Conversion Factors for Predicting Unshielded Dose Rates in Shielded Waste – 9162

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ABSTRACT

This document describes the methodology developed and used by the Advanced Mixed Waste Treatment Project for determining the activity content and the unshielded surface dose rate for lead lined containers contaminated with transuranic waste. Several methods were investigated:

- Direct measurement of the dose rate after removing the shielding.
- Use of a MicroShield® derived dose conversion factor, $(\text{mRem/hr}_{\text{unshielded}})/(\text{mRem/hr}_{\text{shielded}})$, applied to the measured surface dose rate to estimate the unshielded surface dose rate.
- Use of a MicroShield® derived activity conversion factor, $\text{mRem/hr}_{\text{unshielded}}/\text{Ci}$, applied to the measured activity to estimate the unshielded dose rate.
- Use of an empirically derived activity conversion factor, $\text{mRem/hr}_{\text{unshielded}}/\text{Ci}$, applied to the measured activity to estimate the unshielded dose rate.

The last approach proved to be the most efficacious by using a combination of nondestructive assay and empirically defined dose rate conversion factors. Empirically derived conversion factors were found to be highly dependent upon the matrix of the waste. Use of conversion factors relied on activity values corrected to address the presence of a lead liner.

INTRODUCTION

The Advanced Mixed Waste Treatment Project (AMWTP), operated by Bechtel BWXT Idaho, LLC. (BBWI), is responsible for characterization, treatment, and shipment of transuranic (TRU) waste to the Waste Isolation Pilot Plant (WIPP). The vast majority of the AMWTP TRU waste is classed as Contact Handled (CH) having surface dose rates that are below 200 mRem/hr. There is a population of waste, however, packaged in lead lined 55-gallon drums that may in fact be Remote Handled (RH). The uncertainty in the classification arises due to the presence of the lead shielding. Shielding may not be used to change the classification of waste from RH >200mRem/hr to CH. The WIPP characterization criteria for CH and RH are different. Therefore, to ensure proper characterization, it is necessary to determine the surface dose rate that would exist if the lead liner were removed. Once the unshielded dose rate is determined it may then be compared with the 200 mRem/hr limit to determine which classification is appropriate.

The Department of Energy Idaho Field Office (DOE-ID) requested BBWI to develop and formally document a methodology that will either remove the subject containers from the suspect RH-TRU population, or identify them as candidate RH-TRU containers. This report presents the technical approach and methodology for identifying candidate RH-TRU containers.

Characterization Data

There is a wide range of AMWTP characterization data available when attempting to determine a container's unshielded surface dose rate. This data includes: Non-Destructive Assay (NDA), Real-Time Radiography (RTR), and measured surface dose rates. Each is discussed below.

NDA Data

The NDA data that is available includes gamma-ray spectral data, passive neutron data, and in some cases active neutron interrogation data. A review of this data indicates that, in the majority of cases, the lead lined drums contain waste that has an elevated Am-241 content. An initial review of the NDA data also indicates that the results are significantly biased due to the presence of the lead liner. A detailed review, and reanalysis, of the NDA data associated with each drum was carried out to correct the reported results for the affect of the lead liner. The aim was to make an accurate determination of the activity content for each drum.

It was found that the best method for determining the activity content is based on an analysis of the gamma-ray spectral data. To account for the lead liner thickness, an attenuation correction is applied to the detection efficiency of each photopeak. The photopeak efficiency is determined during normal NDA analysis based on the density of the waste within the waste drum. However, the calibration that is used does not account for the presence of a lead liner. For this reason an additional correction must be applied to the NDA to account for the attenuation caused by the lead liner. The mass attenuation coefficient values from the NIST X-Ray attenuation database[2] were applied to the peak efficiencies. This attenuation data is presented graphically in Figure 1, and is used to correct the detection efficiency values using the Equation 1. Where;

ε'_E is the detection efficiency at energy E corrected for the lead liner thickness.
ε_E is the detection efficiency at energy E not corrected for the lead liner thickness.
μ_E is the mass attenuation coefficient at energy E.
ρ is the density of the lead liner.
t is the thickness of the lead liner.

