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REVIEW OF THE PHYSICAL STABILITY AND DISPERSION CHARACTERISTICS OF UNIRRADIATED MIXED OXIDE (MOX) NUCLEAR FUEL INVOLVED IN MARITIME INCIDENTS AND ACCIDENTS

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PHYSICAL STABILITY AND DISPERSION OF MOX

SUMMARY

The nuclear industry approach to nuclear safety for the transportation of mixed oxide fuels (MOX) and other nuclear fuels rely upon the absolute containment being provided by the transportation flask. Originally designed for road and rail transportation of irradiated uranium fuels, the flasks have been adopted but not that much adapted for both sea transportation and for packaging of other radioactive consignments and, specifically considered here, MOX fuels that contain a high component of plutonium oxide.

This review considers the physical stability and dispersion characteristics of MOX during and in the aftermath of a severe accident at sea.

It is shown that the forces and, particularly, temperatures and fire duration associated with ship accidents at sea would be very challenging to the flask containment. Once a flask had failed there are a number of opportunities available for the fuel components to break up and dissociate to form a release to atmosphere. In circumstances where a high temperature fire continues to rage around the failed flask(s), which is not considered to be exceptional, the fuel cladding would be expected to fail and the body of the fuel break down and aerosolise. These extended, high temperature fire conditions could result in a significant release of all of the fuel bound radionuclides of which the plutonium could present a very significant health hazard to populations exposed to the resulting atmospheric plume.

The Review does not expand in any detail on the health consequences, although it is noted here that models developed by the United States Nuclear Regulatory Commission (NRC) assess the consequences of a 5 kilogram atmospheric release of plutonium, over land in a relatively moderately populated area (250 persons/km²) to point towards an excess total number of cancers exceeding 16,000 over a 100 year period.

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PHYSICAL STABILITY AND DISPERSION OF MOX

INTRODUCTION

Over recent years the nuclear fuel industry has increased its capacity to fabricate and supply mixed oxide (MOX) nuclear fuel. This type of fuel, comprising a blend of plutonium and depleted uranium, is being used as part replacement of the core load of nominally enriched uranium oxide fuel in a number of commercial boiling and pressurised water reactors in Europe and Japan.

Whereas uranium enriched fuels, prior to irradiation, are not considered to represent a major health hazard in the event of a release as a result of an accident during delivery to the nuclear power plant, the release of the highly radio-toxic plutonium from a delivery transportation accident involving MOX fuel could result in a very significant health detriment indeed.

For this reason the response and durability of MOX fuel during the accident and its aftermath are very important and determine the surety requirements of the transportation package and, indeed, this will reflect on the choice of transport mode and its route.

In justifying the safety case for the delivery transportation of unirradiated MOX fuel, the nuclear industry relies upon two strings of argument. First, it claims that the flasks used for the transportation of the fuel are failsafe under all reasonably foreseeable accident conditions and that, second should, inconceivably, a flask fail it defines¹ MOX fuel as a low dispersible material (LDM).

By assuming the flask to be failsafe the nuclear industry gives no cognisance to the mode of transport, so the differing extremities of accident conditions between road, rail and sea transportation are effectively discounted.²

LDM means that if the normally fully clad fuel is exposed to air, particularly where the fuel is subject to a fire environment, then the formation and release of particulate from the outer surface, or broken surfaces, of the fuel pellet will be minimal. Setting an upper limit to the release fraction in this way also sets an upper limit to the environmental impact and health detriment arising from accidents involving MOX under transportation.

