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## Cadmium Personnel Doses in an Electrorefiner Tipping Accident

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### 1. Introduction

This chapter estimates airborne cadmium concentrations caused by the facility design base earthquake (DBE) at the INL Fuel Conditioning Facility which damages the MK-IV electrorefiner (ER) vessel so that cadmium spills out onto the floor. In addition, the seismically qualified safety exhaust system (SES) is assumed to fail. The SES is a safety grade system that is large enough to keep the flow through any DBE caused breach into the cell. But with SES inoperative, failure of non -seismically qualified cell boundary penetrations allows release of cadmium vapor to the facility workers, site workers, and general public. Consequence categories are designated by estimating airborne concentrations at specific personnel locations and comparing them to applicable exposure guidelines. Without the failure of the SES, there would be negligible doses to all workers and the general public.

This scenario involves a spill of 587.4 kg of 500°C liquid cadmium from the MK-IV electrorefiner to the argon cell floor. Based on the methods and assumptions described in this chapter, the maximum potential chemical toxicity consequences of this beyond DBA release scenario are: Moderate for facility workers, low for collocated workers, low for bus staging area, and negligible for the off-site public. This safety assessment is concerned only with the cadmium release. Bounding calculations are used as required in safety assessments to assure that exposures would not be greater than calculated. Consequences of potential releases of the radiological materials caused by the DBE alone are analyzed elsewhere.

The main metabolic feature of cadmium is a long biological half-life, resulting in a virtually irreversible accumulation of this metal in the body throughout life. In non-occupationally exposed subjects, ingested and inhaled cadmium content in the body increases continuously with age. Cadmium poses a chemical toxicity hazard to kidneys, lungs, and/or liver if ingested or inhaled. Repeated or prolonged exposure to cadmium can damage target organs. Severe over-exposure can result in death.

### 2. Description of facility

The Idaho National Laboratory (INL) Fuel Conditioning Facility (FCF) uses an engineering scale pyrometallurgical process developed by Argonne National Laboratory (Till and Chang, 1989 and Till, et. al., 1997) to reprocess metallic fast reactor fuel. It is currently being used to treat fast reactor spent fuel from the Experimental Breeder Reactor II. Operations in

the facility began June 7, 1996. The process is necessary because the Experimental Breeder Reactor II fuel design uses sodium metal bonding between the fuel pin and stainless steel cladding. The use of sodium prevents the spent fuel from being suitable for direct geologic disposal. Since the Experimental Breeder Reactor II fuel is metallic rather than oxide, a pyrometallurgical process can be used for treatment rather than the traditional solvent extraction method. Since this is a nuclear facility, safety analyses of possible incidents and highly unlikely accidents are required in order to license and run such a facility. The design basis accident (DBA) for this facility is an earthquake which causes failure of pipes that go through the walls to supply argon, cell atmosphere purification, and remote operations. An exhaust system was designed and installed to mitigate this accident.

FCF is pictured in Figure 1. FCF is in the foreground with the Experimental Breeder Reactor II, located in the domed containment building, in the background. The two facilities are connected, allowing direct spent fuel transfers from the reactor to the processing facility so that spent fast reactor fuel could be processed and returned to the reactor for further power generation. Although the mission has changed and the Experimental Breeder Reactor II has been shut down and decommissioned, the spent fuel from this reactor must still be treated for long term storage.



Fig. 1. Fuel Conditioning Facility.

The entire pyrometallurgical process is conducted remotely in a hot cell environment. Figure 2 shows the FCF hot cell layout. The Air Cell, the rectangular portion, contains an air atmosphere and is used for spent fuel storage, fuel assembly disassembly into individual rods, and product storage. An operating corridor surrounds the whole facility. The Argon Cell, the annular portion, contains an inert argon atmosphere which allows metallic fuel rods to be disassembled and processed. The release of radioactive gases when the fuel rods are chopped and the exposure of heavy metal to the Argon Cell atmosphere in the form of chopped fuel or metal extracted in the refining process presents the possibility of radiological releases due to cell breaches caused by an earthquake. The vessel of the MK-IV electrorefiner shown at the top of Figure 2 is hypothesized to crack open due to the design basis earthquake causing the cadmium to spill on the floor.

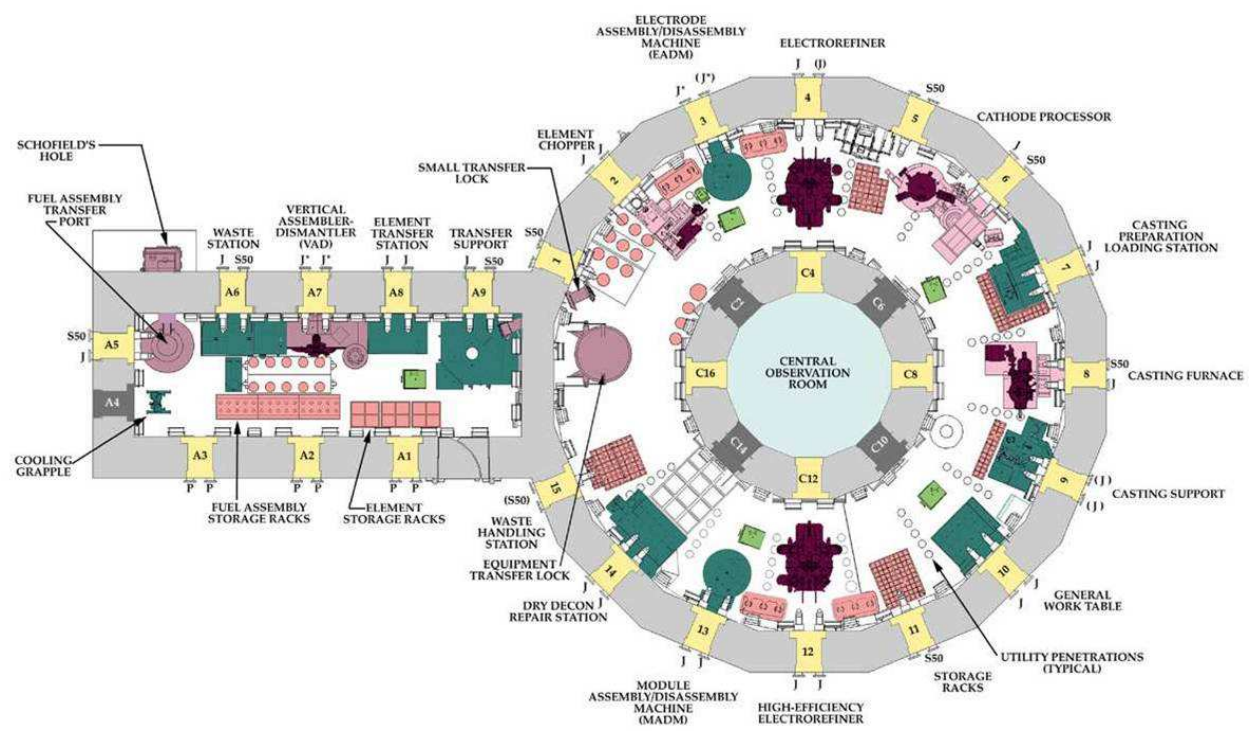


Fig. 2. Facility Layout.

The process starts with chopping spent fuel elements and loading them into baskets. Once filled, a basket (anode) is lowered into an electrorefining vessel (Figure 3) which contains molten salt (LiCl-KCl) electrolyte and in the case of the MK-IV electrorefiner, a pool of cadmium below the salt. A stainless steel mandrel (cathode) is also lowered into the salt. The basket walls are perforated allowing molten salt to contact the element segments.