Equation 1

$$\varepsilon'_E = \varepsilon_E \cdot e^{-\mu_E \rho \cdot t}$$

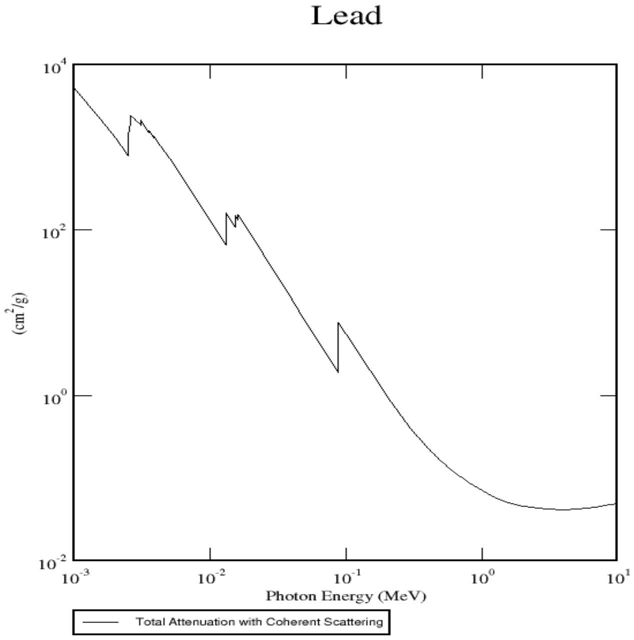

Figure 1. NIST X-Ray Mass Attenuation values for lead

This method must be treated with some caution, however, since the lead liner is not accounted for in the NDA calibration or the analysis that is automatically performed by the drum assay system. This analysis first determines the lead liner thickness using a differential attenuation model and then corrects the detection efficiency for the impact of the lead liner. This analysis takes as its inputs the following information:

1. The Am-241 photopeak areas obtained from the gamma-ray spectral analysis
2. The live time of the gamma-ray spectral acquisition
3. The Am-241 branching ratios for the selected gamma-rays
4. The detection efficiency as determined by the NDA system
5. The mass attenuation coefficients for lead at the gamma-ray energies used
6. The density of lead

An example of the data used to determine the lead liner thickness and to correct the NDA results for its presence is presented in Table 1.

The basis for the lead thickness determination is that the Am-241 activity is constant for all of its gamma-rays. If the reported activities for the various gamma-rays show a trend then it indicates that the detection efficiencies used in the NDA analysis are incorrect. As can be seen in the graph that is given in Figure 2, the Am-241 activity results obtained using the detection efficiencies reported by the NDA system, show a clear trend upwards with increasing photopeak energy. This is a feature of an analysis that has under corrected for the presence of an attenuator. In this case it was assumed that this under correction was due to the fact that the calibration of the system does not account for the presence of a lead liner. Based on this assumption a lead correction algorithm was carried out using a lead attenuator with variable thickness. The lead thickness was adjusted until the corrected Am-241 activity showed no dependence on the gamma-ray energy. The results of this correction are given in Table 1 and are graphically presented in Figure 2.

The lead liner thickness that was determined during the analysis described above was 0.44 cm, this is given as the *effective* lead liner thickness at the top of Table 1. A review of the RTR data indicates that the lead liner for this drum is in fact 1/8″ or 0.3175cm. This discrepancy may at first glance be attributed to an overestimate in the liner thickness by the NDA technique described above or as an error in the Rocky Flats historical data. However, on closer inspection it becomes clear that the path length, of a gamma-ray produced within the lead lined drum, through the lead liner would at a minimum be equal to the lead liner thickness. This minimum path length would only correspond to those gamma-rays that were produced in the same horizontal and vertical plane as the gamma-ray detector. All other gamma-rays produced within the drum would have a longer path length through the lead. This increase in the path length through the lead is geometry dependant and although it was not specifically determined during this review it does seem reasonable that the *effective* lead liner thickness that was determined here is consistent with the presence of a lead liner with a thickness of 1/8″.