Classifying the extremely radiotoxic MOX fuel as LDM influences crucial safety case factors related to the transportation and storage of MOX. For example, the claimed minimal dispersal from damaged MOX lessens the requirement for absolute surety of the transport container which means that the container itself need not be so massive and failsafe; if it is acceptable to transport MOX in lighter containers, then the mode of transportation for the present longer haul routes might switch from sea to air transport with its accompanying harsher accident impact forces and fire environment; and/or if the release fraction is claimed always to remain at a low ceiling, this might result in larger consignments or batches of MOX making up any one single shipment, whatever the transport mode

MOX FUEL

Composition

Although the nuclear industry is strident in its claim that MOX is LDM,³ it offers little

technical substantiation in support, there being just two publicly available technical papers on this. 4,5,6,7

Mixed Oxide (MOX) nuclear fuel is a blend of uranium and plutonium oxides, typically containing between 3 to 10% plutonium Pu^{239} depending on the specific design of the host nuclear reactor,⁸ with the remainder bulk being depleted uranium (U²³⁸).⁹

Manufacture and Fabrication

The oxides best suited for subsequent pressing and sintering into pellets are made from uranium/plutonium compounds precipitated from uranyl salt and plutonium nitrate (by oxalate precipitation) solutions that are subsequently reduced to the respective oxides (UO_2 and PuO_2) by firing. The oxide products have a large surface to volume ratio, low bulk and packing density, and a moderate to high oxygen to uranium/plutonium ratio.

The two oxides are thoroughly mixed with additives to produce a homogenised powder, thereafter these are blended and milled and then tumbled in a spheroidiser to produce granulated powder. During these processes a dry lubricant/conditioner (a zinc based stearate and a porosity control agent) is added. The granulated powder is milled, pressed and sintered under a reducing argon-hydrogen atmosphere to produce a sintered, fused matrix of (crystalline) ceramic dioxide in the form of individual, cylindrical pellets of approximately 20 to 25mm length and 10 to 12 mm diameter, depending on the host reactor requirements.^{10,11}

When sintering UO_2 -PuO₂ green pellets, the uranium plutonium and oxygen atoms interdiffuse to form a solid solution of UPuO₂. However, because the rate of diffusion of the metal atom is very slow (10⁻¹² cm²/s) it is likely that pure PuO₂ particles of about 30 micron diameter will not completely convert into MOX particles because of the slow rate of U-Pu diffusion. Plutonium homogeneity throughout the pellet aims to reduce plutonium agglomeration to 400 microns or less, although the UK MOX producer, BNFL, claims that in practice its product contains few agglomerates or 'clumps' as large as 20 to 30 microns.^{11,12}

The final stage of pellet manufacture is to grind the end and radial surfaces to within a final dimensional tolerance.

The final (BNFL) pellet (specified) properties are as follows:-¹²

| Geometric Density | 10.45 +/-0.17 g/cm ³ | Green pellet density $>6 \text{ g/cm}^3$ |
|--------------------|---------------------------------|---|
| Surface Roughness | 0.43 micro-radians | 0.159 micro-radians sd |
| Hydrogen Inclusion | 0.27 ppm | 0.14 sd – no in store uptake |
| Oxygen/Metal Ratio | ~2.000 | |
| Grain Size | 7.4 micron average | 0.54 micron sd |
| Sinter temperature | 1,650°C max 24 hours | 4%Hz/Ar atm + trace CO2 grain growth via Oxygen potential to limit Pu reduction to +3 valency state |
| Open Pore Density | GD-ID 0.016 W/cm^3 | Water immersion + Wetting agent |

A stack of pellets is encapsulated in a zirconium alloy sheath that is purged with helium to form a sealed fuel rod of 2 to 3 m length. These fuel rods are arranged in arrays with lightweight bracing to form fuel assemblies that are inserted into the reactor core.



MOX Characteristics - Oxidation at Elevated Temperature

The plutonium dioxide component of MOX is alpha active resulting in a degree of self-heating from absorption of the energy of the alpha particles, although equilibrium temperatures are low.¹³ In the longer term, the continuing growth of the Pu²⁴¹ daughter product Am²⁴¹ (americium) renders MOX increasingly more gamma active¹⁴ and, continuing emissions of alpha particles from both the americium and plutonium convert to helium atoms which results in a build up of pressure inside the pellet internals and cladding sheath (cladding gap) over any prolonged period of storage before loading into the reactor core.¹⁵