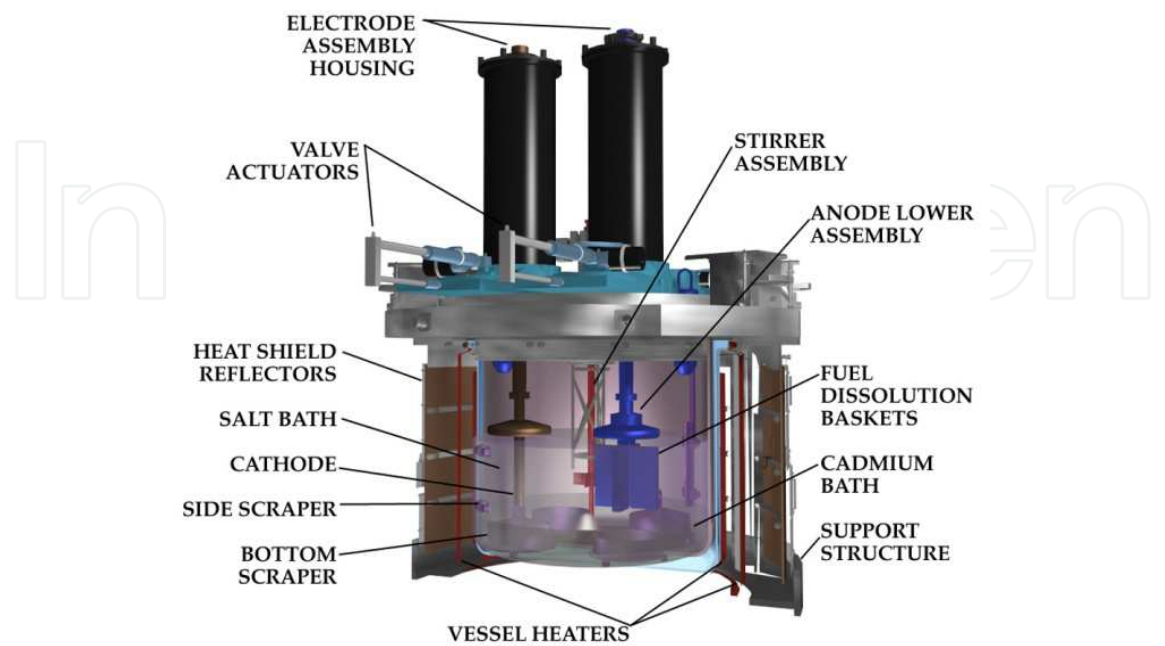


Fig. 3. Electrorefiner.



In the electrorefiner, active metal fission products, transuranic metals, and sodium metal undergo chemical oxidation and form chlorides. Voltage is applied between the basket, which serves as an anode, and a rod (mandrel), which serves as a cathode. This causes metallic uranium in the spent fuel to undergo electro-chemical oxidation, thereby forming uranium chloride and simultaneously, uranium chloride to undergoes electro-chemical reduction at the cathode depositing uranium metal dendrite onto the mandrel (Figure 4). Fission products, plutonium, and other transuranic metals remain as chlorides in the salt during this step but are removed later on.

Upon removal from the electrorefiner, the uranium metal dendrites are stripped from the mandrel. These and adhering fission product laden salt are placed in a crucible. The crucible is transferred to a distillation furnace where the uranium metal melts, forming an ingot, and the salt vaporizes under vacuum conditions and subsequently condenses in a separate crucible.

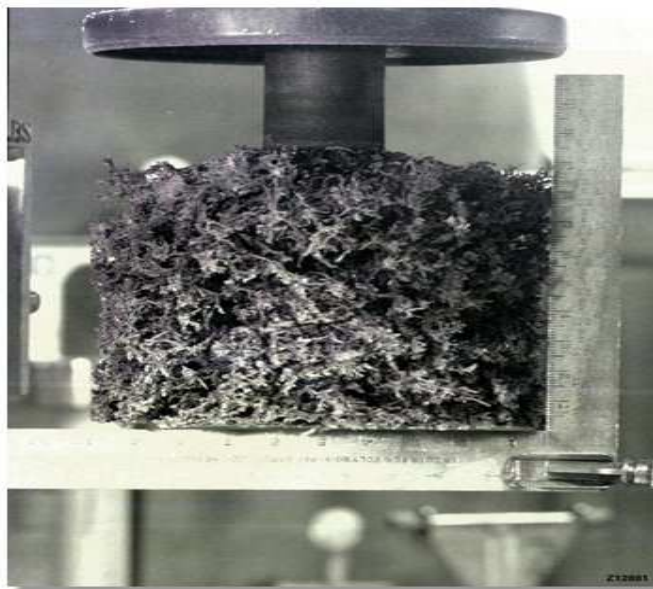


Fig. 4. Cathode Deposit.

Pyroprocessing spent metallic nuclear fuel requires the use of an inert atmosphere to maintain product purity and because pyrophoric materials, such as the cathode deposits, are produced at certain points in the process. In addition to providing an inert atmosphere, radioactive contamination and special nuclear material security concerns can be satisfied by thick concrete cell walls (1.5 m thick), a subatmospheric inert (i.e., argon) environment, remote operations, and specialized windows (1.5 m thick) to enable operations with the remote controls.

The argon cell has airborne and surface radioactive contamination and is therefore of concern for possibly releasing radioactivity. More importantly, much of the pyrophoric material is laden with radioactive fission products that can become airborne if the material begins burning. In addition, the electrorefiner contains cadmium and radioactive fission product compounds in the electrolyte salt. During normal operation, the cell pressure is kept at 745 Pa below atmospheric pressure to ensure that no radioactive material leaks out to the operating corridor which surrounds the cell or the environment.

Vertical cross sections of the circular argon cell and below grade support are shown in Figure 5. A dashed line shows the confinement boundary of the argon cell (that portion of the boundary which is not considered to fail). The boundary between the cooling cubicles and argon cell is sheet metal and is assumed to fail when the earthquake occurs if that cooling cubicle is assumed to have a breach. The failed cooling cubicles initially contain air, which is assumed to mix with the argon in the cell. The locations of the three potential breach areas are also indicated. The SES, which initiates when the cell pressure increases to  $-248 \text{ Pa g}$ , draws on the argon cell and both cooling cubicles. (Note  $248 \text{ Pa} = 1 \text{ iw g.}$ )

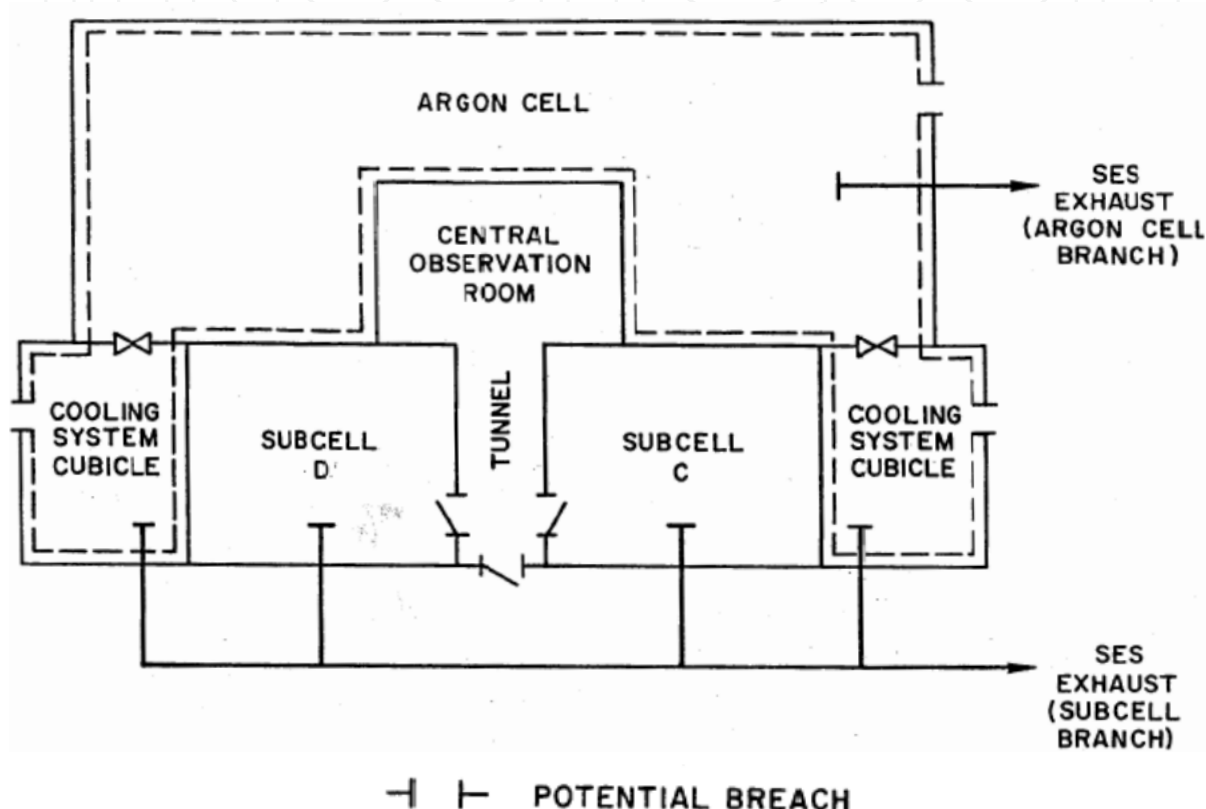


Fig. 5. Schematic of the Argon Cell.

