IN SITU VITRIFICATION OF TRANSURANIC WASTES: SYSTEMS EVALUATION AND APPLICATIONS ASSESSMENT

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ABSTRACT

In situ vitrification is an emerging technology, suitable for the stabilization of radioactive waste. In just under three years in situ vitrification has moved from a concept tested in the laboratory to an achievable reality, with a series of 18 laboratory (engineering-scale) tests and 7 field (pilot-scale) tests. A radioactive make-up site of TRU wastes and mixed fission products has been stabilized with the pilot-scale system. In this test, approximately 25 kg of contaminated soil, containing about 600 nCi/g of ^{239}Pu/^{241}Am, was successfully vitrified without release of radionuclides to the environment. The ISV tests have been supplemented by public and occupational exposure calculations which show that releases fall below the limits set by the federal government for both routine and accident conditions. Performance analysis has verified the process effectiveness in terms of preventing the migration of radionuclides into the biosphere in the far-distant (1000 and 10,000 year) future under an array of intrusion scenarios including inadvertent and deliberate human intrusion. Cost analyses have shown that for both Hanford and a generic site, processing costs are less than the cost of disposal of low-level waste at U.S. Department of Energy disposal sites.

INTRODUCTION

In situ vitrification (ISV) is the conversion of contaminated soil into a durable glass and crystalline waste form through melting by joule heating. The technology for in situ vitrification is based upon electric melter technology developed at the Pacific Northwest Laboratory (PNL) for the immobilization of high-level nuclear waste. In situ vitrification was initially tested by researchers at PNL in August 1980 (U.S. Patent 4,376,598).[1] Since then ISV has grown from a concept to an emerging technology through a series of 18 engineering-scale (laboratory) tests and 7 pilot-scale (field) tests. The program has been sponsored by the U.S. Department of Energy's (DOE's) Richland Operations Office for application to Hanford sites. Additional support has been provided by the National Transuranic Waste Management Program since FY 1982.

The ISV development program is utilizing three sizes of vitrification systems. The distinguishing characteristics of each system are power level, electrode spacing, and mass of block produced, as shown below:

System	Power	Electrode Spacing	Block Mass
Engineering	30 kW	30 cm	50 kg
Pilot	500 kW	1.2 m	10 t
Large	3750 kW	5 m	175 t

The most recent pilot-scale test, completed in June 1983, vitrified a makeup site in which 25 kg of soil containing 600 nCi/g transuranic (TRU) waste simulated a highly radioactive area (or "hot spot"). The made-up source also contained mixed fission products with a total activity of 30,000 nCi/g, which exhibited a surface exposure rate of 100 R/h before it was emplaced in the test site. During the vitrification the material was distributed uniformly within an 8 t block. No radionuclides were released to the environment during the vitrification process.

With the successful completion of the radioactive test, the focus of the program has turned to the conceptual design of a large-scale system. This system is expected to be fabricated and acceptance testing completed early in FY 1985, with vitrification of actual sites planned in later fiscal years.

Major advantages of in situ vitrification as a means of stabilizing radioactive waste are:

- long term durability of the waste form
- cost effectiveness
- safety in terms of minimizing worker and public exposure
- applicability to different kinds of soils.

This paper describes ISV technology that is available as another viable tool for in-place stabilization of waste sites.

PROCESS DESCRIPTION

In situ vitrification is a process for stabilizing and immobilizing contaminated soil by vitrifying the waste and/or contaminated soil. To begin the process, which is shown in Fig. 1, graphite electrodes are inserted vertically in the ground in a square array. Graphite is placed on the surface of the soil between the electrodes to form a conductive path, and an electrical current is passed between the electrodes, creating temperatures high enough to melt the soil. The molten zone grows downward, encompassing the contaminated soil and producing a vitreous mass. Convective currents within the melt distribute the wastes evenly. During the process, off gas emitted from the molten mass is collected by a hood over the area and routed through a line to a treatment system. When power to the system is turned off, the molten volume begins to cool. The product is a block of glasslike material resembling natural obsidian. Any subsidence can be covered with uncontaminated backfill to the original grade level.

