc)	Max. Permissible Concentration in Air for 40-hr. Week	
			μc/cc	μg/cc
Pu-239	Bone	0.04	2×10^{-12}	3.2×10^{-11}
Pu-240	Bone	0.04	2×10^{-12}	9×10^{-12}
Pu-241	Bone	0.90	9×10^{-11}	1.7×10^{-14}
Pu-242	Bone	0.05	2×10^{-12}	5.1×10^{-10}