The purpose of this chapter is to estimate airborne concentrations of cadmium at personnel locations due to damage to the electrorefiner vessel caused by an earthquake that also results in penetration of physical boundaries and to assess potential toxicological exposure. The electrorefiner operates in a radiological cell with an argon environment to prevent metal fuel reaction with air. It also prevents reaction of the cadmium with air. The accident proceeds with the electrorefiner splitting open or tipping over due to the design base earthquake. Thirty centimeters of liquid salt are on top of 15 centimeters of cadmium in a one meter diameter vessel, at  $500 \text{ C}$ . The cadmium spills out on the floor of the argon filled reprocessing cell and is assumed to be uncovered by the salt. The mass transfer of cadmium to the argon atmosphere takes place in two phases. The first phase occurs as the cadmium is spilling on the cell floor with the cadmium splashing back up and spreading on floor. The second phase occurs after the cadmium spreads out over the floor. The floor is concrete

covered by a steel liner. Cadmium evaporates from the surface until it solidifies. This chapter evaluates the mass transfer in each phase and estimates the transport and health effects to personnel in the facility, to personnel on the site, and to the general public from exposure to the cadmium.

The cooling system is assumed to trip off due to a power trip which occurs when an earthquake is sensed. Combustion cannot occur in this facility unless enough oxygen is introduced into the cell to bring the oxygen concentration above the 4% spontaneous combustion limit. The facility has a gas purification system which limits the oxygen concentration to below 50 ppm during normal operation which is far below the 4% combustion limit of metal fuel exposed to a mixture of oxygen and nitrogen or argon. A breach of the cell boundary followed by the SES initiation can cause sufficient oxygen to be introduced into the cell to cause a fire. The flow rate of the SES is large enough to limit or inhibit the unfiltered outflow of cell atmosphere. It causes the pressure to become subatmospheric by removing cell atmosphere from the cell. The SES flow is all filtered through HEPA filters before being exhausted to the atmosphere. It has the adverse effect of causing significant inflow of air into the cell in sufficient quantity to cause combustion. In order for people to be exposed to the cadmium from the rupture of the electrorefiner vessel, the SES must fail to allow outflow of the argon cell atmosphere to the operating corridor or to the outside of FCF.

### 3. Cadmium physical, chemical, and toxicity properties

Cadmium is a solid at normal atmospheric temperatures and pressures. Abundance earth's crust: 150 parts per billion by weight, 30 parts per billion by moles. Cadmium and its compounds are highly toxic. Cadmium is a soft, malleable, ductile, bluish-white metal, which is easily cut with a knife. It is an excellent electrical conductor and shows good resistance to corrosion and attack by chemicals. It is similar in many respects to zinc in its chemical properties. Cadmium tarnishes in air and is soluble in acids but not in alkalis. The metal burns in air to form brown cadmium oxide (CdO). Cadmium most often occurs in small quantities associated with zinc ores, such as sphalerite (ZnS). Greenockite (CdS) is the only cadmium mineral of any consequence. Almost all cadmium is obtained as a by-product of zinc, copper, and lead ore refining operations.

Most cadmium is used in batteries (especially rechargeable nickel-cadmium, NiCad, batteries). As a result of its low coefficient of friction and its high fatigue resistance, cadmium is used in alloys for bearings. Cadmium is used in low melting alloys and is a component of many kinds of solder. It is also used in electroplating. Compounds containing cadmium are used in black and white television phosphors, and in the blue and green phosphors for color television picture tubes. Cadmium sulfide is used as a yellow pigment, and cadmium selenide is used as a red pigment, often called cadmium red. (Chemicool, 2011).

Cadmium and tellurium can be compounded into CdTe thin-film photovoltaic modules whose physical characteristics are ideal for solar cell production. They are relatively low cost and have an almost perfect bandgap for solar energy harvesting. (Chemicool, 2011).

| Substance Name   | Cadmium                            | Cadmium Oxide                      | Cadmium Chloride                   |
|--|------------------------------------|------------------------------------|------------------------------------|
| Molecular formula  | Cd                                 | CdO                                | CdCl <sub>2</sub>                  |
| Molecular weight   | 112.41                             | 128.41                             | 183.32                             |
| Density [ g/cm <sup>3</sup> ]                            | 8.646                              | 8.15                               | 4.048                              |
| Boiling point  | 765 – 767°C<br>1,410 – 1,413°F     | 1,385 – 1,559°C<br>2,525 – 2,838°F | 956.9 – 969.6°C<br>1,754 – 1,777°F |
| Melting point  | 321°C<br>610°F                     | 1,430°C<br>2,606°F                 | 567.9°C<br>1,054°F                 |
| Vapor pressure [ mmHg ]                                  | 1 mmHg @ 394°C<br>760 mmHg @ 765°C | 1 mmHg @ 1,000°C                   | 10 mmHg @ 656°C                    |
| References:       Knovel, 2003, Patty, 2001, Perry, 1997 |                                    |                                    |                                    |

Table 1. Physical and chemical properties of cadmium and selected cadmium compounds.

**Solid Phase:** Solid cadmium is a soft, blue-white, metal or a gray-white powder. Cadmium metal dust has an auto-ignition temperature of 250°C (482°F). Cadmium is insoluble in water and soluble in ammonium nitrate, nitric acid, and hot sulfuric acid. Cadmium is combustible and burns in air or oxygen, producing cadmium oxide. In moist air, cadmium slowly oxidizes to form cadmium oxide.

**Liquid Phase;** At atmospheric pressure, cadmium metal melts at 321°C (610°F) and forms a liquid. Liquid cadmium has a relatively high vapor pressure, as shown in Table 1. A release of molten cadmium is expected to cool quickly to ambient temperatures and solidify. If the ambient atmosphere is above the melting point, 321°C (610°F), then a fraction of the material is expected to transfer to the vapor phase based on the vapor pressure of cadmium at the ambient temperature.

**Vapor Phase:** Cadmium is a vapor at atmospheric pressure and temperatures above approximately 765°C (1,410°F). In ambient air, cadmium vapor rapidly oxidizes to produce cadmium oxide. Cadmium vapor or fumes may contain elemental cadmium or cadmium oxide as particulate matter in the respirable range. In addition, cadmium vapor reacts with other typical stack gases, as shown in Table 2.

|   |                                    |        |  |
|---|------------------------------------|--------|--|
| Cadmium vapor in the presence of:                                       | ambient air                        | forms: | cadmium oxide (CdO)                      |
|   | carbon dioxide (CO <sub>2</sub> )  |        | cadmium carbonate (CdCO <sub>3</sub> )   |
|   | water vapor (H <sub>2</sub> O)     |        | cadmium hydroxide (Cd[OH] <sub>2</sub> ) |
|   | sulfur dioxide (SO <sub>2</sub> )  |        | cadmium sulfite (CdSO <sub>3</sub> )     |
|   | sulfur trioxide (SO <sub>3</sub> ) |        | cadmium sulfate (CdSO <sub>4</sub> )     |
|   | hydrogen chloride (HCl)            |        | cadmium chloride (CdCl <sub>2</sub> )    |
| These compounds may be formed in stacks and emitted to the environment. |                                    |        |  |
| Reference: Patty, 2001  |                                    |        |  |

Table 2. Compounds formed by atmospheric cadmium vapor.