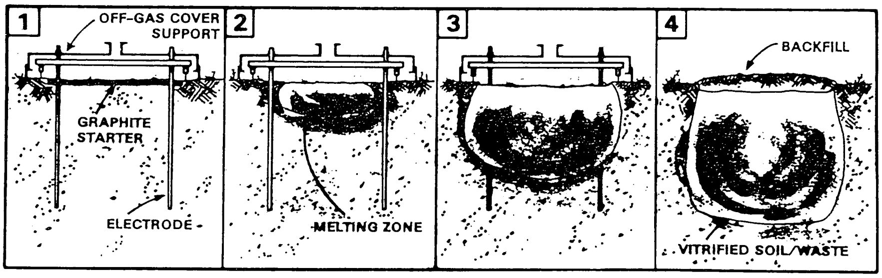

Fig. 1. In Situ Vitrification Process Sequence.

The principle of operation is joule heating, which occurs when an electrical current passes through the molten media. As this molten mass grows, resistance decreases, so to maintain the power level high enough to continue melting the soil, the current must be increased. This is accomplished by a transformer equipped with multiple voltage taps. The multiple taps allow more efficient use of the power system by maintaining the power factor (the relationship between current and voltage) near maximum. The process continues until the appropriate depth is reached. Melt depth is limited as the heat losses from the melt approach the energy deliverable to the molten soil by the electrodes.

To contain off gases that are released from the melting process, an off-gas hood that is operated under a slight vacuum covers the vitrification zone. The hood also provides support for the electrodes. The off gases are routed to a treatment system, which scrubs and filters hazardous components.

A more detailed description outlining the power system design and the off-gas treatment system for the large-scale system, the pilot-scale radioactive test system, and the engineering-scale system follows.

Power System Design

The power system design is similar for all three scales of the ISV program. A transformer connection converts three-phase alternating current electrical power to two single-phase loads. The single-phase loads are connected to two of the electrodes, which are arranged in a square pattern, creating a balanced electrical load on the secondary. The even distribution of current within the molten soil produces a vitrified product almost square in shape to minimize overlap among adjacent settings. Multiple voltage taps and a balanced load allow a near constant power operation, which shortens run time and thus minimizes cost.

Off-Gas Treatment System

In both the pilot-scale radioactive test and the large-scale systems, the off-gas containment and electrode support hood collects the off gas, provides a chamber for the combustion of released pyrolyzed organics, and supports the four electrodes embedded in the soil. Much of the heat generated during the ISV process is released to the off-gas stream. The heat is removed in the off-gas treatment system, so that the temperature of the gas which exits after treatment is close to ambient.

There are three major kinds of treatment for the off-gas system (see Fig. 2): first, the gases are scrubbed in two stages with a quencher and tandem nozzle scrubber. These scrubbers remove particles down through the submicron range. Second, the water in the saturated gas stream is removed by a vane separator and condenser followed by a vane separator, and then heated, insuring an unsaturated gas stream at a temperature well above the dewpoint. In the third stage the off gas is filtered with two stages of high efficiency particulate air (HEPA) filters. The off-gas treatment for the radioactive test system is similar to that of the large-scale system. Both systems are trailer mounted and therefore mobile.

The off gas from the engineering-scale system is treated by the air handling system of the facility in which it is located.

PERFORMANCE ANALYSIS

The ability of the waste form to retain the encapsulated or incorporated radionuclides (some with very long half-lives) is of prime importance in the usefulness of the ISV process.

Vitrified soil blocks were analyzed to determine their chemical durability with a series of tests including 24 hour soxhlet leach tests. The soxhlet leach rate for all radionuclides was less than 1×10^{-5} g/cm^2/day, an acceptable value. These rates were comparable to those of Pyrex® or granite, and much less than those of marble or bottle glass, as shown in Fig. 3.

A 28 day Materials Characterization Center test (MCC-1)[2] was also conducted on a contaminated soil sample that was vitrified in the laboratory at 1600°C. The overall leach rate of the vitrified soil is comparable to the 76-68 glass and other TRU waste forms. The release rate of Pu from the vitrified soil (2×10^{-7} g/cm^2/day) was higher than those for the borosilicate and aluminosilicate glasses. Higher vitrification temperatures like those experienced in the field are expected to lower the observed Pu leach rate.