The melting point of the MOX refractory ceramic is approximately $2,700^{\circ}C^{16}$ but surface oxidation and comminution initiates at a significantly lower temperature of around $250^{\circ}C^{17}$ if the fuel is exposed to air.¹⁸ At relatively low temperatures exposed MOX pellets produce respirable-sized particles¹⁹ following relatively short exposure periods. For example, 1.87% of the initial mass was rendered respirable-sized particulate when MOX fuel is exposed at 430°C for 15 minutes, as compared to 0.01% at $800^{\circ}C$.¹⁷ Pulverisation at the lower temperature could result in substantial particulate release in smouldering type fires that could last for many hours.^{20,21}

MOX Characteristics - Mechanical Damage

MOX ceramic pellets are brittle and will shatter when exposed to high-energy impacts.

For example, experiments on depleted uranium fuel pellets subjected to impact energy of 0.1 J/g resulted in a release of 0.5% (of the initial mass) particles of dispersible size,^{17,22} although realistic accident impact levels would be considerably higher²³ at about 4 J/g or higher under extreme air or ship transport conditions.

PERFORMANCE OF MOX FUEL IN ACCIDENT AND POST ACCIDENT CONDITIONS

Maritime Transportation Incidents and Accidents

Transportation of MOX fuel consignments qualifies for containment within flasks compliant with the International Atomic Energy Agency (IAEA) *Safety Series 6 Type B Multilateral* requirements.

Although adopted by the International Maritime Organisation (IMO) it is generally accepted that collision and impact forces and, particularly, the duration and temperature of shipboard fires are likely to very much exceed the IAEA test conditions (\sim 13m/s or \sim 30 mph and 800°C for 30 minutes).²⁴ Ramming and collisions forces between ships at sea can invoke energy levels of around 3.10⁶kN-m with temperatures and, particularly, the duration of fires on board being significantly higher and longer than the IAEA thermal test. IMO records giving the mean duration of ship fires at sea to be about 23 hours with mean temperatures in excess of 980°C, sometimes reaching in excess of 1000°C.

Test and development work on Type B flask designs²⁵ show that the Excellox type flask could withstand a hydrocarbon fire (\sim 800°C - kerosene) of 100 minutes but that at 90 minutes the internal lead liner had completely melted and the flask commenced to expel and continued to expel for 120 minutes from the onset of the fire. The transportation flasks for MOX

consignments at sea are similar to the Excellox flask, being previously used irradiated fuel flasks adapted to receive an inner MOX fuel rack.

Once that flask surety has failed the remaining containment is limited to the fuel cladding, which comprises a thin sheath of zirconium alloy (zircalloy) forming the fuel pin that encapsulates the stack of fuel pellets. Zircalloy is not reactive at low temperatures but violently exothermic reactions occur in the region of 1,000°C, particularly in the presence of superheated steam,²⁶ evolving hydrogen which can subsequent rapidly burn or explode.

The next potential stage for dispersion of the fuel is oxidation of the uranium matrix of the fuel from UO_2 to U_3O_8 , which commences in air exposures at around $282^{\circ}C.^{27}$ Since the oxidation chemistry of plutonium is very similar to that of uranium, the same dispersal mechanisms will apply but, because the milling size of the plutonium oxide is minimal, to prevent nuclear 'clumping' within the fuel pellet, the particle size of the release may be smaller than that experience for a pure uranium fuel.^{28,29}

CONCLUDING COMMENTS AND OBSERVATIONS

The nuclear industry approach to nuclear safety for the transportation of MOX and other nuclear fuels relies upon the absolute containment being provided by the transportation flask. Originally designed for road and rail transportation of irradiated uranium fuels, the flasks have been adopted but not that much adapted for sea transportation and for other radioactive consignments, such as vitrified high level waste and, as considered here, MOX fuels that contain a high component of plutonium.