**Reactions:** Combustion reaction: Cadmium metal burns in air to form cadmium oxide ( $\text{CdO (s)}$ ). Combustion processes produce very fine cadmium-containing particles. Cadmium metal does not react with water.

The overall hazards presented by cadmium are listed in the International Chemical Safety Cards (ICSC, 2005). Cadmium accumulates in the liver and kidneys and has a biological half-life, from 17-30 years in man. After uptake from the lung or the gastrointestinal tract, cadmium is transported in blood plasma initially bound to albumin. Cadmium bound to albumin is preferentially taken up by the liver. Cadmium is widely distributed in the body, with the major portion of the body burden located in the liver and kidney. Liver and kidney cadmium concentrations are comparable after short-term exposure, but the kidney concentration exceeds the liver concentration following prolonged exposure.

The concentration of cadmium in the liver of occupationally exposed workers generally increases in proportion to intensity and duration of exposures to values up to  $100 \mu\text{g/g}$ . The concentration of cadmium in the kidney rises more slowly than in the liver after exposure and begins to decline after the onset of renal damage at a critical concentration of  $160\text{-}285 \mu\text{g/g}$ .

Most non-occupationally exposed people are exposed to cadmium primarily through the diet. Cadmium can be detected in virtually all tissues in adults from industrialized countries, with greatest concentrations in the liver and kidney. Average cadmium concentrations in the kidney are at birth near zero, and rise roughly linearly with age to a peak (typically around  $40\text{-}50 \mu\text{g/g}$  wet weight) between ages 50 and 60, after which kidney concentrations plateau or decline. Liver cadmium concentrations also begin near zero at birth, increase to typical values of  $1\text{-}2 \mu\text{g/g}$  wet weight by age 20-25, then increase only slightly thereafter. (Corrosion-doctors, 2011)

Cadmium is classified as an "A2-Suspected Human Carcinogen" by the American Conference of Governmental Industrial Hygienists (ACGIH). In addition, cadmium poses a chemical toxicity hazard to kidneys, lungs, and/or liver if ingested or inhaled. Repeated or prolonged exposure to cadmium can damage target organs. Severe over-exposure can result in death. Skin contact is considered an irritant or sensitizer, and eye contact is considered an irritant.

The main routes of non-occupational exposure to cadmium are inhalation and ingestion of fumes or dust; absorption through the skin is negligible. The major routes of occupational exposure to cadmium are inhalation of dust and fumes and incidental ingestion of dust from contaminated hands, cigarettes, or food. Inhalation includes (1) inhalation of cadmium-containing particles from ambient air; (2) inhalation of cigarette smoke; and (3) occupational inhalation of cadmium fumes and dusts.

Ingestion includes (1) consuming food -- food is the major source of non-occupational cadmium exposure for nonsmokers, with the largest contribution from grain cereal products, potatoes, and other vegetables; (2) drinking water -- drinking water normally has very low levels of cadmium; and (3) incidental ingestion of cadmium-contaminated soil or dust. Table 3 lists the harmful effects of exposure to cadmium.

|   |  |
|---|--|
| Short Term (Acute) Exposure                       | Cadmium is much more dangerous by inhalation than by ingestion. High exposures to cadmium that may be immediately dangerous to life or health occur in jobs where workers handle large quantities of cadmium dust or fume; heat cadmium-containing compounds or cadmium-coated surfaces; weld with cadmium solders; or cut cadmium-containing materials such as bolts. Severe exposure may occur before symptoms appear. Early symptoms may include mild irritation of the upper respiratory tract, a sensation of constriction of the throat, a metallic taste, and/or a cough. A period of 1–10 hours may precede the onset of rapidly progressing shortness of breath, chest pain, and flu-like symptoms with weakness, fever, headache, chills, sweating and muscular pain. Acute pulmonary edema usually develops within 24 hours and reaches a maximum by three days. If death from asphyxia does not occur, symptoms may resolve within a week. |
| Long Term (Chronic) Exposure                      | Repeated or long-term exposure to cadmium, even at relatively low concentrations, may result in kidney damage and an increased risk of cancer of the lung and of the prostate.   |
| CFR, 2007,<br>29 CFR 1910.1027, Subpart Z, (2007) |  |

Table 3. Harmful effects of cadmium exposure.

4. Source term evaluation

4.1 Mass transfer during the spilling phase

The amount of cadmium droplets and vapor which is transferred to the argon gas as a result of the electrefiner cracking open and spilling the liquid cadmium may be estimated using the five-factor formula from DOE-HDBK-3010-94, (2000). This formula takes into account the material which sprays, evaporates and droplets which splash back up from the floor.

$$ST = MAR \times DR \times ARF \times RF \times LPF$$

(1)

- where ST = Source term [mass entrained in the gas phase]
- MAR = Material-at-risk = mass, spilled cadmium
- DR = Damage ratio = fraction of mass involved
- ARF = Airborne release fraction = stays in gas
- RF = Respirable fraction = fraction assumed inhaled
- LPF = Leak path factor = fraction which reaches people

The airborne release fraction, ARF, is evaluated from DOE-HDBK-3010-94, (2000) Volume 1, page 3-33 using the classification of a concentrated heavy metal solution. DOE-HDBK-3010-94 (2000), Section 3.2.3.1 discusses free-fall spill results from heights of 1 and 3 m for aqueous solutions with densities of approximately 1.0 (uranine) and approximately 1.3

(uranium in nitric acid – UNH). Section 3.2.3.1 further states that any solution containing heavy metal salts where the liquid alone has a density in excess of approximately 1.2 g/cm<sup>3</sup> is considered a “concentrated heavy metal solution” for assigning ARF and RF values and assigns a bounding ARF/RF of 2x10<sup>-5</sup>/1.0 to concentrated heavy metal solutions.

Liquid cadmium has a density of 7.78 g/cm<sup>3</sup>, a surface tension of 600 mN/m, and a viscosity of 2.5 cP (Crawley, 1972). For comparison, water at 20°C has a density of 1.0, a surface tension of 73 mN/m, and a viscosity of 1 cP. It is, therefore, reasonable to conclude that molten cadmium will have an ARF and RF more like concentrated heavy metal solutions than like aqueous solutions with densities of approximately 1.0 because it is more viscous, much more dense, and has a much higher surface tension than H<sub>2</sub>O.

Each of DR, RF, and LPF are set equal to one which is very conservative. The LPF for a specific accident should be estimated as part of physical model development for the movement of the argon gas with the entrained cadmium. For unmitigated release models, the LPF is conservatively assigned as 1.0 (the maximum value).

Ballinger and Hodgson (1986) and Sutter, et. al. (1981) state that viscosity, density, and surface tension are interrelated, and that increases in all three cause a decrease in the mass airborne since more viscous solutions require more energy to break up the liquid into microscopic particles. (It is worth noting, though, that those properties which contribute to a lower ARF also contribute to a higher RF, so that nearly all of the material that becomes airborne is respirable. This issue was addressed by conservatively setting the RF equal to 1.0.) The resulting source term is included in Table 4.

| MAR      | DR | ARF    | RF | LPF | ST <sub>Cd, spill</sub> = MAR×DR×ARF×RF×LPF |
|----------|----|--------|----|-----|---|
| 587.4 kg | 1  | 2.0E-5 | 1  | 1   | 1.175E-2 kg = 11.75 g                       |

Table 4. Source Term Parameters for a Release of Cadmium Vapor.

4.2 Evaporation mass transfer during the solidification

Experiments have shown that the spill of a liquid takes place quickly, then the liquid spreads out on the floor uniformly to form a thin liquid layer. It’s spread is limited by the surface tension of the liquid on the floor or by some obstruction which keeps it from flowing further. The area is estimated from the volume of liquid spilled and the thickness of the liquid layer. In the case of the cadmium, it spills onto a steel lined concrete floor, the high conductivity of the cadmium and the steel causes it to solidify quickly. Calculations, shown later, indicate in less than 3 seconds. The heat transfer can be represented by transient one dimensional heat transfer in the vertical direction, and the mass transfer can be estimated from mass transfer for flow over a flat plate.