Another indication of the durability of the ISV waste form is found in a study of the weathering of obsidian, a glasslike material physically and chemically similar to the ISV waste form.[3] In the natural environment, obsidian weathers at a rate of 1 to 20 μm^2 per 1000 years.[4] A value of 10 μm^2 per 1000 years,

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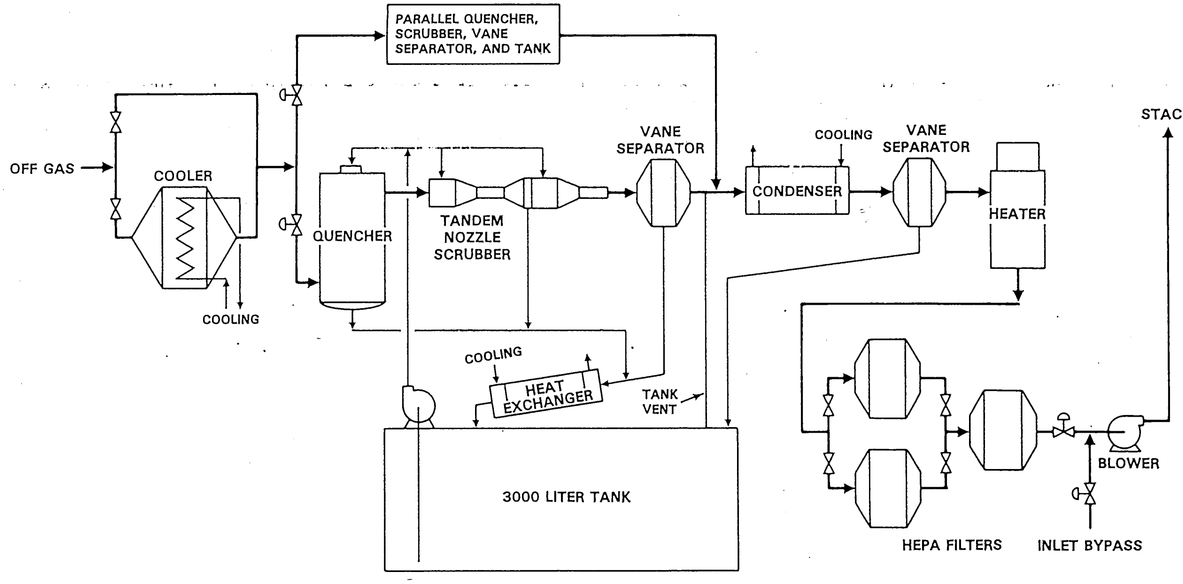

Fig. 2. Schematic for the Large-Scale Off-Gas System.

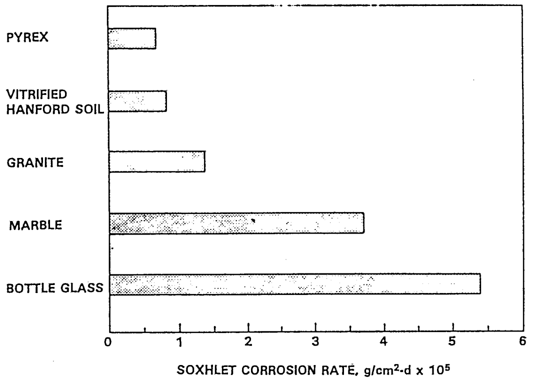

Fig. 3. Leach Resistances of Selected Materials.

assuming a linear weathering rate, yields a very conservative estimate of 1 mm of weathered ISV waste form for a 10,000 year time span.

Another important factor to consider in the waste form evaluation is the migration of the radionuclides once they are a part of the molten waste form. In the pilot-scale field tests, the radionuclides did not move beyond the vitrified block. Furthermore, analysis of the blocks from the tests revealed that the radionuclides did not concentrate in the block, but instead were uniformly distributed.

Also studied was the release of radionuclides to the off gas. The higher the decontamination factor (the mass of an element in the soil divided by the mass released to the off-gas treatment system), the less of the radionuclide that is released. Based on results from the pilot-scale system, it was estimated that for the large-scale system, soil-to-off-gas-hood DFs for less volatile elements such as Pu, Sr, and U will be 1×10^3 to 1×10^4. More volatile elements such as Cs, Co, and Te should have DFs of about 1×10^2. Element retention increases with depth of burial and the presence of a cold cap and decreases with the presence of gas generating materials. Decontamination factors for the off-gas treatment system (hood to stack) are as follows: for the semivolatiles (Cs, Co, and Te), 1×10^4, and for the less volatile nuclides Sr and Pu, 1×10^5. The soil-to-stack DFs are 1×10^6 for the semivolatiles and 1×10^8 to 1×10^9 for less volatile materials. For particulates the DFs are about 1×10^{11}. In estimating plutonium DFs, Pu should be considered a nonvolatile in TRU contaminated soil sites, and a particulate in solid waste burial sites.