It is excepted that the forces and. particularly, temperatures and fire duration associated with ship accidents at sea could be very challenging to the flask containment and, in the event of flask failure, there are a number of opportunities available for the fuel components to break up and dissociate to form a release to atmosphere. In circumstances, where a high temperature fire continues to rage around the failed flask(s), which is not considered to be an exceptional circumstance (as chance would have it), the fuel cladding would be expected to fail and the body of the fuel break down and aerosolise. These extended, high temperature fire conditions could result in a significant release of all of the fuel-bound radionuclides of which the plutonium could present a very significant health hazard to populations exposed to the resulting atmospheric plume.³⁰

The fact that the source of the release is a ship at sea that may have been, because of the circumstances of the accident, abandoned to drift out of control means that near-coast and coastal populations are at risk from a mobile source over which they have very little control.

There is some ambiguity on whether the nuclear industry defines MOX as LDM, although BNFL certainly implied that MOX is LDM with its press release *Air Transport of Mixed Oxide (MOX) Fuel: The Facts,* 12 June 1997 which stated *"Even in a hypothetical scenario where the container was damaged to such an extent that it was split open, the MOX fuel inside would not disperse in air – it is hard, stone-like and cannot be disintegrated to a powder"* although it subsequently retracted this or a similar claim in a letter to G L Cox of 13 October 1997, noting that *"With regard to MOX and LDM, I apologise for implying that MOX fuel has in any way been officially classified as Low Dispersible Material. This was not a deliberate attempt to mislead you. BNFL, in collaboration with other European transport organisations and MOX fuel fabricators, is currently developing a reproducible and robust test regime to demonstrate LDM under funding from the EU."*