The molar vapor generation rate per unit area from an evaporating pool of liquid to an ambient gas above it is a function of the saturation mole fraction of the evaporating liquid at the temperature of the liquid, the mole fraction of the contaminant in the ambient gas, and the mass transfer coefficient between the evaporating liquid and the ambient. For simplification, the properties of air are used as the ambient gas instead of argon. The flux rate by convection is given by (Bird, et. al., 1960) as

The mass transfer coefficient for flow over a smooth flat plate is taken from Reinke and Brosseau, (1997, equations 3A and 4A) and depends upon the Reynolds Number, the Schmidt Number, and the gas velocity over the liquid pool. It is given by

$$k = 0.664 \times Re_L^{(-0.5)} \times u \times Sc^{(-2/3)} \text{ for } Re_L < 15,000 \quad (3a)$$

$$k = 0.036 \times Re_L^{(-0.2)} \times u \times Sc^{(-2/3)} \text{ for } 15,000 \leq Re_L \leq 300,000 \quad (3b)$$

where  $Re_L$  = Reynolds Number [ unitless ]

$u$  = air velocity over liquid pool [ m / s ]

$Sc$  = Schmidt Number [ unitless ]

The velocity in the argon cell is estimated at 0.5 m/s. All the properties in the above expression are evaluated at standard cell conditions of  $T_{Cell} = 35^\circ\text{C}$  and  $P_{Cell} = 630 \text{ mmHg}$  (from Perry, 1997) which is the standard operating conditions of the argon cell. The value of  $L$  is taken to be 2.75 m which is the smallest value of length considered reasonable. The mass transfer coefficient varies inversely to  $L$  so the smaller value of  $L$  is conservative in producing more mass flux.

$$Re = \frac{uL\rho_{air}}{\mu_{air}} = \frac{0.5 \frac{m}{s} * 2.75m * 0.95 \frac{kg}{m^3}}{1.74 * 10^{-5} \frac{kg}{m s}} = 74,900 \quad (4)$$

$$Sc = \frac{\mu_{air}}{\rho_{air} * D_{Cd,air}} = \frac{1.74 * 10^{-5} \frac{kg}{m s}}{0.95 \frac{kg}{m^3} * 1.70 * 10^{-5} \frac{m^2}{s}} = 1.08 \quad (5)$$

Correlation 3b is chosen due to the value of Reynolds number so substituting these values into Equation 3b, the mass transfer coefficient is

$$k = 0.00181 \text{ m/s}$$

Reinke and Brosseau (1997) give two other models for estimating the mass transfer. The penetration model gives numbers that are a bit larger (10%) than the flat plate. It is much more reasonable to use a well researched correlation like the flow over a flat plate so that is used here. It is realized that the accuracy aim for developing a model for the source term is to be accurate at best by a factor of 2.

The heat balance on the cadmium should contain a loss for the vaporization. This term has been neglected in the evaluation of the heat transfer which will produce a higher temperature and a higher mass flux. So this neglect is conservative. Following is the prediction of the transient temperature of the cadmium. This is followed by an evaluation of the mass transferred to the gas phase during solidification using the temperature profile and cadmium vapor pressure.

#### 4.2.1 Cadmium transient temperature

To determine the transient temperature of the cadmium pooled on the floor of FCF, it is necessary to estimate its depth. This analysis assumes that the entire quantity of molten

cadmium present in the MK-IV ER, 587.4 kg, spills on the floor. The volume of material released,  $V_{pool}$  is calculated from mass and density (mass/volume) as:

$$V_{pool} = \frac{m}{\rho} = \frac{587.4 \text{ kg}}{7778 \frac{\text{kg}}{\text{m}^3}} = 0.0755 \text{ m}^3$$

Three different depths of the spill (pool) were considered: 1.27 cm, 1 cm, and 0.39 cm. Dividing the volume by these three depths gives the respective areas of 5.95 m<sup>2</sup>, 7.55 m<sup>2</sup>, and 19.36 m<sup>2</sup>. The deeper the pool, the longer the cadmium takes to solidify. The second depth is recommended by Reinke and Brosseau,1997. The third depth is calculated from a model presented in Solbrig and Clarksean, 1993 which determines the depth from a balance between gravity and surface tension forces to estimate when a spill will stop spreading. The first depth is taken as a conservative estimate of the thickest depth and is used to determine the temperature time and solidification time for the cadmium. The third depth produces the largest area of the spill. These two values are used in conjunction to produce the largest flux (1.27 cm) and the largest area 19.36 m<sup>2</sup> to estimate the total cadmium evaporated.

The results of the thermal analysis of the cadmium spilling directly onto the steel liner is shown in Figure 6. On the upper surface of the cadmium, the heat transfer coefficient is 8.72 W/m K convecting to the 35°C cell gas, and the emissivity is 0.8 radiating to a 35°C cell environment. The cadmium starts at 500°C, cools down and solidifies at 321°C. The cadmium thickness, 1.27 cm, is the same as the steel liner. The cadmium is completely solidified by 3 seconds.

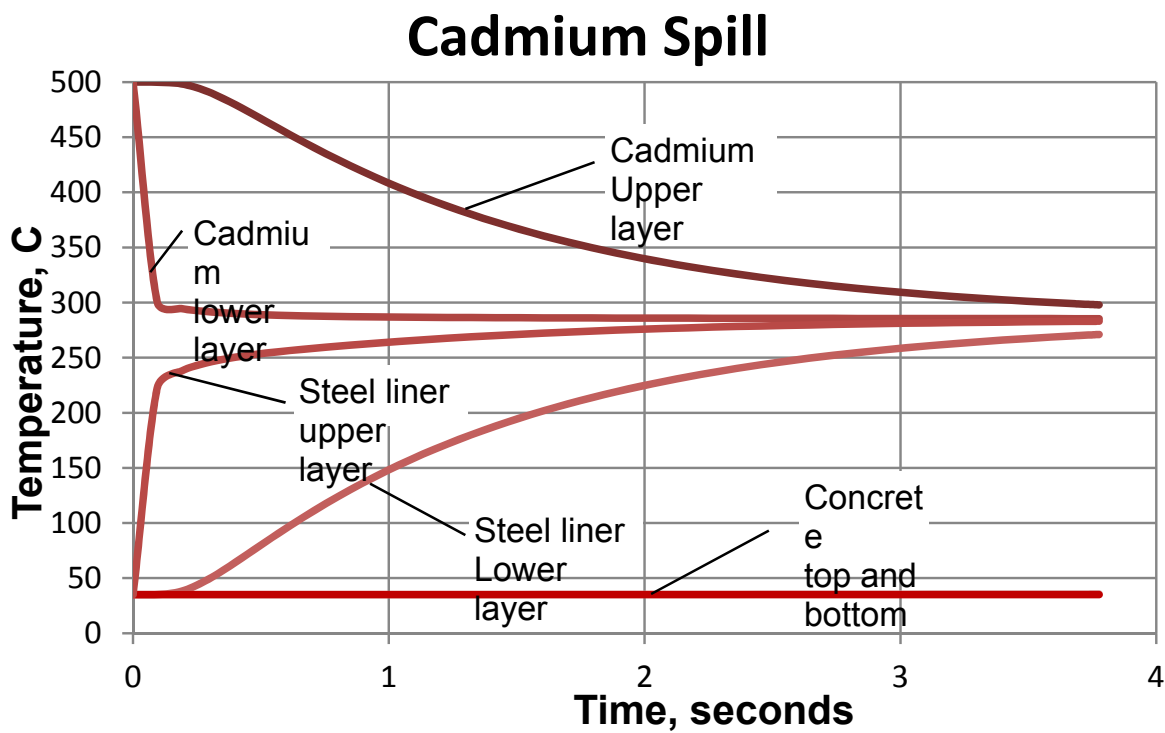


Fig. 6. Temperatures from spilling 500°C cadmium on 35°C steel-lined concrete floor

The upper surface temperature of the cadmium projects a vapor pressure which determines the mass transfer to the ambient.