PROCESS PARAMETERS

PNL studied the effects of soil properties such as chemical composition, thermal conductivity, fusion temperature, specific heat, electrical conductivity, viscosity, and bulk density on nine soils from waste sites all over the U.S. None of the minor variations in properties among the soils significantly impacts ISV operation. While soil moisture does affect the ISV process by increasing the power requirements and run time, it is not a barrier to its use, having only a small effect on the attainable melt depth. Soil moisture is an economic penalty, not a process impediment.

The effect of buried materials, particularly those that are commonly found in radioactive waste sites, has been considered. These materials include metals, cements and ceramics, combustibles, and sealed containers. While there are some limitations to the ISV process due to waste inclusions, they are not significant.

The potential for criticality due to the presence of fissionable materials has been addressed. The Pu areal limit for the ISV process is approximately 1 kg Pu per square meter of soil. Sites containing Pu levels approaching or surpassing this point should consider exhumation and recovery treatment prior to ISV as a stabilization option.

A mathematical model was devised to predict the behavior of the ISV system for waste burial sites with differing geometries and to assist in scale-up to the large-scale system without the need for extensive field testing. The effects on process performance of changes in soil properties, power system capability, and waste site geometry were evaluated using the model.

Information produced included energy consumption, mass vitrified, operating time, melt depth, and melt width for various ISV configurations. The model was also used to determine the effect of soil moisture on the ISV process. As part of the assessment of the effectiveness of the model as a predictive tool, model predictions were compared to results from the pilot-scale field tests, and the predicted and actual values were very close, with usually less than 10% variance.

ECONOMIC ANALYSIS

The cost of using ISV as an in-place stabilization technique was estimated for a TRU contaminated soil site. The components that contribute to the basic cost of ISV are site preparation activities, annual equipment charges, operational costs such as labor, and consumable supplies such as electricity and electrodes. Five different configurations for TRU contaminated soil sites, employing the large-scale system, were evaluated using the four basic cost-contributing categories. The results are provided in Table I.

When using the cost figures in Table I, it is recommended that ranges be employed for making cost estimates. For example, to estimate the cost of selectively vitrifying portions (a volume of 2900 m^3) of the 216-Z-1A site at Hanford, as shown in Fig. 4, the lower boundary of the range should be case 3 (local power, above average manpower, average heat losses): \$138/m^3, for a total cost of 2900 m^3 x \$138/m^3, or \$400,200 or \$400 K. The upper boundary of the range should be a combination of cases 1, 2, and 3 (local power, above average manpower, and high heat losses), which calculates to be 2900 m^3 x \$138/m^3 x [ratio of heat loss effects: 142 (case 1)/116 (case 2)] = \$489,900, or \$490 K.

For comparison, the charge for disposal of low level defense waste at Hanford is \$145/m^3, and the cost of placing TRU waste in 20 year retrievable storage is \$370/m^3. Estimates for exhumation, processing, certification, and emplacement at WIPP range from \$12,000 to \$25,000/m^3,[5,6] depending on the complexity of the site.

ANALYSIS OF OCCUPATIONAL AND PUBLIC SAFETY

To analyze the occupational and public safety of routine and nonroutine ISV operations for both the near and far term, a Hanford waste site (the 216-Z-1A tile field in the 200 area) was selected as a reference. Radionuclide release rates from the soil during vitrification were estimated, and the 216-Z-1A waste inventory reported by Owens[7] was the basis for the radionuclide source term.

