EVALUATION OF SHIPPING DOSES AND COMPOSITIONS FOR VITRIFIED WASTE

by

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ABSTRACT

Shipments of radioactive materials must adhere to dose limits specified in the Code of Federal Regulations. This paper discusses methods for evaluating shipping doses of vitrified waste. A methodology was developed for evaluating the change in vitrification composition required to maintain shipping dose rates within limits. The point kernel codes QAD and Microshield were used to evaluate dose equivalent rates from specified waste forms and radioactivity measurements. The Origen code was utilized to provide the gamma-ray activity as a function of time from isotopic activity measurements. This gamma-ray activity served as source input for QAD. Microshield developed its own source from the given isotopic activities.

I. INTRODUCTION

The Fernald Environmental Restoration Management Corporation (FERMCO) is planning to vitrify waste stored in silos at the Fernald site in Ohio, and ship the vitrified waste to a disposal site in Nevada. The amount of waste is over 200,000 cubic feet in volume, so the vitrification and shipping process will be a major undertaking. Shipping must adhere to federal regulations which specifies maximum surface doses on the shipping configuration. The activity levels in the silo waste is expected to vary, so that the vitrification composition must change to accommodate increases in silo activity while maintaining shipping dose limitations. This paper discusses methods for evaluating shielding requirements and additive dilution in the vitrification compositions to maintain shipping limits.

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II. SOURCE TERM

A. FERMCO MEASUREMENTS

Radioactive isotopes and average activities in the silo material have been identified and measured by FERMCO. The measured activities indicated that the main radioactivity chain in the silo is associated with the Uranium-238 series, with some additional isotopes from the U-235 series and the Thorium-232 series. The measured activities are given in Table II.1. Uranium had been extracted from the original ore, leaving the residue of isotopes. The most active isotope is Radium-226. Radium-226 decays to Bismuth-214 and Lead-214, both of which provide troublesome decays.

B. ORIGEN CODE CALCULATIONS

The Origen code\(^1\) will calculate the decay and buildup of radioisotopes from a given source. The source values of Table II.1 were converted to mass and used as input for Origen decay calculations.

Gamma-ray release rates for 18 energy groups as a function of time are provided by Origen. These values are used to establish the source term for shielding calculations. The most important isotopes for each group are shown in Table II.2, along with the percentage of the total dose as calculated by Origen. The isotopes Pb-214 and Bi-214 are very significant contributors to the total.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>(T_{1/2}) Yrs.</th>
<th>pCi/g</th>
<th>Chain</th>
<th>Isotope</th>
<th>(T_{1/2}) Yrs.</th>
<th>pCi/g</th>
<th>Chain</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ac-227</td>
<td>21.77</td>
<td>7670</td>
<td>(^{239})U</td>
<td>Ra-226</td>
<td>1620</td>
<td>477000</td>
<td>(^{238})U</td>
</tr>
<tr>
<td>Bi-214</td>
<td>3.8-5</td>
<td>441000</td>
<td>(^{239})U</td>
<td>Th-228</td>
<td>1.91</td>
<td>7360</td>
<td>(^{232})Th</td>
</tr>
<tr>
<td>Pa-231</td>
<td>32760</td>
<td>4040</td>
<td>(^{239})U</td>
<td>Th-230</td>
<td>77000</td>
<td>76200</td>
<td>(^{231})U</td>
</tr>
<tr>
<td>Pb-210</td>
<td>22.26</td>
<td>202000</td>
<td>(^{239})U</td>
<td>Th-232</td>
<td>1.4+10</td>
<td>1100</td>
<td>(^{232})Th</td>
</tr>
<tr>
<td>Pb-211</td>
<td>6.9-5</td>
<td>19000</td>
<td>(^{239})U</td>
<td>U-234</td>
<td>244500</td>
<td>1160</td>
<td>(^{239})U</td>
</tr>
<tr>
<td>Pb-214</td>
<td>5.1-5</td>
<td>438000</td>
<td>(^{239})U</td>
<td>U-235</td>
<td>7.0+8</td>
<td>94</td>
<td>(^{235})U</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(^{239})U</td>
<td>U-236</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Po-210</td>
<td>0.379</td>
<td>281000</td>
<td>(^{218})U</td>
<td>U-238</td>
<td>4.5+9</td>
<td>1120</td>
<td>(^{238})U</td>
</tr>
<tr>
<td>Ra-223</td>
<td>0.031</td>
<td>16000</td>
<td>(^{218})U</td>
<td>Total</td>
<td></td>
<td>1972754</td>
<td></td>
</tr>
</tbody>
</table>

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Table II.2
Significant Isotopes and Percent of Total Dose