4.2.2 Vapor generation determination

The vapor pressure curve for cadmium is shown in Figure 7. Due to lack of data below 420°C, the curve is conservatively approximated from 321°C to 420°C.

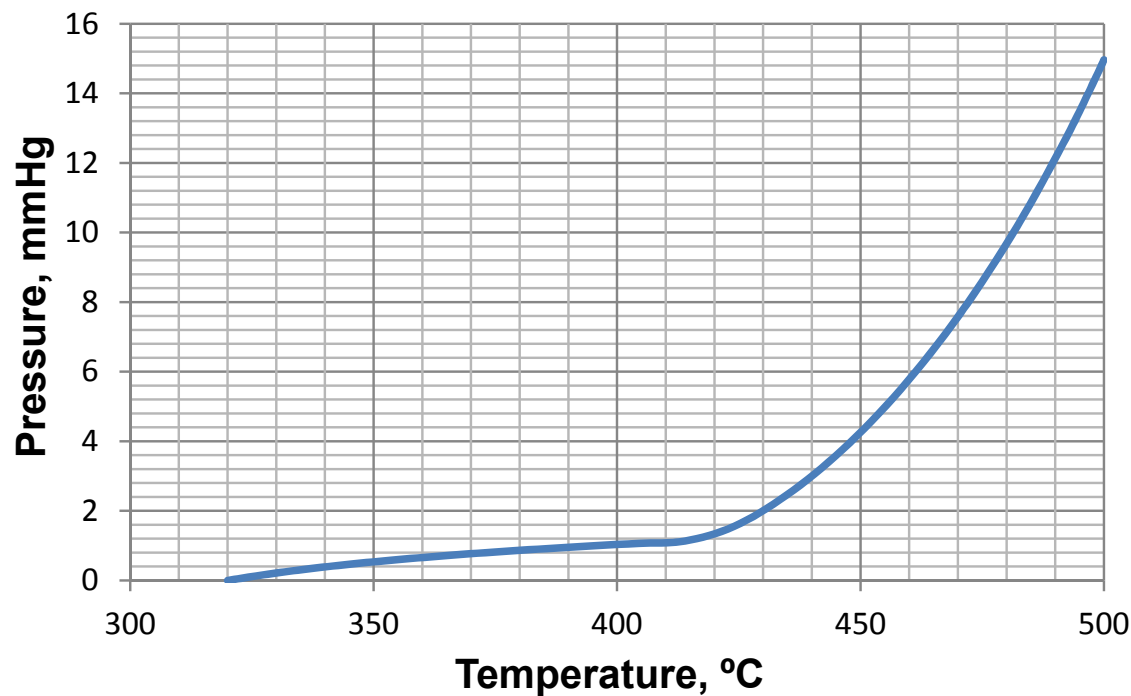


Fig. 7. Vapor Pressure of Cadmium

The temperature data in Figure 6 and the vapor pressure in Figure 7 can be combined to yield the vapor pressure of the cadmium versus time as shown in Figure 8.

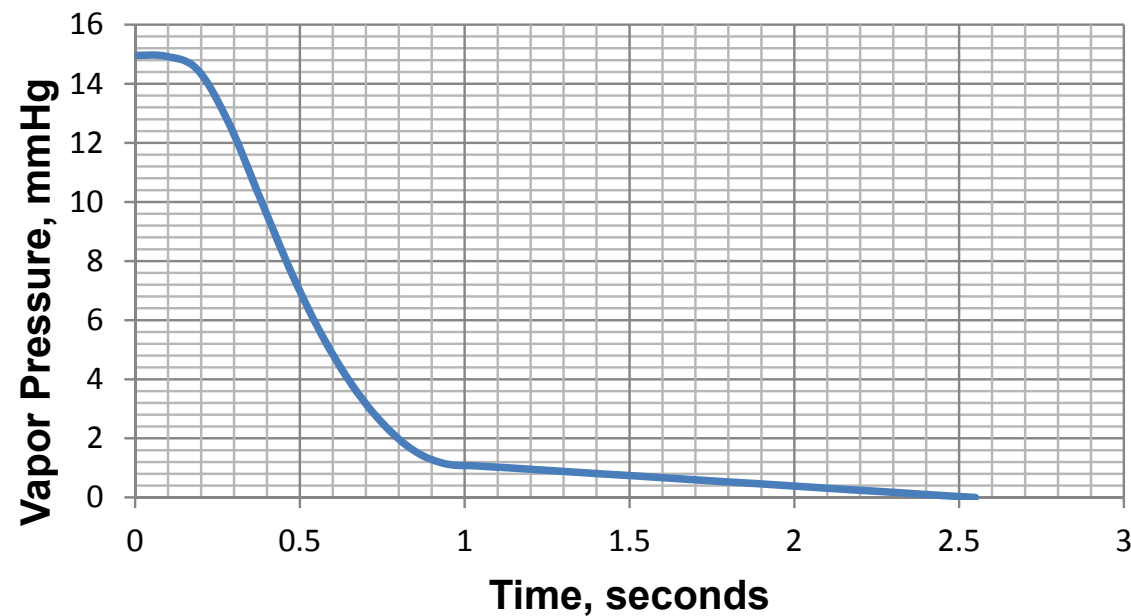


Fig. 8. Vapor Pressure during Solidification

Now, Equation 2 can be used to estimate the molar flux rate leaving the surface by utilizing Dalton’s law which states that the saturation mole fraction may be evaluated as the ratio of the saturation pressure to the ambient pressure since it is applicable when contaminant concentrations are low and pressure is near atmospheric.

$$y_{Cd,sat} = \frac{P_{Cd,sat}}{P_{cell}}$$

The mole fraction in the ambient is set equal to zero to maximize the calculated flux. Thus Equation 2 reduces to

$$\frac{dn}{dt} = k\tilde{\rho} \left( \frac{P_{Cd,sat}}{P_{cell}} \right)$$

Thus, the molar flux rate is proportional to the vapor pressure. By substituting in the values for k, into this equation, the molar flux can be determined and is shown in Figure 9.

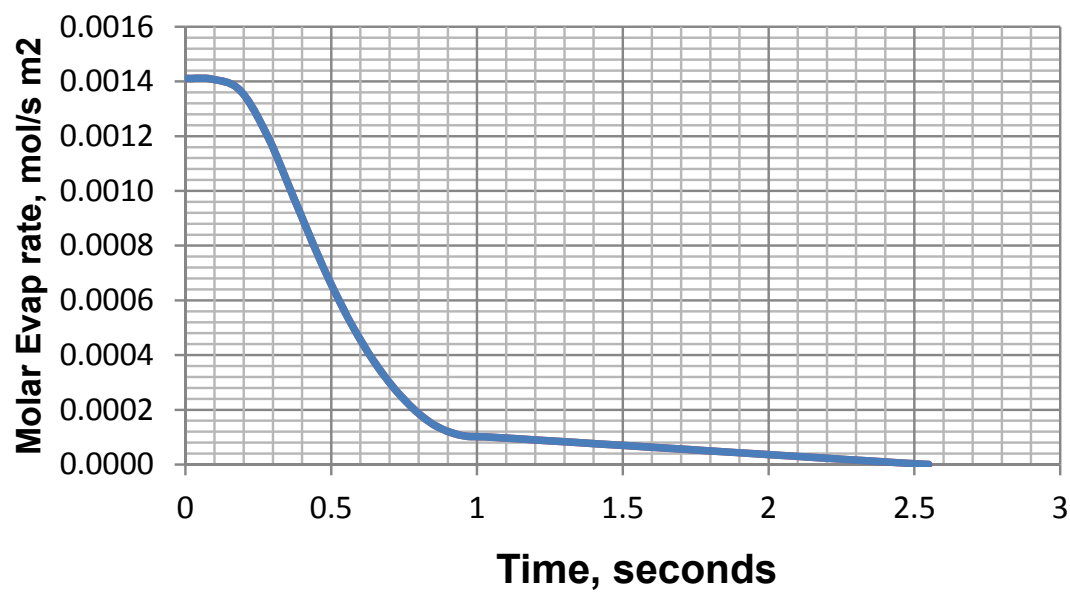


Fig. 9. Molar flux of Cadmium

The total molar flux over 3 seconds is obtained by integrating the instantaneous molar flux in Figure 9 to obtain  $7.42 \times 10^{-4} \text{ mol/m}^2$ . The mass flux is obtained by multiplying this by the molecular weight of cadmium and converting from kg to mg, to obtain  $83.5 \text{ mg/m}^2$ . The total mass over the whole surface is obtained by multiplying this by the  $19.36 \text{ m}^2$  area to obtain  $ST_{Cd,Evap} = 1620 \text{ mg}$ .

$$ST_{Cd,Evap} = 7.42 \times 10^{-4} \frac{gmol}{m^2} * 0.112 \frac{kg}{gmol} * 19.36m^2 = 1620 \text{ mg}$$

5. Evaluation of airborne concentrations

5.1 Airborne concentrations in the argon cell operating corridor

Indoor airborne concentrations are evaluated based on the source terms described in Section 4 assuming an instantaneous release with rapid mixing throughout the available

argon cell plus operating corridor volume and no subsequent losses, such as those due to leakage, ventilation, plate-out, or deposition.