Tables II and III give the radiation doses from routine operations in the near term for both the ISV worker and the public, respectively. For all routine exposures, radiation doses are estimated to be well below federal guidelines set by DOE. Of all activities associated with ISV operations, the maximum occupational dose is expected to occur while the worker is placing electrodes. The low exposure levels can be seen in Table II, where the occupational dose for this activity is compared to the dose that would be received during the same time period from natural background. The maximum exposed worker would receive a dose roughly the same as background radiation. The doses calculated for ISV operation at this reference site are substantially below the DOE regulations on exposure for routine operations to both workers and the general public.[8]

TABLE I Cost Estimates for Five ISV Large-Scale Configurations

Number	Site	Power	Heat Loss	Manpower Level	Total Cost of Soil Vitrified, 1982 \$/m^3
1	Hanford	Local	High	Average	142
2	Hanford	Local	Average	Average	116
3	Hanford	Local	Average	Above Avg.	138
4	Generic	Local	Average	Average	135
5	Generic	Portable	Average	Average	179

Abnormal exposures for both the ISV worker and the public from the vitrification process were calculated using worst-case scenarios. The most serious abnormal condition is a break in the off-gas line. For the specific exposures calculated, see Tables IV and V. Accidents that result in any individual receiving a total body dose >25 rem must be immediately reported, and any that result in a dose >5 rem for the total body must be reported within 72 hours.[9,10] None of the public doses from potential accidents investigated for ISV application at site 216-Z-1A falls within these categories.

Far term (beyond institutional control) exposures were calculated for transients, inadvertent intruders, intentional intruders, and permanent residents in the vicinity of the waste site. Doses were calculated for these categories for individuals 1000 and 10,000 years in the future using three scenarios: doses resulting from a site that is 1) left in its present state, 2) covered with an engineered barrier, and 3) selectively vitrified in the highly contaminated areas and covered with an engineered barrier (see Table VI). Pathways of concern were determined and evaluated using the allowable residual contamination level (ARCL) techniques described by Napier.[11] The pathways are direct irradiation; inhalation of resuspended material; and ingestion of contaminated crops, groundwater, and animal products.

For transients the undisturbed site does not pose a direct radiation hazard at the surface because the radiation levels are at background. This is true even for the unaltered site due to the shielding properties of the soil cover. However, for permanent residents the growing of gardens and eating of the garden produce could produce an exposure of 10 rem/yr from the unaltered site for both the 1000 and 10,000 year scenario. With an engineered barrier and selective vitrification, the dose would be reduced to 1 x 10^{-2} rem/yr, as shown in Table VI.

The scenarios that best distinguish the capabilities of the selectively vitrified sites from unaltered sites and barrier sites involve intentional and unintentional human intrusion (see Table VII). For a vitrified site, the worst case is drilling into the glass block because the relatively large quantity of respirable fines generated may result in the inhalation of the resuspended particles.

The worst case for the nonvitrified material with or without a barrier is excavation into the waste zone, shown in Table VII. While the barriers are designed to prevent surface erosion and plant or animal intrusion into the site, there is no way to predict whether future human populations may intrude into the waste zone. Although ISV cannot prevent an intrusion, it can mitigate its consequences.

ASSESSMENT OF WASTE SITE APPLICATIONS

Preliminary studies[12] indicate that a combination of selective vitrification and appropriately scaled