<table>
<thead>
<tr>
<th>Gamma Ray Group</th>
<th>Energy MeV</th>
<th>Isotope</th>
<th>Percent of Total Dose Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.01</td>
<td>Pb-210, Pb-211, Pb-214, Bi-210, Bi-214, Ra-223, Ra-226</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>0.025</td>
<td>Pb-211, Pb-214, Bi-210, Bi-214</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>0.038</td>
<td>Pb-211, Pb-214, Bi-210, Bi-214</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td>0.058</td>
<td>Pb-210, Pb-211, Pb-214, Bi-210, Bi-214, Th-230</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>0.085</td>
<td>Pb-212, Pb-214, Bi-210, Bi-214, Ra-223, Ra-226</td>
<td>0</td>
</tr>
<tr>
<td>6</td>
<td>0.125</td>
<td>Pb-211, Pb-214, Bi-210, Bi-214, Ra-223, Th-230</td>
<td>2.23-3</td>
</tr>
<tr>
<td>7</td>
<td>0.225</td>
<td>Pb-212, Pb-214, Bi-210, Bi-214, Ra-223, Ra-226</td>
<td>0.83</td>
</tr>
<tr>
<td>8</td>
<td>0.375</td>
<td>Pb-214, Bi-214</td>
<td>3.9</td>
</tr>
<tr>
<td>9</td>
<td>0.575</td>
<td>Tl-208, Pb-211, Pb-214, Bi-214</td>
<td>13.73</td>
</tr>
<tr>
<td>10</td>
<td>0.850</td>
<td>Pb-211, Pb-214, Bi-212, Bi-214</td>
<td>6.34</td>
</tr>
<tr>
<td>11</td>
<td>1.25</td>
<td>Pb-211, Bi-214</td>
<td>27.31</td>
</tr>
<tr>
<td>12</td>
<td>1.75</td>
<td>Bi-214, Ra-223, Ra-226, Th-228, Th-230</td>
<td>33.22</td>
</tr>
<tr>
<td>13</td>
<td>2.25</td>
<td>Bi-214, Ra-223, Ra-226, Th-228, Th-230, U-238</td>
<td>13.41</td>
</tr>
<tr>
<td>14</td>
<td>2.75</td>
<td>Tl-208, Bi-214, Ra-223, Ra-226, Th-228, Th-230, U-238</td>
<td>1.19</td>
</tr>
<tr>
<td>15</td>
<td>3.5</td>
<td>Bi-214, Ra-223, Ra-226, Th-228, Th-230, U-238</td>
<td>0.66</td>
</tr>
<tr>
<td>16</td>
<td>5.0</td>
<td>Po-210, Po-214, Po-218, Rn-222, Ra-223, Ra-226, Th-228, Th-230</td>
<td>0</td>
</tr>
<tr>
<td>17</td>
<td>7.0</td>
<td>Po-210, Po-214, Po-218, Rn-222, Ra-223, Ra-226, Th-228, Th-230</td>
<td>0</td>
</tr>
<tr>
<td>18</td>
<td>9.5</td>
<td>Po-210, Po-214, Po-218, Rn-222, Ra-223, Ra-226, Th-228, Th-230, U-238</td>
<td>0</td>
</tr>
</tbody>
</table>
III. COMPOSITION OF VITRIFIED MATERIAL

The composition of the vitrified material must be broken down into its elemental form for input to shielding codes. The composition includes the silo material mixed with a vitrification additive. FERMCO measurements of the silo material provide the weight fraction of compound or element in the silo material. In addition, the additive composition is given. For an assumed additive weight fraction, the elemental weight fractions in the vitrified mixture is given by

\[ WF_i = \frac{n_iA_i}{\sum_{j=1}^{J} n_jA_j} WF_C(1-WF_{ADD}) \]

where,
- \( WF_i \) = Weight fraction of element i in vitrified mixture
- \( n_iA_i \) = number of atoms of i in the compound multiplied by atomic weight of i
- \( n_jA_j \) = number of atoms of j in the compound multiplied by atomic weight of j
- \( J \) = number of elements in compound
- \( WF_C \) = Weight fraction of compound in silo material
- \( WF_{ADD} \) = Weight fraction of additive in vitrified mixture

The total weight fraction of element i is then obtained by summing over all the individual weight fractions of element i in the mixture. To obtain the number density of a given element, the vitrified mixture density is calculated. The mixture density is obtained from

\[ \rho_{MIX} = \frac{1}{WF_{ADD}(1-WF_{ADD})} (1-VF) \]

where,
- \( \rho_{MIX} \) = Mixture Density
- \( \rho_{ADD} \) = Additive Density
- \( \rho_{SILLO} \) = Silo Material density
- \( VF \) = Void fraction of vitrified gems or pellets in shipping configuration

The number density of element i is then,
\[ N_I = \frac{\text{Avogadro's Number}}{A_I} \cdot WF \cdot \rho_{mix} \]

IV. FEDERAL REGULATIONS

Shipping of radioactive materials is controlled by the Department of Transportation (DOT) and Nuclear Regulatory Commission regulations. The DOT regulations are specified in Chapter 49 of the Code of Federal Regulations, primarily in Parts 173 and 178. The NRC regulations apply to NRC licensees, and are found in Chapter 10 of the Code of Federal Regulations, Part 71. This study was based on two shipping limits: the activity of the shipping configurations must not produce a dose equivalent rate of more than 200 mrem/hr at the surface of the container, nor more than 10 mrem/hr at 2 meters from the container.