- In effect the United States will not permit air transportation of MOX fuel because it recognises higher accident conditions thereby requiring considerable upgrading of the IAEA test conditions see Ref 7.
- ³ As yet MOX has not been officially accepted as LDM.
- 4 Requirements for Very Low Dispersible Material (VLDM), TC-946, F Lange, F Nitsche, F-W Collin and M Cosack, Working Paper No 11, IAEA Technical Committee Meeting, Vienna, 15-19 May 1995
- ⁵ Contribution to Technical Committee Meeting, Working Paper 8, F Lange and F Nitsche, IAEA Vienna, 29 August 2 September 1994
- ⁶ Refs 4 and 5 do not actually specify that any of the presently manufactured MOX fuels qualify as LDM but rather set a qualification test for any radioactive substance that is to qualify for air transportation materials which must be LDM see LAEA Safety Series 6, Regulations for the Safe Transportation of Radioactive Materials which introduces the so-called Type C qualification test that applies an impact velocity of 90m/s and a 1 hour fire at 800°C. The substance qualifies as LDM if, during and following the tests, does not release an amount of activity greater than 100 times the A2 index in gaseous and particulate forms of up to 100 microns in diameter.
- ⁷ The IAEA SS6 Regulations apply as Type C qualification but the matter of LDM remains under investigation by the parties with the IAEA Transport Safety Standards Advisory Committee (TRANSACC) which recommended at its March 1997 meeting that "the new Coordinated Research Programme (CRP) on 'Accident Severity during air transport of radioactive material' be advances as planned, particularly including for the participation of the International Civil Aviation Organisation"
- The reactor in-core thermal and mechanical performances are similar to UO₂ fuel, although there is a reduced drop of reactivity with burn-up, leading to increasing fission gas release and higher rod inner pressure at the end of life of MOX fuel; also the nuclear self shielding is more pronounced and, hence greater heat is generated at the periphery of MOX fuel and this mitigates the generally poorer thermal properties of MOX fuel *In-Pile Performance of Mixed Oxide Fuel with Particular Emphasis of MIMAS Fuel*, Deramaix P and Haas D, Nuclear Technology 102, April 1993
- ⁹ For reactor grade reprocessed plutonium, at about 65% Pu-239 with Pu-238, 240, 242 isotopes forming the remainder, 7% Pu-239 content MOX is equivalent to 4.5% U-235 enrichment.
- ¹⁰ Fundamental Deficiencies in the Quality Control of Mixed-Oxide Nuclear Fuel, F Barnaby and S Burnie, Greenpeace International, Fukushima City, Japan March 2000
- ¹¹ The Microstructure Of Unirradiated SBR Max Fuel, R Eastman and S Tod, BNFL, IAEA-SM-358/9 states that all Pu rich areas are smaller than 40 microns with the greater 90% being below 25 microns and with Pu concentrations by volume being above 20% at 2%.
- ¹² MOX Fuel Manufacture at Sellafield, Edwards, J and Brennan, J, Intl Sem on MOX Fuel, Institute of Nuclear Engineers, Windermere, England, June 1996 the BNFL MOX manufacturing technique is referred to as the Short Binderless Route (SBR which incorporates a single blending route) whereas Belgonucleaire and Cogerna use the MIcronised MASterBlend (MIMAS two blending stages) for which it is claimed the agglomerates are of a size of 30 to 100 microns MOX Fuel Fabrication: A Mature Industry, Vandergheynst A and van Vliet J, Uranium Institute, 1994 Mixed-Oxide Fuel Fabrication Technology and Experience at the Belgonucleair and CFCa Plants and Further Developments for the Melax Plant, Haas D, Vandergheynst A, van Vliet J, Lorenzelli R and Nigon J, Nuclear Technology, April 1994 Elimination of the Homogenisation Step in the Manufacture of MOX Assemblies for LWRs by means of a Reactivity Equivalence Formulation, Hesketh K, Jahrestagung Kerntechnik, May 1992.
- ¹³ The equilibrium temperature is about 50°C for 4.5 kg of plutonium metal suspended in moderately static air.
- ¹⁴ There is also the decay of Pu²³⁶ into Bi²¹² and Tl²⁰⁸ yielding gamma, all at about one-fifth the rate of the Am²⁴¹ accumulation the rate of gamma activity increase depends on the 'quality' of the plutonium used and, of course, the decay series cited commence at and are dated from the in-core life of the original fuel source of the plutonium.
- ¹⁵ The helium production rate is approximately 0.3 micromole/day per kg of Pu. The Americium rate increases by approximately 0.5% pr year. Over longer storage periods the generation of helium bubbles in the fuel matrix could have a significant impact on the fissioning processes of the in reactor core fuel, although several if not many years of storage would be required for this.
- ¹⁶ As given by Ref 20 although a lower melt temperature of ~1,800°C might more realistically apply since the melting point of stoichiometric MOX (at 5% plutonium) is about 20°C below that for UO₂.
- ¹⁷ Durchfuhrung Experimenten zur Unterstutzung de Annahmen zur Freisetzung von Plutonium bei einem Flugzeugabsturz, H Seehars, D Hochrainer Franhofer-Institute, SR 0205A, March 1982
- Here it is assumed that the thermo-mechanical forces occurring during the incident are sufficient to fail the fuel transport or storage package and break through the zirconium alloy cladding.
- ¹⁹ Less than 10 µm in equivalent aerodynamic diameter.
- 20 Inadequazy of the IAEA's Air Transport Regulations: The Case of MOX Fuel, E Lyman, Nuclear Control Institute, Washington, October 1997
- ²¹ Import/Export of Irradiated Fuel and Radioactive Waste to and from the United Kingdom, Large & Associates, Greenpeace UK R1924-1, 1995
- ²² Dispersible-sized' particles are usually taken as being less than 100 microns strictly these results apply to uranium dioxide pellets, although the plutonium oxide component should particularise similarly.
- 23 The Assessment of Risks Associated with the Carriage by Air of Radioactive Material, A C Walton, Cranfield Impact Centre August 1997
- ²⁴ Import/Export of Irradiated Fuel and Radioactive Waste to and from the United Kingdom, Large & Associates, R1924, August 1995
- ²⁵ An Assessment of Acidental Thermal Testing and Analysis Procedures for Radioactive Materials Shipping Package, Pope R et al, ASME/AIChernE, July 1980 – see also, Burgess M H Thermal Trials on a Water Cooled LWR Flask, PATRAM 1983
- ²⁶ The reaction strips oxygen and liberates hydrogen thus Zr + 2H₂O > ZrO₂ + 2H₂ which liberates -9,878kJ/kg of energy (heat) the reaction is slow at 870°C but thereafter rapidly increases following a phase change of the unoxidised zirconium above 997°C.
- ²⁷ Fuel Acident, Letter of 7 November 1983 relating to PIVR Fuel Oxidation Incident of 1983 at the Battelle Columbia Laboratory, MacDonald see also Uranium Properties and Nuclear Applications, Bell J, AE (USA), 1961 and The Release of Fission Products from Zircalloy Clad UO₂ Fuel at around 1000°C, Hillary J et al, UKAEA, TRG 2433(W), 1973
- 28 Hence smaller particulate size, greater respiratory uptake and the potential for reconcentration of plutonium in the organs.

