Establishment of the Low-Level Radwaste Classification Using the Dose-To-Curie Technique at the Lan-Yu Temporary Storage Site, Taiwan

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ABSTRACT

The Fuel Cycle and Materials Administration (FCMA) in Taiwan announced a Supplementary Regulation for Classification of Low Radioactive Wastes, as well as the Regulation for Disposing of Low Radioactive Wastes and its Facility Safety Management in July 17, 1997, and September 10, 2003, respectively. The latter regulation states that in the future, before delivering low-level radioactive waste to a final land disposal site, each waste drum must specify the nuclide activity and be classified as class A, B, C or greater than C. The nuclide activity data for approximately 100,000 drums of low-level radwaste at the Lan-Yu temporary storage site accumulated in 1982-1995, therefore, must be established according to the above regulations.

The original waste database at the Lan-Yu site indicates that the data were absent for about 9% and 72% of Co-60 and Cs-137 key nuclide activities, respectively. One of the principal tasks in this project was to perform whole drum gamma radioactivity analysis and contact dose rate counting to establish the relationship of dose-to-curie (D-to-C) of specific waste stream to derive gamma radioactivity of counting drums for 2 trenches repackaged at the Lan-Yu site. Utilizing regression function of Microsoft Excel and collected gamma data, a dose-to-curie relationship for the whole-drum radwaste is estimated in this study. Based on the relationship between radioactivity of various nuclides and the surface dose rate, an empirical function of the dose rate (Dose) associated with product of nuclide activity (Curie) and energy (Energy), CE is set up. Statistical data demonstrated that 838 whole drums were counted employing D-to-C approach to classify other 3,279 drums, and only the contact dose rate was detected for roughly 75% of the drums to estimate gamma radioactivity of whole drums, which can save considerable cost, time, and manpower. The 4,508 drums were classified as A and 7 drums as C after repackaging was complete. The estimation of D-to-C relationship was near 80% in those sorted drums**.** This methodology can provide a simple, easy and cost-effective way for inferring gamma nuclide activity.

Keywords: Low-level radwaste, whole drum waste scanning, cement solidified waste, dose-to-curie, waste classification

INTRODUCTION

The Lan-Yu temporary storage site received radioactive waste, roughly 100,000 drums, from the Taiwan nuclear power plant, and medicine, agriculture, engineering and research institutes in 1973-1995. According to new regulations for waste management introduced by the Fuel Cycle and Materials Administration (FCMA), Atomic Energy Council of the Executive Yuan in Taiwan in 1997 and 2003, waste drums must be classified as A, B, C and greater than C classes according to nuclide concentrations [1,2,3]. At the Lan-Yu temporary storage site, only the contact dose rate and nuclide types were recorded on waste drums in the early years of operation. Consequently, the waste drums cannot meet current regulations [4,5,6] for final disposal. Therefore, using current technology to re-evaluate the nuclide data for waste drums for classification is necessary.

According to regulations, drum number, surface dose rate, contamination and nuclides activities must be reported for low-level radwaste. The nuclide types and activities in each drum must be recorded on the shipping manifest for “radioactive material awaiting treatment” and a computer database must be established. Several methods provide a relatively high degree of confidence in determining the radioactivity of waste drums. Methods currently include (1) using a whole drum gamma scanner, (2) direct sampling and (3) dose-to-curie (D-to-C) conversions. The most common methods for waste characterization are using a gamma scanner and scaling factor developed in 1997 at Institute of Nuclear Energy Research (INER) of Atomic Energy Council and have been applied for waste classification at the Lan-Yu site. However, the largest problem at the Lan-Yu site is a shortage of resources and limited time to retrieve large amounts of existing waste drums. The D-to-C methodology used at the Savannah River Site (SRS) [7] and in other countries [8,9,10] were taken to develop the calculation model and reduce manpower and time required to achieve the same reliability for waste classification at the Lan-Yu site.

The primary waste streams are categorized as concentrated waste (BCCLIQ), cleanup residue (BCRESI), resin (BIRESI), and sludge (BSLUDG) based on treatment process for the numbers of 8-2 and 13-2 trenches. Only 109 drums of miscellaneous wastes were stored in the 8-2 trench [11]. In this investigation, the gamma analytical report and data for the whole drum were transferred to Excel software, utilizing a screening function based on specific waste streams as the unit and in excess of 20 drums were measured for each waste stream. About 17 waste streams were also established by D-to-C relationship. Based on the relationship between radioactivity of different nuclides and surface dose rate, an empirical function of dose rate D related to nuclides activity (Curie) and energy (Energy, CE) is set up [12].