V. SHIELDING CODES APPLIED TO SHIPPING CONFIGURATIONS

Two shielding codes were evaluated and applied to the calculation of dose equivalent rates. Both codes are point kernel codes which utilize build-up factors to account for scattered gamma-rays to augment the uncollided dose equivalent rates. Both codes run on an IBM compatible PC. The codes are Microshield Version 4.1, and QADCGGP. Microshield is a microcomputer version of the mainframe code ISOSHIELD, and QADCGGP is a recent version of the DOE QAD codes which uses combinatorial geometry and geometric progression evaluation of build up factors (hence, CGGP).

A. COMPARISON OF RESULTS BETWEEN MICROSHIELD AND QAD.

Detailed calculations were done on vitrified waste cylinders with Microshield version 4.1 and QAD to compare the two codes. The two codes agreed to within a few percent of each other. This agreement is considered very good since the source terms for the two codes were calculated independently. The QAD source was determined from an Origen calculation, while Microshield calculates a source from isotopic activity input.

In order to obtain this agreement, the new and improved build-up factors based on geometric progression had to be utilized in QAD rather than the old QAD buildup factors.

VI. COMPUTER CODE FOR EVALUATING ADDITIVE FOR LIMITING DOSE RATES

It is anticipated that the silo extractions will vary in activity, so the dose equivalent rates may exceed transport limits unless the vitrification process is designed to counter increases in activity. A computer code was designed and written to provide the proper additive weight fraction to limit dose equivalent rates to allowable transport limits. The code uses the QAD

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shielding code in conjunction with some auxiliary coding and subroutines to limit the surface dose equivalent rate for a given transport configuration to 200 mrem/hr, and the dose equivalent rate 2 meters from the transport configuration to 10 mrem/hr. QAD was chosen over Microshield for several reasons: QAD comes with the source code written in Fortran 77, so it is amenable to alterations and additions, while Microshield does not provide a source code; the file structure of Microshield is difficult to modify, especially without access to the source code.

The code which was developed is named TCDOSE, for transport configuration dose, and works as follows: The Main Program reads a file which provides measured activity and composition data. The code then calls QAD which reads a QAD input file, to obtain information which will not change during the course of the calculation. This information will include the geometry of the transport configuration, shielding materials and compositions, gamma-ray energies, dose conversion factors, and detector locations. Initial values for the total source activity and vitification compositions are also read, and a preliminary calculation of dose equivalent rates is made. QAD must be run to put the data in the proper common blocks or storage areas. Two input areas must change in the initial QAD file to obtain the first iterate of the dose equivalent, based on the measured data. These include the total source activity and the composition of the source or vitrified material. The Main Program will call a subroutine to reevaluate the silo material composition in accordance with the measured input data. This data is stored in a common block in the QAD code. In addition, the total source will be changed based on the measured data. The total source strength in photons/sec is also stored in common. Prior to making the first iterate with the QAD code, some parameters in a common block have to be restored to their initial values, since these values are changed when QAD is run. A subroutine which restores this data to its initial values is called before each subsequent QAD run. QAD is then run with the source strength and composition provided by the measured data file for the surface detector. If the dose equivalent rate is less than 200 mrem/hr, then the dose equivalent rate and additive weight fraction are printed. If the surface dose equivalent rate is greater than 200 mrem/hr, the additive weight fraction is increased, and the necessary changes made to the composition array and total source, and QAD is rerun. Iterations are made until the surface dose falls between 190 and 200 mrem/hr, after which the dose rate and additive weight fraction are printed. The vitrified composition is also printed to the output file. The procedure is then repeated for the dose equivalent rate at 2 meters, starting with the weight fraction developed for the surface detector. When the dose equivalent rate at 2 meters is between 9.9 and 10 mrem/hr, the dose equivalent rate calculation is complete. All output information will be shown on the screen and in an output file. Figure VI.1 depicts the essentials of the calculational procedure.

Some results are shown in Figure VI.2. These results are for a truckload of gems shielded with 6 inches of concrete. The results indicated that the limiting parameter for transport is the dose equivalent rate at 2 meters. The surface dose equivalent rate of 200 mrem/hr was met with the standard additive weight fraction of 0.15.
Figure VI.1 Essentials of Computer Code for Evaluating Additive Weight Fraction

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Figure VI.2 Weight Fractions for 2 Meter Dose Limit

References


2) "Microshield version 4.10", Grove Engineering, Inc., 15215 Shady Grove Road, Rockville, MD 20850, 1993


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