²⁹ Under these conditions plutonium differs inasmuch that the volatility of decay product americium-241 may be enhanced in reducing conditions.

Because of its long radioactive half-life (24,300 years) plutonium, remains a hazard for something like half a million years. Unlike chemical or biological hazards, plutonium is essentially impossible to destroy (except by irradiation in a fast-breeder reactor). Thus plutonium dispersed in an accident will be present in the environment essentially forever. Contaminated materials containing plutonium (PCMs), arising from clean-up operations, would need to be very securely disposed of to prevent eventual migration back to the environment over hundreds of thousands of years.

The chief hazard from plutonium derives from the alpha particles emitted during its slow but steady radioactive decay. The combined physical properties of alpha particles (large mass and diameter, double positive charge) emitted by plutonium cause large amounts of energy to be transmitted from the alpha particles to living tissues when the particles travel through human or animal bodies and until the particles are absorbed. Typically over 100,000 ionisations of atoms and molecules might be caused by one alpha particle. Each such ionisation absorbs about 35 eV of energy from the alpha particle and results in electrons being released from some molecules in the living tissues and leaving behind positively charged atoms (radicals), and being absorbed by others. This process causes changes in the chemical structure in the area of the ionisations.

Cells within about 10 microns of a plutonium-dioxide particle will be killed by this ionising radiation, whilst cells from 10 microns to 50 microns away are likely to have their genetic materials changed. Such changed cells are potential cancer cells. Cells that are not killed by the radiation may have various end results depending on how well or poorly the DNA is repaired and what sort of cell has been affected. The cells may become cancerous, weaken the body against infection (eg, in lymph nodes) or, in reproductive organs, cause birth defects. The alpha particles emitted by plutonium atoms which have lodged in bone (especially in the areas of the periosteum, endosteum and trabeculae) attack the radio-sensitive haematopoietic tissue in the bone marrow, leading to a reduction in the number of red blood corpuscles and serious effects on the body.

After study of these various effects, plutonium has generally come to be considered several times more hazardous to human health than the element radium, also an alpha-particle emitter and bone-seeker, and which itself is exceedingly toxic. This analysis is most applicable in the case of direct inhalation, which is the primary access route to the human body in the aftermath of a plutonium release accident. There is considerable doubt as to what the increased hazard factor of plutonium over radium is, though it is probably in the range of 5 to 30.

Plutonium (along with other actinides) has been linked to many forms of health injury. These include increased risks of cancer at several sites in the body, excess mortality from cancer, elevated risk of leukaemia, elevated risk of brain tumour, increased mortality from non-malignant respiratory disease, elevated risk of malignant skin cancer, increased risk of lung cancer, excess risk of gastrointestinal cancer and fibrosis of the lung, weakness against infection and micro-organisms, birth defects, and other health injuries.

The major risks to health from plutonium result from inhalation of plutonium dust, from entering the body through cuts or abrasions, and to a lesser extent via ingestion and absorption through the gastrointestinal tract. Unlike more common toxins that may be inhaled or ingested and then relatively quickly expelled from the body, plutonium is retained in the body for essentially the rest of the exposed individual's life. That is, the excretion rate of plutonium from the body takes somewhat the same mathematical function as the radioactive decay of the particles themselves. However, the base excretion period - the biological half-life over which time half the absorbed amount in an organ is excreted from the body - is typically a combination of exponential functions rather than a single exponential function as for radioactive decay half-life, making estimates of total exposure and predicted health injury even more complex. Inhalation, retention, and thereafter absorption by the human body of aerosolised plutonium (plutonium dust mixed in air with particles of other materials) is dependent on several factors including the size, shape and density of particles, and their chemical composition (including whether organic or inorganic, and the pulmonary clearance classification classes of the compounds. None of these factors can be predicted with certainty for accident situations and this will lead to further uncertainties regarding where in the body the plutonium will be retained, and how avid (long) the retention will be.