Having determined the effective thickness of the lead liner it is possible to make an estimate of the actual Am-241 activity present within the drum. In the example given this turns out to be approximately 80 Ci.

Live time (sec)	529.42
Density of lead (g/cc)	11.32
Lead liner *effective* thickness (cm)	0.44

Energy keV	Branching ratio	net peak area	efficiency	Activity - As reported by NDA2000		Mass Attenuation Coefficient for Lead cm^2/g	Activity - Corrected for lead liner	
				Bq	Ci		Bq	Ci
208.01	0.00000791	9695	0.00007292	3.17E+10	0.86	0.91	2.95E+12	79.79
322.52	0.000001518	25257	0.00006599	4.76E+11	12.87	0.348	2.70E+12	72.85
332.35	0.00000149	29729	0.00006497	5.80E+11	15.68	0.328	2.97E+12	80.31
335.37	0.00000496	97499	0.00006469	5.74E+11	15.51	0.322	2.85E+12	77.13
368.65	0.00000217	52566	0.00006144	7.45E+11	20.13	0.269	2.84E+12	76.86
370.94	0.000000523	14085	0.00006125	8.31E+11	22.45	0.266	3.12E+12	84.44
376.65	0.000001383	34703	0.0000607	7.81E+11	21.10	0.259	2.84E+12	76.67
383.81	0.000000282	7836	0.00006005	8.74E+11	23.62	0.25	3.04E+12	82.06
419.33	0.000000287	8606	0.000057	9.94E+11	26.86	0.214	2.89E+12	77.97
426.47	0.000000246	7525	0.0000564	1.02E+12	27.69	0.208	2.89E+12	78.02
454.66	0.000000097	3026	0.00005421	1.09E+12	29.38	0.187	2.76E+12	74.56
619.01	0.000000594	22582	0.00004452	1.61E+12	43.59	0.12	2.93E+12	79.25
653.02	0.000000377	15326	0.00004302	1.78E+12	48.24	0.112	3.12E+12	84.27
662.4	0.00000364	147190	0.0000426	1.79E+12	48.46	0.11	3.10E+12	83.81
688.72	0.000000325	12637	0.00004151	1.77E+12	47.82	0.105	2.98E+12	80.67
722.01	0.00000196	78041	0.00004026	1.87E+12	50.49	0.0994	3.06E+12	82.83
755.9	0.000000076	2888	0.00003905	1.84E+12	49.68	0.0944	2.94E+12	79.50

Table 1. Data used in the calculation of the lead liner thickness and correction of the NDA results associated with a lead lined drum.