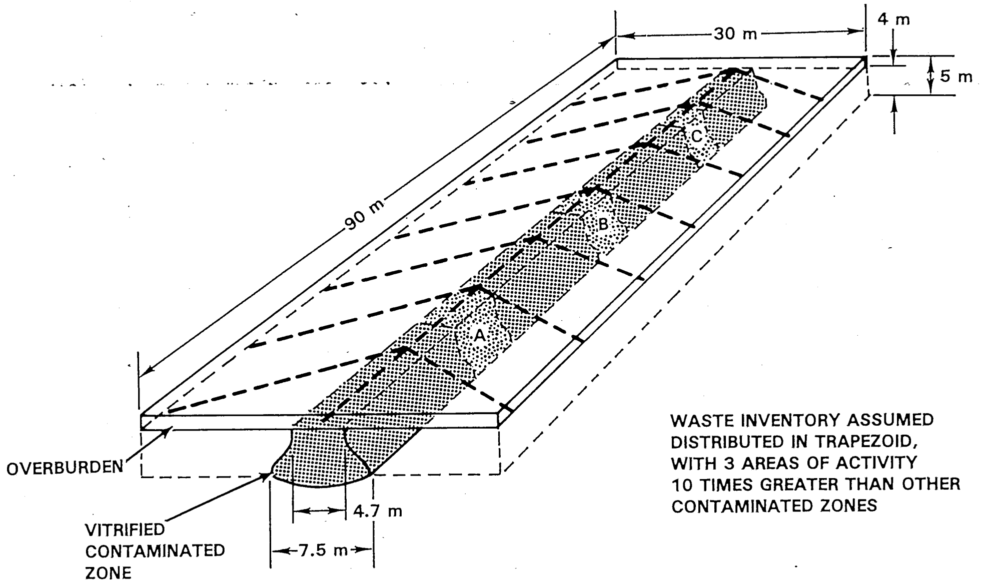

Fig. 4. Waste Distribution in a 90 x 4 x 4 m Site.

TABLE II. Whole Body Radiation Doses from Routine Operations

Occupational Dose (Electrode Emplacement)	
All Workers:	0.09 man-rem
Maximum worker:	0.02 rem
Natural Background	
Dose Rate:	7 μR/h
Total (1880 h):	0.01 rem

TABLE III. Public Dose Commitments from Routine Operations (critical organ)

	Max. Exposed Indiv., rem	Population, man-rem
1st yr dose (lungs)	3×10^{-8}	9×10^{-3}
50 yr dose (bone)	1×10^{-5}	5×10^{-1}

TABLE IV. Occupational Doses from Accidental Releases; 120 Hour Run, 15 Sets (concentrated inventory)

Accident	Number of Personnel	Length of Exposure	1st Year Dose Commitment to Each Worker, rem Total Body	 Bone	 Lung
Uncontrolled venting	1	1 min	1×10^{-3}	2×10^{-2}	2×10^{0}
Break in off-gas line	1	5 min	6×10^{-3}	1×10^{-1}	1×10^{1}
Excess overburden removal	2	10 min	3×10^{-3}	4×10^{-2}	5×10^{0}

barriers may be the most cost effective in-place stabilization technique for those Hanford TRU sites requiring remedial action. This approach is consistent with the findings of the National Academy of Sciences,[13] which stated that retrieval of TRU contaminated soils for disposal in a geologic repository could be more hazardous then disposing the waste in place. This approach is also consistent with the long-range master plan for defense transuranic waste management,[14] which states that "deep geologic disposal may not be the most economical means of safe disposal for all TRU wastes." DOE Order 5820.1[15] allows field organizations to establish new or alternative TRU waste management practices. ISV is one of the engineered permanent disposal alternatives being examined for greater confinement than shallow land burial, also referred to as greater confinement disposal.

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TABLE V. Public Dose Commitments from Postulated Abnormal Occurrences

	Maximum Exposed Individual, rem	Population, man-rem
Uncontrolled Venting		
1st year (lungs)	5×10^{-5}	2×10^{-1}
50 year (bone)	5×10^{-4}	2×10^{0}
Off-Gas Line Break		
1st year (lungs)	3×10^{-2}	1×10^{2}
50 year (bone)	3×10^{-1}	1×10^{3}
Excessive Overburden Removal		
1st year (lungs)	1×10^{-2}	3×10^{1}
50 year (bone)	9×10^{-2}	3×10^{2}

TABLE VI. Public Dose Estimates for Far-Term Routine Scenarios

	Year 1,000	Year 10,000
Direct Irradiation	Maximum Annual Total Body Dose, rem	
Unmodified site	Background	Background
Vitrification and engineered barrier	Background	Background
Engineered barrier	Background	Background
Ingestion	Maximum Annual Bone Dose, rem	
Unmodified site	10	10
Vitrification and engineered barrier	0.01	0.01
Engineered barrier	0.01	0.01

TABLE VII. Public Dose Commitments for Far-Term Intrusion, rem

	Year 1,000		Year 10,000	
	1st Year[a]	50 Year[b]	1st Year[a]	50 Year[b]
Drilling				
Unmodified site	24	880	16	550
Vitrification and engineered barrier	6	15	3	8
Engineered barrier	24	880	16	550
Excavation				
Unmodified site	48	1600	33	1100
Vitrification and engineered barrier	0.006	0.02	0.003	0.008
Engineered barrier	48	1600	33	1100
Vitrified curio[c]	0.2	10	0.1	5

(a) Lung dose.
(b) Bone dose.
(c) Total body dose.

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