The release occurs in the argon cell with subsequent release to the argon cell operating corridor through the small area opened up in the cell wall by the earthquake assuming no resistance for interchange between the cell and the corridor, the volume over which the cadmium is dispersed is the combined volume of the argon cell and the argon cell operating corridor. Assume (1) the mass vapor generation rate remains constant over the release duration; (2) rapid mixing occurs throughout the available dispersion volume; and (3) there are no subsequent losses, such as those due to leakage, ventilation, plate-out, or deposition.

$$V_D = 3,568 \text{ [ m}^3 \text{ ]}$$

Corridor Airborne Concentration for the spilling phase: The cadmium source term determined in section 4.1 is

$$C_{Cd,indoor,spill} = ST_{Cd, spill} / V_D$$

where

$C_{Cd,indoor,spill}$  = indoor airborne concentration of cadmium [ mg/m<sup>3</sup> ]

$ST_{Cd, spill}$  = airborne respirable source term of cadmium [ mg ]

$V_D$  = available dispersion volume [ m<sup>3</sup> ]

Corridor Airborne Concentration for the Evaporation Phase: The cadmium source term determined in section 4.2 is

$$C_{Cd,indoor,spill} = ST_{Cd, spill} / V_D$$

where

$C_{Cd,indoor,evap}$  = indoor airborne concentration of cadmium [ mg/m<sup>3</sup> ]

$ST_{Cd, evap}$  = airborne respirable source term of cadmium [ mg ]

$V_D$  = available dispersion volume [ m<sup>3</sup> ]

## 5.2 Outside airborne concentrations

Outside airborne concentrations are evaluated based on the source terms described in Section 4 and applicable dispersion coefficients, assuming a release of material to the outside atmosphere with no losses within the building. Thus the source term is actually used twice. Once, assuming all the cadmium stays in the FCF building. The second time, it is assumed that all the material leaves the building and is dispersed in the atmosphere. Three separate locations are tabulated for the possible dose to an individual at that location. These are:

100 m Collocated worker

280 m The distance from the FCF to the bus parking lot is 280 m (0.2 miles).

5,000 m Off-site public

The distance from the FCF to the nearest site boundary is 5,000 m (3 miles). Consequences to the off-site public are evaluated at 5,000 m (3 miles) although no one would actually park and stay at the site boundary.

**Dispersion Coefficient ( $\chi/Q$ )**An effective dispersion coefficient ( $\chi/Q$ ), based on variable parameters such as release height, distance to receptor, and receptor height, is applied when evaluating outside releases and environmental exposures. The factor ( $\chi/Q$ )describes Gaussian plume dispersion and may be hand calculated or determined using Radiological Safety Analysis Computer Program (RSAC, 2003) based on meteorological and other input parameters. The dispersion coefficient is used to estimate the quantity of contaminants at a location of interest as the result of an airborne release, based on Pasquill stability class and distance from point of release to receptor. In calculations, it is used as a reduction factor to account for the effects of transporting the contaminant from the release point to the receptor location. For the purposes of this evaluation, the maximally exposed individual (MEI) is assumed to be at the distance corresponding to the maximum dispersion coefficient. This analysis uses very non dispersive meteorology which would persist 95% or greater for outdoor dispersion estimates. The result is an extremely conservative evaluation. For this reason, an actual release scenario is expected to result in consequences less severe than those estimated in this chapter.

These calculations conservatively neglects the particle deposition that would normally occur as the material is transported from the point of release to the receptor location, which would cause a further reduction of the airborne concentration at the receptor location. Note that dispersion methodologies are not precise and provide only estimates of the consequences of an accidental release. The ( $\chi/Q$ ) values used in this evaluation were developed using INL’s Radiological Safety Analysis Computer Program (RSAC, 2003) and are listed in Table 5. Higher dispersion coefficients mean that the weather is calmer so that less dispersion takes place. For conservatism, the larger, ( $\chi/Q$ ) values obtained using the Markee method are used for all dose consequence calculations resulting in higher estimates of the cadmium exposure.

|   |  |  |  |
|---|--|--|--|
| Distance from release point to receptor                                     | 100 m                                      | 280 m                                      | 5,000 m                                    |
| $\chi/Q$ using Hilsmeier-Gifford method<br>(release duration <15 minutes)   | $3.217 \times 10^{-2}$<br>s/m <sup>3</sup> | $5.638 \times 10^{-3}$<br>s/m <sup>3</sup> | $5.697 \times 10^{-5}$<br>s/m <sup>3</sup> |
| $\chi/Q$ using Markee method<br>(15 minutes ≤ release duration ≤60 minutes) | $4.081 \times 10^{-3}$<br>s/m <sup>3</sup> | $1.309 \times 10^{-3}$<br>s/m <sup>3</sup> | $4.120 \times 10^{-5}$<br>s/m <sup>3</sup> |
| Reference:  | RSAC, 2003                                 |  |  |

Table 5. Dispersion coefficients for the Fuel Conditioning Facility

**Outside Airborne Concentration – Spilling Phase**

Outside airborne concentrations are evaluated based on the source terms described in Section 4, release duration, and applicable dispersion coefficients, assuming a release of material to the outside atmosphere with no losses within the building, and the entire quantity of material is released over the release duration (t). Although the release takes place very quickly, in order to compare it to the limits, the average rate over 60 minutes must be used.

**Outside Airborne Concentration – Spilling Phase**

$C_{Cd,outside,spill} = ST_{Cd,spill} \times (\chi/Q) / t$   
where  $C_{Cd,outside,spill}$  = outside airborne concentration of cadmium [ mg/m<sup>3</sup> ]  
 $ST_{Cd,spill}$ = airborne respirable source term of cadmium [ mg ]  
 $t$  = release duration [ s ]=15 min  
 $Q$  = dispersion coefficient [ s/m<sup>3</sup> ]

Outside Airborne Concentration -- Evaporation Phase

$C_{Cd,outside,evap} = ST_{Cd,evap} \times (\chi/Q) / t$   
where  $C_{Cd,outside,evap}$  = outside airborne concentration of cadmium [ mg/m<sup>3</sup> ]  
 $ST_{Cd,evap}$ = airborne respirable source term of cadmium [ mg ]

6. Toxicological exposure parameters

The dose guidelines that are used in this chapter are based upon Temporary Emergency Exposure Limits (TEEL) Protective Action Criteria (PAC) for exposure to the guideline concentration for 60 minutes that are defined at the DOE website (DOE, 2011). The PACs (PACs, 2009) used in this chapter are defined as follows:

**TEEL-0:** This is the threshold concentration below which most people will experience no appreciable risk of health effects. This PAC is always based on TEEL-0

**PAC-1:** The maximum concentration in air below which it is believed nearly all individuals could be exposed without experiencing other than mild transient adverse health effects or perceiving a clearly defined objectionable odor.

**PAC-2:** The maximum concentration in air below which it is believed nearly all individuals could be exposed without experiencing or developing irreversible or other serious health effects or symptoms that could impair their abilities to take protective action.

**PAC-3:** The maximum concentration in air below which it is believed nearly all individuals could be exposed without experiencing or developing life-threatening health effects.

