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1	WHC-SD-EN-TI-040		0	Carbon Tetrachloride Vapor Extraction/Possible Plutonium and Americium Contamination in the Extracted Vapor	4	2	1	

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16. KEY

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2		Cog.Eng. T. E. Moody	T. E. Moody		H4-55	EDMC			H4-22	3	6
2		Cog. Mgr. R. C. Roos	R. C. Roos	7-30-92	H4-55	~~IRA Clearance~~			~~H4-17~~	~~3~~	~~6~~
		QA				M. R. Adams			H4-55	3	6
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18. T. E. Moody, T. E. Moody 07/28/92. Signature of EDT Originator Date	19. Authorized Representative for Receiving Organization Date	20. R. C. Roos, R. C. Roos 7-30-92. Cognizant/Project Engineer's Manager Date	21. DOE APPROVAL (if required) Ltr. No. [] Approved [] Approved w/comments [] Disapproved w/comments

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Part 1

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WHC-SD-EN-TI-040

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7. Abstract

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9. Impact Level 4

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1.0 INTRODUCTION

Plutonium and americium will not be extracted by the 850 millibar or less vacuum, but will be applied to remove the carbon tetrachloride (CCl_4) in the 200 West Area as described in the expedited response action (Swanson 1991). This position will be substantiated utilizing:

- Vapor pressure characteristics of liquids, gases, and metals
- Soil plutonium and americium characterization
- The mobility of plutonium and americium, given the specific soil characteristics of the 200 West Area.

2.0 VAPOR PRESSURE CHARACTERISTICS OF LIQUIDS, GASES, AND METALS

The process of the vapor extraction technique relies on the process of vaporization (the liquid is converted to a vapor). The ability of an element or compound to enter into the vapor phase or to volatilize is dependent on the vapor pressure, which is the pressure of the vapor in equilibrium with the liquid or solid from which it originates. The vapor pressure is a characteristic property of a given liquid or solid and varies with the strength of the intermolecular forces. The vaporization process allows molecules to continually leave the substance in question until the starting substance is exhausted, exemplified in an open system or until an equilibrium is reached, and exemplified in a closed system. The vapor extraction technique emulates an open system by preventing equilibrium between the gas and the liquid. Sisson and Ellis (1990) depict the maximum vacuum to exist in the ground using the vapor extraction technique to be 850 millibar pressure, or 638 mm Hg. This vacuum is not substantial, being slightly less than atmospheric pressure, but inhibits the equilibrium between the liquid and gas, thus increasing the vaporization rate. An analogy to this is boiling water at temperatures lower than 100°C. Evacuating the volume containing the water to 24 mm Hg at a temperature of 25°C causes water to boil and vaporize more rapidly. Material with higher vapor pressures than water will evaporate quicker or vaporize more readily.

CCl_4 is characteristic of a liquid with a much higher vapor pressure than water. To obtain a vapor pressure of 760 mm Hg, a temperature of 76.7°C is required; compared to 100°C needed for water. In terms of a constant temperature (20°C), CCl_4 exhibits a vapor pressure of 90 mm Hg, and water exhibits a vapor pressure of 17.5 mm Hg.

Of the 106 known elements, 81 are classified as metals. Metals do not volatilize in the range of normal atmospheric pressures and temperatures. Mercury is the easiest to volatilize; requiring a temperature of 357°C to maintain a vapor pressure of 760 mm Hg. As an example of the low volatility of the transuranics, uranium has a melting point of 1132°C, and a temperature requirement of 3800°C to maintain a vapor pressure of 760 mm Hg. The melting points of plutonium and americium metals are 640°C and 1173°C, respectively. A temperature of 2600°C is required for americium to vaporize. Inducing

volatility of the transuranics obviously requires a substantial energy input. Comparing the temperature required for CCl_4 to maintain a vapor pressure of 760 mm Hg (76.7°C) to that of uranium (3800°C) and realizing the high melting point temperatures of plutonium and americium, it can be concluded that to induce volatility of the transuranics is quite difficult. However, it must be noted that these temperatures and pressures reference the pure metallic forms of plutonium and americium.

Price et al. (1979) indicates that the acid liquid effluent containing plutonium and americium hydrolyzed the mineral constituent in close proximity. Plutonium and americium, not complexed at the exchange sites of the soil, were involved in reactions with the alkaline earth metals released from the hydrolysis. This resulted in the oxide formation of the respective metals PuO_2 and AmO_2. Benedict et al. (1981) references the melting point of PuO_2 to be 2400°C; substantially higher than the pure metal form. Although the melting point for AmO_2 is not referenced, stability of this compound is indicated up to 1000°C. These facts further corroborate that volatility of plutonium and americium in the soil at 20 to 25°C in an open evacuated system of 638 mm Hg will not occur.