At a simple level of approximation, the hair-like cilia in nasal passages and other breathing systems within the human body filter out all particles over 10 microns in size (diameter), and some 95% of particles over 5 microns. However, particles smaller than 5 microns (or perhaps 7 microns, which is sometimes taken as the critical size) are easily absorbed through the lungs into the bloodstream and other organs. The percentage of the dust that can enter the body and be a hazard is termed the 'respirable content'.

Besides the bodily retention effects due to particulate size, the retention period in the lungs is also a function of the pulmonary clearance classification of inorganic compounds of plutonium. For example, one compound considered likely to be present in a serious MOX fuel accident where there arises a release of fine oxides of plutonium, is plutonium-dioxide. This is an insoluble compound, and has the highest (worst) classification (Class Y, avid retention) with a pulmonary biological half-life measured in years. Plutonium-dioxide exists either as a powder or as a particle attached to carbon or dust.

The empirically averaged <u>biological</u> half-lives for plutonium in the critical organs of the liver and bones are 40 years and 100 years respectively. In bone, for example, plutonium deposits on the endosteal surfaces of the mineral bone where it lodges as non-soluble phosphate compounds, and redistributes slowly through the bone volume by resorption and burial. Plutonium absorbed into bone causes malignant tumours (osteosarcomas). Once deposited in the lungs (pulmonary system), some of the plutonium will pass through the body relatively quickly (via the gastrointestinal, system), whilst other plutonium will enter the bloodstream or lymph nodes and thence pass to various organs at various rates (half-lives). Once in the bloodstream, the plutonium deposits in a variety of organs, but primarily the liver (about 45%) and skeletal system (45%). Of course, depending on its chemical form, the plutonium may be excreted more slowly or more quickly, may be absorbed into the bloodstream to higher or lower percentages, may deposit to greater or lesser degrees in the liver or bone, etc. For example, if the plutonium is present in a soluble form, it may stay in the blood longer and later deposit to a higher degree in bone.

Another critical organ of some concern regarding plutonium exposure is the gonads. Besides the known possibility of radiation-induced sterility, the possibility for transmitted genetic damage must be further investigated. At present, it is believed that the amount of absorbed plutonium (from inhalation) reaching the gonads is about 0.035% for males and 0.011% for females. The concentration of plutonium in gonads of humans exposed to fall-out has been found to be approximately 5x10-4 nanocuries per gram of gonad tissue (5x10-13 Ci/g). This is higher than the lung or bone concentration, but less than for the liver or thoracic lymph nodes. This plutonium is considered to lodge in these organs permanently, and is a cause for much concern regarding genetic damage. Moreover, even small quantities of absorbed plutonium can cause degeneration of sexual glands, more destructively even than beta-particle emitters such as strontium-90.

Absorption of ingested plutonium compounds (through food or water) into the bloodstream and thence to other organs from the gastrointestinal tract varies depending on the chemical composition, but is only on the order of 1 part in 10,000 under most conditions (eg for plutonium-239 oxide, which is highly insoluble). However, intestinal absorption is higher in children by an uncertain factor: typically the increase

is thought to be a factor of at least 100 for children under 1 year of age; 1000 times in nursing animals; or perhaps 3000 times higher based on studies on rats. Absorption is also higher if the plutonium compounds are organic.

Moreover, one body of research has found that if ingested water has been chlorinated (eg, by the water district in a treatment plant) the valancy states the determining factor for chemical reactions -of plutonium change, and this change increases the absorption through the gastrointestinal tract by as much as a factor of 1,750. The effect is probably that the presence of chlorine -a much stronger chemical reagent than oxygen will break down the insoluble plutonium-dioxide present in the water, and result in more soluble plutonium, which is much more readily absorbed by the body.