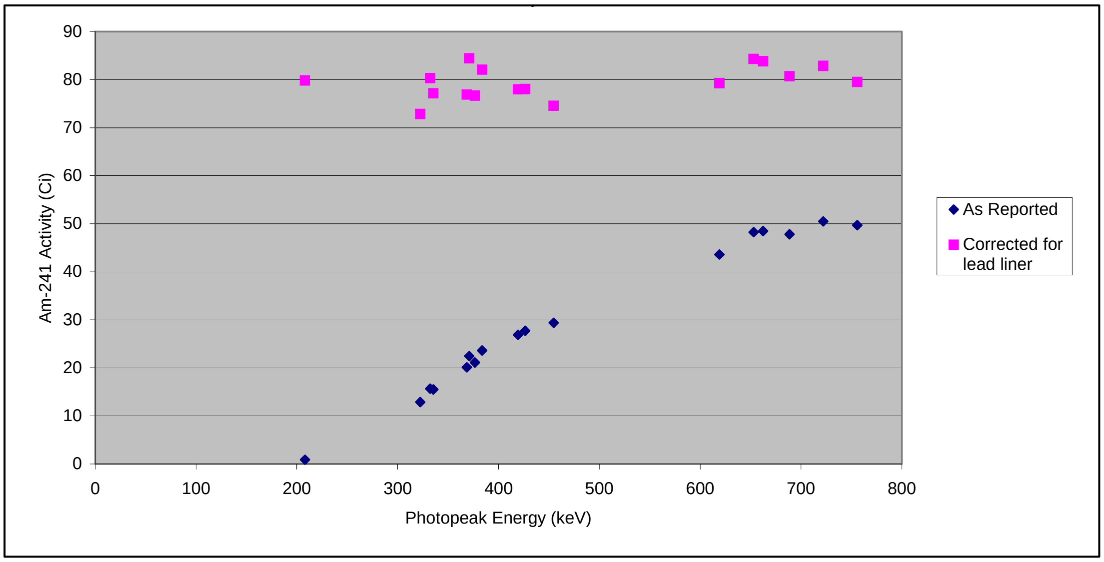

Figure 2. A graphical representation of the Am-241 activity data that is presented in Table 1

RTR Data

The results associated with the RTR data provide information regarding the presence of a lead liner and its thickness. The thickness is determined by observation based on the RTR operator's training and experience. The thickness determination is aided by the fact that there were two standard lead liner thicknesses in common usage at Rocky Flats; these were 1/8″ and 1/16″.

Surface Dose Rate Data

The surface dose rate data was obtained at the AMWTP inspection station during retrieval operations. This data is used to identify drums that required special handling due to elevated dose rate readings.

Determination of the Unshielded Surface Dose Rate

Multiple techniques were investigated to determine the unshielded dose rate. Each technique is discussed below.

Direct Measurement of Dose Rate

Removing the lead liner and directly measuring the surface dose rate is unattractive for several reasons. The work required to pull the lead liner or the drum rigid liner is time consuming, expensive, and would result in unnecessary dose to AMWTP personnel. Therefore this option was not pursued.

MicroShield® Modeling

To assist in the determination of the unshielded dose rate a commercial software program MicroShield® was used. MicroShield® is a comprehensive photon/gamma ray shielding and dose assessment program that is widely used for designing shields and estimating source strength from radiation measurements.

Models were run with various isotopic sources and it was determined that for Rocky Flats waste the surface dose rate is significantly dominated by Am-241. Since all of the lead lined containers are from Rocky Flats, the MicroShield® results are only quoted for Am-241.

MicroShield® models were run for unshielded and shielded drums with various waste matrix types and lead liner thickness. These model results were used to determine the ratio between the unshielded and the shielded surface dose rates. This ratio was then used in combination with measured surface dose rate data to determine the unshielded surface dose rate associated with lead lined drums. The results from the unshielded models were also used to determine dose conversion factors; these represent the unshielded dose rate that would be observed per Curie of Am-241 contained within the drum. These factors were then used in combination with the Am-241 activity, from NDA measurements; to determine the unshielded surface dose rate associated with lead lined drums.

MicroShield® models were run using the following input parameters:

1. A 1 Curie (Ci) source of Am-241 was defined using nuclear data obtained from the Table of Isotopes[1].
2. The drum was defined as a 55-gallon drum with a height of 88.27 cm, a radius of 29.77 cm; the drum wall was defined as iron with a density of 7 g/cc and a thickness of 0.17 cm.
3. The content of the drum was assumed to be either debris or sludge;
   - Debris was assumed to be a mixture of carbon (0.25g/cc) and iron (0.25g/cc) with a bulk density of 0.5g/cc
   - Sludge was assumed to be concrete with a bulk density of 1.1g/cc.
4. For each pair of models, the first model included a lead liner with a thickness of either 0.15875cm (1/16”), or 0.3175cm (1/8″) and a density of 11.32g/cc. In the second model the lead liner was replaced with air of density 0.00122g/cc to simulate the removal of the lead liner.

The results of these MicroShield® models are presented in Table 2.