The whole drum gamma scanning system

Since gamma counting is very closely related to efficiency, the counting object geometry, homogenous distribution of radionuclides, matrix and density of materials need to be considered. Therefore, energy and efficiency calibration is extremely important. Although theoretical modeling was carried out assuming that the concentration of nuclides inside the drums is homogeneous and material density is constant, these assumptions no longer hold. To reduce the influence of matrix density and improve the homogeneous distribution of radionuclides, the scanning system is mounted on a mobile 20’×8’ ft container that can be moved and kept as the same counting geometry during measurement of 55-gallon solidified cement drums [13].

In this investigation, HPGe detector, counting software and standard source were installed, in addition to a waste drum rotator, and cement standard drum were to set up equipment operational procedures. For whole drum gamma scanning of a cement standard drum and actual waste drums, Genie-PC Gamma Waste Assay Software v 2.0 (Canberra, Model S431) was used to acquire gamma radionuclide activity and contact dose rate [13]. An electrically refrigerated cooler for the HPGe detector was assembled to consider that liquid nitrogen supply is more difficult at the Lan-Yu storage site than the other sites. Following the establishment of the whole drum counting system in a container room, calibration and sample counting was performed.

Preparation of cement solidified standard drum

The standard 55-gallon drum conditioned with the same composition as cement matrix of low-level waste weighed 311 kg. Next, 6 pieces of NIST certified Eu-152 line sources supplied by Isotope Products Laboratories (IPL), California with total activity of 2.85 mCi were inserted into the supporter inside the drum, thereby simulating a drum with homogeneous distribution of radioactivity on the rotator. Rotation speed was controlled at 10-20 turns per minute. Drum weight was measured automatically. Standard drums were calibrated for energy and efficiency based on quantitative analysis of waste drums.

Results and Discussion

Effect of waste density

The D-to-C technique is a simple and easy way for estimating container nuclide activity when used properly. To improve the practicality of the method, numerous assumptions were required, as were limitations for its use.

Figure 1 presents the result of MicroShield calculations for k values (dose conversion factor, Ci per R/hr) in a 55-gallon drum as a function of a fraction of Co-60 for various assumed waste densities. The k value was not sufficiently sensitive to the density of waste material, especially for waste with a density of 1.0 g/cm^3. Based on statistical data for most drums from various waste streams at the Lan-Yu storage site, average density was 1.29-1.36 g/cm^3; therefore, the density effect can be ignored.

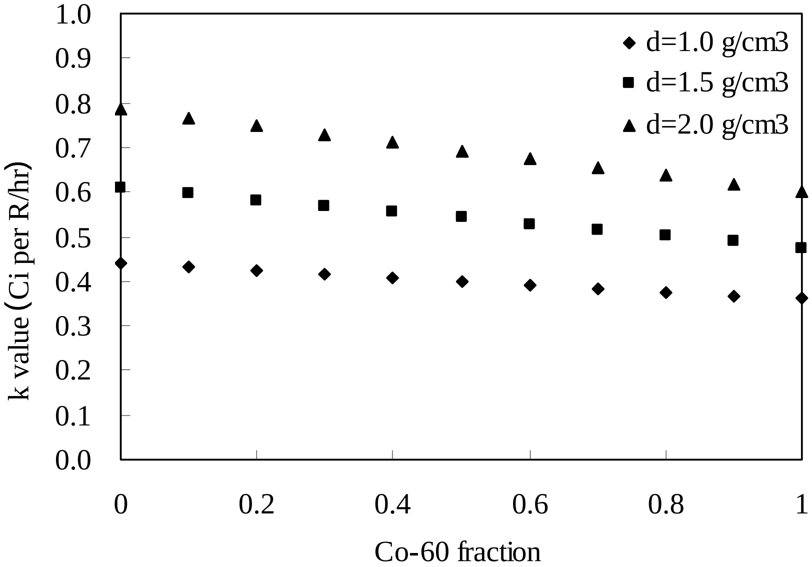

Fig. 1 Variation of k values versus fraction of Co-60 under different densities

Experimental vs. theoretical contact dose rate for all waste streams

Based on all contact dose rate values obtained with a survey meter (Model 621B Intelligent Dosimeter, Eberline Instruments, Germany) and total activity for 1883 drums, the results of experimental contact dose rate *versus* drum modeling were generated using cylindrical geometry with MicroShield 5.0 code for all drums sampled from various waste streams. The correlation coefficient of 0.8 indicated statistically high relevance, demonstrated in Fig. 2.