These are:

1. The dose a person could be exposed to for an eight hour day without experiencing adverse health effects 0.005 mg/m<sup>3</sup>.
2. The maximum concentration in air below which it is believed nearly all individuals could be exposed without experiencing other than mild transient adverse health effects or perceiving a clearly defined objectionable odor. 0.03 mg/m<sup>3</sup>.
3. The maximum concentration in air below which it is believed nearly all individuals could be exposed without experiencing or developing irreversible or other serious health effects or symptoms that could impair their abilities to take protective action. 1.25 mg/m<sup>3</sup>.
4. The maximum concentration in air below which it is believed nearly all individuals could be exposed without experiencing or developing life-threatening health effects for 60 minutes. 9.0 mg/m<sup>3</sup>

A 60 minute exposure or longer to the dose of 9.0 mg/m<sup>3</sup> would be expected to produce life-threatening health effects. Based on the above guidelines, Table 6 lists airborne concentrations that bound consequence categories.

| Persons Receiving Dose | Negligible<br>if less than:   | Low<br>if less than:         | Moderate<br>if less than:   | High<br>if greater than     |
|------------------------|-------------------------------|------------------------------|-----------------------------|-----------------------------|
| All workers            | 0.03 [ mg / m <sup>3</sup> ]  | 1.25 [ mg / m <sup>3</sup> ] | 9 [ mg / m <sup>3</sup> ]   | 9 [ mg / m <sup>3</sup> ]   |
| Off-site public        | 0.002 [ mg / m <sup>3</sup> ] | 0.03 [ mg / m <sup>3</sup> ] | 1.25 [mg / m <sup>3</sup> ] | 1.25 [mg / m <sup>3</sup> ] |

Table 6. Chemical Toxicity Consequence Thresholds For Cadmium.



Since 9 mg/m<sup>3</sup> is the concentration 60 minute exposure limit for morbidity, then other exposures may be compared to the product of these two so the morbidity concentration time limit is 540 mg minutes/m<sup>3</sup>. Thus, an exposure of 1.00 mg/m<sup>3</sup> for 9 hours or an exposure of 18 mg/m<sup>3</sup> for 30 minutes would meet the criteria for life-threatening health effects as well.

7. Consequences of a vapor release due to liquid cadmium spill

The consequences are based on the DOE protective action criteria (PAC) described above, and are in the form of maximum allowable airborne concentrations that correspond to consequence levels of negligible, low, moderate and high. The evaporation phase is also very short. Heat transfer calculations show that the cadmium solidifies in less than 3 seconds. This mass is assumed to be uniformly distributed over the FCF argon cell and the operating corridor. In addition at the same time, this same mass is assumed to be released to the atmosphere to expose workers on site, workers at the bus area, and members of the people at the site boundary. This release is averaged over 15 minutes. The morbidity PAC limit of 9 mg/m<sup>3</sup> is an airborne concentration which is breathed for 60 minutes, it is really the integral of the concentration times the time increment at that concentration over time. Using an averaged value over 15 minutes is appropriate in analyzing the effects of a much higher release concentration which occurs only over 3 seconds. Applying these conservative methods result in the following doses and consequences

The airborne concentration levels are calculated in Table 7 assuming that the release occurs over a 15 minute period.

| Receptor            |  | Concentration<br>(mg/m <sup>3</sup> ) | Consequences |
|---------------------|--|---------------------------------------|--------------|
| Facility worker     | $11.75 \times 10^3 \text{ mg} \div 3568 \text{ m}^3$                                   | 3.29                                  | MODERATE     |
| On Site (100m)      | $11.75 \times 10^3 \text{ mg} / 900\text{s} \times 4.081 \times 10^{-3} \text{ s/m}^3$ | 0.0533                                | LOW          |
| Bus Area<br>(280 m) | $11.75 \times 10^3 \text{ mg} / 900\text{s} \times 1.309 \times 10^{-3} \text{ s/m}^3$ | 0.0153                                | NEGLIGIBLE   |
| Public<br>(5,000 m) | $11.75 \times 10^3 \text{ mg} / 900\text{s} \times 4.120 \times 10^{-5} \text{ s/m}^3$ | 0.00054                               | NEGLIGIBLE   |

Table 7. Consequences of Cadmium Vapor Release Due To MK-IV ER Spill – Spilling

The dose consequence of the evaporation phase is included in Table 8. . The total evaporated mass is conservatively estimated to be 1620 mg in less than 3 seconds. As in the spilling phase, the evaporation rate is average over 15 minutes.

| Receptor          |   | Concentration<br>(mg/m <sup>3</sup> ) | Consequences |
|-------------------|---|---------------------------------------|--------------|
| Facility worker   | $1620 \text{ mg} / 3568 \text{ m}^3$                                      | 0.454                                 | LOW          |
| Site worker 100 m | $1620 \text{ mg} / 900\text{s} \times 4.081 \times 10^{-3} \text{ s/m}^3$ | 0.0073                                | NEGLIGIBLE   |
| Bus Area (280 m)  | $1620 \text{ mg} / 900\text{s} \times 1.309 \times 10^{-3} \text{ s/m}^3$ | 0.0023                                | NEGLIGIBLE   |
| Public (5,000 m)  | $1620 \text{ mg} / 900\text{s} \times 4.120 \times 10^{-5} \text{ s/m}^3$ | 0.00007                               | NEGLIGIBLE   |

Table 8. Consequences of Cadmium Vapor Release Due To MK-IV ER Spill – Evaporation

The sum of the two phases is included in Table 9 along with the consequences.

| Receptor               | Concentration<br>(mg/m <sup>3</sup> ) | Concentration<br>(mg/m <sup>3</sup> ) | Concentration<br>(mg/m <sup>3</sup> ) | Consequences |
|------------------------|---------------------------------------|---------------------------------------|---------------------------------------|--------------|
| Facility worker        | 3.29                                  | 0.073                                 | 3.74                                  | MODERATE     |
| Site worker<br>(100 m) | 0.0533                                | 0.358                                 | 0.0606                                | LOW          |
| Bus Area (280<br>m)    | 0.0153                                | 0.115                                 | 0.0186                                | NEGLIGIBLE   |
| Public (5,000<br>m)    | 5.38E-04                              | 0.003                                 | 0.00061                               | NEGLIGIBLE   |

Table 9. Consequences of Cadmium Vapor Release Due To MK-IV ER Spill – Combined Spilling and Evaporation

So in conclusion, with the many conservative assumptions involved in this analysis, the consequences of this possible accident are extremely small for collocated workers and the public. For a facility worker who remains in the building for 60 minutes the exposure is only moderate, however, staying there for 180 minutes would be detrimental to his health and could cause death. Without the failure of the SES, there would be negligible doses to all workers and the general public since all releases would be through two sets of seismically qualified HEPA filters which would reduce the releases outside the building by a factor of four and eliminate the dose to the facility workers.

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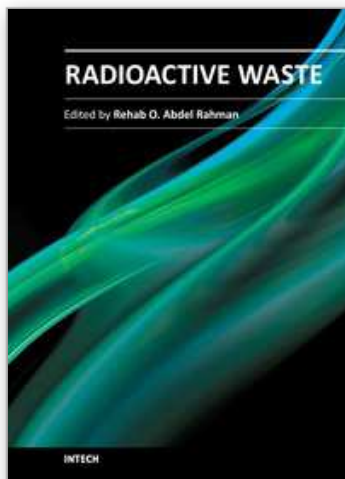
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## **Radioactive Waste**

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The safe management of nuclear and radioactive wastes is a subject that has recently received considerable recognition due to the huge volume of accumulative wastes and the increased public awareness of the hazards of these wastes. This book aims to cover the practice and research efforts that are currently conducted to deal with the technical difficulties in different radioactive waste management activities and to introduce to the non-technical factors that can affect the management practice. The collective contribution of esteemed international experts has covered the science and technology of different management activities. The authors have introduced to the management system, illustrate how old management practices and radioactive accident can affect the environment and summarize the knowledge gained from current management practice and results of research efforts for using some innovative technologies in both pre-disposal and disposal activities.

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