Table 2. MicroShield® Dose Conversion Factors

Model Number	Description	Unshielded (mRem/hr/Ci)	Shielded (mRem/hr/Ci)	Ratio Unshielded to Shielded
1	Debris Drum, 1/16" Lead, 80% fill	8.593	0.03884	221.2
2	Debris Drum, 1/8" Lead, 80% fill	8.593	0.02688	319.7
3	Sludge Drum, 1/16" Lead, 80% fill	3.238	0.01942	166.7
4	Sludge Drum, 1/8" Lead, 80% fill	3.238	0.01398	231.6
5	Debris Drum, no lead, 80% fill	8.593		
6	Sludge Drum, no lead, 80% fill	3.238		
7	Debris Drum, 1/16" Lead, 40% fill	17.15	0.06499	263.9
8	Debris Drum, 1/8" Lead, 40% fill	17.15	0.04486	382.3
9	Sludge Drum, 1/16" Lead, 40% fill	6.467	0.03556	181.9
10	Sludge Drum, 1/8" Lead, 40% fill	6.467	0.02576	251.0

From this data it is clear that the waste matrix type and the fill height of the drum has a significant impact on the ratio between the shielded and unshielded dose rates. It is also clear that the use of these ratios would lead to a high level of uncertainty in the calculated unshielded dose rate for lead lined drums. The reason for this is that the detection threshold of the dose rate measurement is 0.1mRem/hr and a surface dose rate measurement of 1mRem/hr would be sufficient, in most cases, to yield an unshielded surface dose rate of >200mRem/hr. Since a surface dose rate measurement of 1mRem/hr is so low and is also very close to the detection threshold, it makes the measurements very susceptible to variations in the environmental background and measurement uncertainty. These problems are minimized if instead of using measured surface dose rate data as a basis for determining the unshielded surface dose rate, NDA data is used. The advantage of using NDA data is that it requires only one MicroShield® model for each waste matrix type. NDA measurements performed at the AMWTP also have better sensitivity and lower measurement uncertainty than dose rate measurements made at the AMWTP.

MicroShield® Modeling (mRem/hr$_{unshielded}$)/(mRem/hr$_{shielded}$)- Direct Measurement of Shielded Dose Rate

Use of the measured shielded surface dose rate and a simple (mRem/hr$_{\text{unshielded}}$)/ (mRem/hr$_{\text{shielded}}$) conversion factor has the distinct advantage of being very straight forward and simple to apply. However, due to the sensitivity of the measured surface dose rate to the waste matrix composition and the lead liner thickness, this approach was deemed impractical. In addition, to determine the modeled conversion factor two sets of MicroShield® models were required; one for unshielded drums and one for shielded drums containing various waste types and lead liner thickness. These model results were used to determine the ratio between unshielded and shielded surface dose rates. This ratio was then used in combination with measured surface dose rates to determine the unshielded surface dose rates associated with lead lined drums.

MicroShield® Modeling (mRem/hr$_{unshielded}$/Ci)

The use of measured activity data increases the complexity by requiring that a lead attenuation correction is applied to the NDA. The NDA data is then used, in combination with a single dose rate model, to determine the unshielded surface dose rate. However, using the activity data does have the distinct advantage that it removes the uncertainty associated with using the measured surface dose rate data and it also reduces the reliance on modeling results obtained from MicroShield®. If NDA data is used to determine the unshielded surface dose rate associated with lead lined containers, then a single MicroShield® model would be required to determine the dose rate conversion factor.

Empirical Derived Conversion Factors (mRem/hr$_{unshielded}$/Ci)

An alternative method for determining dose rate conversion factors was developed. This method uses measured surface dose rate data and NDA results associated with drums that are not lead lined. Based on a statistical analysis of the available data empirical dose rate conversion factors were determined.

An initial review of the available data indicates that dose rate conversion factors are strongly dependant on the waste matrix. This is consistent with the findings of the MicroShield® modeling exercise that is summarized in the previous section. To account for this dependence the population of drums were sorted in accordance with their Item Description Codes (IDC). For each IDC that considered an analysis was carried out to determine the dose rate conversion factor and its associate uncertainty. This analysis consisted of calculating the ratio between the measured surface dose rate and the measured Am-241 activity for each available drum. These ratios represent the observed dose conversion factors. The data that was used to calculate the dose conversion factors was conditioned to ensure that it produced reliable results. For example, the surface dose rate measurements have a lower threshold of 0.1mRem/hr; for this reason drums that have a reported surface dose rate close to this threshold would yield unreliable ratios. These drums were, therefore, removed from the analysis.