3.0 SOIL PLUTONIUM AND AMERICIUM CHARACTERIZATION

In general, ion exchangers favor the binding of ions of higher charge, decreased hydrated radius, and increased polarizability. Polarizability refers to the ability of an ion's electron cloud to be deformed by neighboring charges. The following is the Lytrophic series, which is a basic description of the preference of cations for binding to negatively charged sites (the predominant charge in soils).

$$Pu^{4+}, Am^{3+}, \gg La^{3+} > Ce^{3+} > Pr^{3+} > Eu^{3+} > Y^{3+} > Sc^{3+} > Al^{3+} \gg$$
$$Ba^{2+} > Pb^{2+} > Sr^{2+} > Ca^{2+} > Ni^{2+} > Cd^{2+} > Cu^{2+} >$$
$$Co^{2+} > Zn^{2+} > Mg^{2+} > (UO_2)^{2+} \gg Tl^{+} > Ag^{+} > Rb^{+} > K^{+} >$$
$$(NH_4)^{+} > Na^{+} > H^{+} > Li^{+}$$

Observing from the series, Pu^{4+} and Am^{3+} are more preferentially held at the exchange site and, consequently, have the highest binding energy. Once plutonium comes in contact with soil or sediment, it becomes firmly attached to the host particles. This strong attraction is exemplified by the high adsorption coefficients in laboratory studies with soils (Rhodes 1957, Prout 1958). The distribution coefficient, Kd, which is defined as the ratio of adsorbed plutonium per unit weight to solution per unit volume, ranged from about 1,000 in laboratory studies to about 100,000 in actual field situations. The high Kd in aged field situations compared to the lower Kd for short term laboratory situations indicates that with time the natural occurring soil and geochemical processes increase the retention of plutonium. The case of the high Kd would apply to the plutonium in the ground under the 200 West Area cribs. Sorption studies of americium on soils is limited compared to plutonium soil sorption.

Routsen et al. (1975) determined the Kd for an arid soil of neutral pH to be greater than 1,200. The high Kd for plutonium and americium indicate soil retention and restricted mobility.

4.0 MOBILITY OF PLUTONIUM AND AMERICIUM GIVEN THE SPECIFIC SOIL CHARACTERISTICS OF THE 200 WEST AREA

The 216-Z-1A crib in the 200 West Area received an estimated 57 Kg of plutonium, 1 Kg of americium, and unknown amounts of actinide bearing acid waste liquids (Price et al. 1979). The actinides of concern are $^{239,240}Pu$ and ^{241}Am. After 10 yr from the last documented release of actinides, the bulk of the $^{239,240}Pu$ and ^{241}Am contamination appears to be contained in the first 15 m of sediments beneath the crib, with a maximum penetration for both $^{239,240}Pu$ and ^{241}Am to 30 m below the facility. The spread was greater in the lateral direction than vertical. The lateral spread of the waste effluent was attributed to the stratification of the soil texture beneath the crib. Medium to very fine sand overlays very coarse pebbly to medium sand. Because of the unsaturated flow principle, this naturally occurring soil textural stratification acted as a natural barrier for the downward migration of the actinides. The bulk of the contamination is contained in the medium to fine sand layer, which has a particle size range of 500μm to 125μm diameter.

The plutonium that attaches itself to host soil particles has been verified directly by microscopic and alpha-track measurement techniques. Mork (1970) studied the size association of plutonium in Yucca Flat on the Nevada Test Site and showed that most of the plutonium was associated with soil particles greater than 44 μm diameter. Tamura (1975) studied soil samples from the Nevada Test Site and found that the particle sizes most closely associated with plutonium was the coarse silt fraction (50μm to 20μm diameter) and the fine sand fraction (125μm-50μm diameter). Since ^{241}Am is a decay product of $^{239,240}Pu$, and exhibits the same affinity for soil complexation as $^{239,240}Pu$, ^{241}Am will exhibit similar particle size associations. Substances with a mass light enough to be pulled from the ground during the vapor extraction are particulates that could be associated with $^{239,240}Pu$ or ^{241}Am. However, the soil textural class at the depth of maximum concentration of $^{239,240}Pu$ and ^{241}Am beneath Z crib is medium to fine sand with a particle size range of 500 μm - 125 μm diameter and a pore space diameter of about 60 μm (Brady 1984). If these particles can be physically moved by the vacuum, which is highly unlikely, either downward or lateral movement of the 500 μm to 125 μm diameter particles is impeded by the 60 μm diameter pore space of the medium to fine sand. From Sisson and Ellis (1990) an in-line high-efficiency particulate air filter system is incorporated in the design of the vapor extraction system to prevent the transport and subsequent accumulation of nondesirable substances. The high-efficiency particulate air filter system entraps 99.97% of airborne particulates > .3 μm diameter.

From the information presented in the previous sections, the conclusion is that there is little chance of outside transport via vapor extraction of $^{239,240}Pu$ and ^{241}Am in the volatile state or associated with soil particles.

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