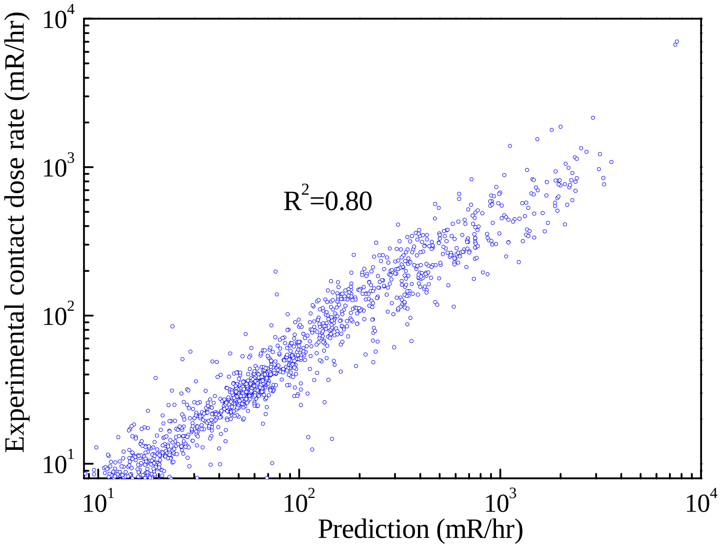

Fig. 2 The relationship between drum modeling and experimental dose rate

The principal gamma energy of radionuclides must be considered when accounting for the contribution of gamma radionuclides with respect to dose rate measurement. According to the formula for the relationship between dose rate and nuclide activity (D=kCE), linearity always exists when dose rate increases with an increase in multiplication of activity and energy. That is, k is a constant and can be described as a function of D=f (C, E). The sample of 81 drums from 1167BSLUDG for a specific waste stream is selected as the case study for determining contact dose rate and activity measurement of Co-60 and Cs-137. Table I presents the calculation steps for drum number 11674127.

Table I. The calculation steps for dose to curie

Order	Calculation steps	Process
1	Contact dose rate	D 2.07 μSv/h 2.07×10^{-4}R/h
2	Gamma nuclide energy	Co-60 energy 2.505 MeV Cs-137 energy 0.661 MeV
3	Whole drum gamma radionuclides counting and total activities	Co-60= 8.90×10^{-6}TBq/m^3 Cs-137= 4.88×10^{-7}TBq/m^3 C=$9.38\text{x}10^{-6}$ TBq/m^3÷$3.7\text{x}10^{10}\text{x}10^{12}$÷5 drums/m^3=$5.07\times10^{-5}$Ci per drum
4	Calculation of activity ratio of Co-60 and Cs-137	Co-60=94.80% Cs-137=5.20%
5	Activity (C) ×energy (E) value	5.07×10^{-5} ×[Co-60 94.8% × 2.505 Cs-137 5.20%×0.661]=1.22×10^{-4}

The relationship between D (contact dose rate) and CE (counting activity ×energy) can be drawn (Fig. 3) for 1167BSLUDG of the specific waste stream. The same approach used for D-to-C calculation can be employed to other waste streams (Table II).