Once the measurement data had been suitably conditioned it was used to create a histogram of the observed dose conversion factors for each IDC. An example of such a histogram is presented in Figure 3 this data is associated with a group of 76 drums of IDC-376 waste that are not lead lined. The data points represent the frequency of each observed dose rate conversion factor over the range 0 – 18 mRem/hr per Curie of Am-241. The line represents the Gaussian function that was fitted to the observed data; the Gaussian function that was used for this fit is given in Equation **2**. The fit parameters that were determined for IDC-376 are presented in Figure 3. A similar analysis was carried out for other IDCs. Unfortunately, it was not possible to complete this analysis for all of the IDCs since measurement data was not available for a sufficient number of drums in all cases. Where the analysis was not possible, an attempt was made to match the IDC in question with an IDC for which the analysis was completed. This pairing of IDCs was based on the waste description and also the calculated bulk density of the waste. The results of the analysis are presented in Table 3.

Equation 2 $$y(x) = y_0 \cdot e^{\left[\frac{-(x-x_0)^2}{2\sigma^2}\right]}$$

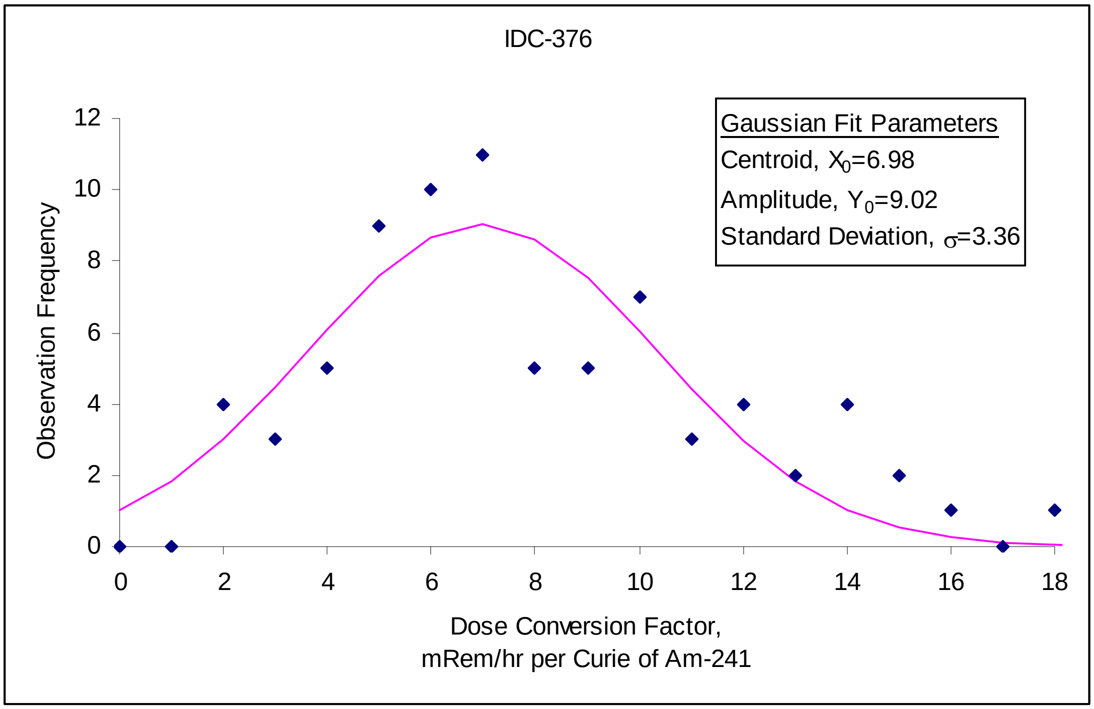

Figure 3. A graphical representation of the observed dose rate conversion factors that are available for drums containing IDC-376 waste.

Table 3. Empirical Dose Conversion Factors

IDC	Description	Dose Conversion Factor (mRem/hr/Ci)		Comments
		Value	Uncertainty	
RF-001	First Stage Sludge	4.76	1.56	
RF-003	Organic Setups, Oil Solids	4.76	1.56	Used RF-001
RF-241	Americium Process Residue	10.83	6.63	Used RF-440
RF-312	Coarse Graphite	10.83	6.63	Used RF-440
RF-320	Tantalum	2.42	2.68	
RF-330	Dry Combustibles	19.64	14.51	
RF-335	Absolute Filters	6.98	3.36	Used RF-376
RF-336	Wet Combustibles	20.41	8.43	
RF-337	Plastic	24.43	10.77	
RF-338	Insulation and Chemical Warfare Service (CWS) Filter Media	11.6	4.44	
RF-339	Leaded Rubber Gloves and Aprons	1.79	1.59	
RF-374	Blacktop, Concrete, Dirt, and Sand	10.83	6.63	Used RF-440
RF-376	Processed Insulation and Filter Media	6.98	3.36	
RF-377	Coarse Fire Brick	8.41	5.87	Used RF-480
RF-391	Crucible and Sand	10.83	6.63	Used RF-440
RF-409	Molten Salt-30% Unpulverized	2.45	1.27	Used RF-410
RF-410	Molten Salt-30% Pulverized	2.45	1.27	Mean and Standard deviation based on 7 observations
RF-411	Electrorefining Salt	12.2	5.1	Used RF-432
RF-414	Direct Oxide Reduction Salt	8.41	5.87	Used RF-480
RF-416	Zinc Manganese Alloy Metals	10.83	6.63	Used RF-440
RF-425	Fluidized Bed Ash	12.2	5.1	Used RF-432
RF-432	Resin, Leached and Cemented	12.2	5.1	
RF-440	Glass (except Raschig Rings)	10.83	6.63	
RF-441	Raschig Rings, Unleached	9.05	2.74	
RF-442	Raschig Rings, Leached	13.88	6.29	
RF-480	Scrap Metal (Non-special source)	8.41	5.87	
RF-481	Leached Metals	3.47	2.1	
RF-900	Low Specific Activity Paper, Plastics, etc.	8.41	5.87	Used RF-480

Confirmation of Empirical Derived Conversion Factors

Data is being collected on drums to validate empirical conversion factors. The results will be inserted here.

Conclusions

The analysis and the results presented in this paper indicate that it is possible to use the NDA derived activity content of a lead lined container to make reasonable estimate of the unshielded surface dose rate. The method that was used to estimate the unshielded surface dose rates involved the use of MicroShield® dose rate modeling software and an analysis of the empirical data associated with drums that do not have lead liners. Both of these methods were used to determine matrix specific dose rate conversion factors. It was determined that the sensitivity of the dose rate conversion factors to the waste matrix composition was so great that even a small inaccuracy in the definition of the waste matrix would result in a large error in the dose rate conversion factor. For this reason it was determined that the analysis of empirical data provides the best estimates of the dose rate conversion factors and the unshielded surface dose rates for the lead lined containers. However, the MicroShield® modeling results provide a useful inter comparison for the empirical data using a completely independent method.

It became clear that MicroShield® modeling is only reliable if the input parameters are accurately defined. In particular, it was found that the source definition must be scrutinized to ensure that it provides an accurate definition of the source it represents. Specifically, it was found that the standard Am-241 source definition provided by MicroShield® yielded some highly suspect results particularly for highly attenuating geometries. This problem was reduced by specifying a *user defined* Am-241 source that provided a far more reasonable representation of an Am-241 source.

When the empirically derived dose rate conversion factors for the various IDCs were applied to AMWTP lead lined drums a group of candidate RH-TRU drums were identified. In each case the estimated unshielded surface dose rate is given together with an estimate of its associated uncertainty. The uncertainty was calculated by propagating the uncertainty in the Am-241 activity reported by NDA with the standard deviation (□) in the empirically derived dose rate conversion factors.

References

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