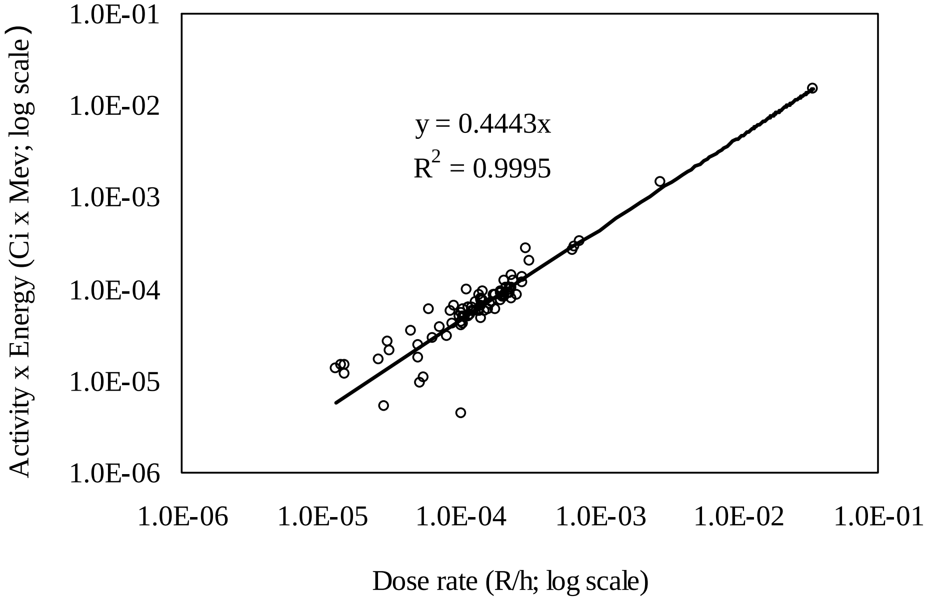

Fig. 3 The relationship between dose rate and activity for 1167BSLUDG of a specific waste stream

According to the concept of quality control, the regression coefficient of the least-square line, $R^2 > 0.7$ is defined as high relevance, 0.3-0.7 is defined medium relevance and < 0.3 is low relevance. In this project, most regression coefficients, R^2 of the D-to-C relationship, are > 0.9, indicating the high relevance and a good relationship between contact dose rate and activity (Table II) [12]. In cooperation with radwaste repackage in the trenches, the D-to-C relationship is utilized to derive radionuclide activitiy in radwaste drums. The classification database can then be established. In this manner, the counting jobs for large quantities of whole drums can be markedly reduced to save time, expense and manpower and meet the requirements of innovation, expertise and safety.

Table II The relationship between dose rate and nuclide activity by counting more than 20 drums in trenches 8-2 and 13-2 at the Lan-Yu temporary storage site

Item	Number of counting drum	Specific waste stream	Equation	R^2	Co-60%	Cs-137%
1	81	1167BSLUDG	Y=0.4443X	0.9995	88.79	11.21
2	31	1171BSLUDG	Y=0.5138X	0.8878	73.14	26.86
3	31	1172BCCLIQ	Y=0.6139X	0.7690	53.72	46.28
4	37	1269BSLUDG	Y=0.5441X	0.8495	91.80	8.20
5	35	1271BCCLIQ	Y=0.5425X	0.8582	65.14	34.86
6	61	1271BIRESI	Y=0.5459X	0.9614	84.86	15.14
7	40	1271BSLUDG	Y=0.4708X	0.9105	83.19	11.81
8	174	2071BCCLIQ	Y=0.7369X	0.9033	96.64	3.36
9	21	1167BCCLIQ	Y=0.6465X	0.9998	83.52	16.48
10	27	1170BSLUDG	Y=0.7053X	0.9064	42.46	57.54
11	24	1171BCCLIQ	Y=0.5188X	0.9239	65.57	34.43
12	22	1267BCCLIQ	Y=0.5497X	0.9818	91.65	8.35
13	20	1268BCCLIQ	Y=0.7459X	0.8761	87.84	12.16
14	26	1270BCCLIQ	Y=0.6172X	0.9898	68.97	31.03
15	20	1270BSLUDG	Y=0.5498X	0.8665	60.50	39.50
16	20	2073BCCLIQ	Y=0.7120X	0.9673	74.68	25.32
17	47	Miscellaneous wastes of Nuclear Power Plant (I)	Y=0.7120X	0.9100	72.79	27.21

Application of D-to-C methodology

Take 1271BCCLIQ waste stream for example, the activities of Co-60 and Cs-137 inside the drum using D-to-C relationship can be verified as follows, based on Table II.

Y 0.5425X, i.e, CE 0.5425R (Eq. 1)

One of non-counted drums for the surface dose rate measured on Dec., 31, 2005 was given by

X 229μSv/h 2.29×10^{-2}R/h (Eq. 2)

As observed in Table II, the activity ratios for Co-60 and Cs-137 are 65.14 % and 34.86%, respectively.

Therefore, the whole drum activity, C, can be calculated as

C×(65.14% x 2.505 MeV+34.86% x 0.661MeV) 0.5425x0.0229

C 6.67×10^{-3} Ci (Eq. 3)

Each average activity of Co-60 and Cs-137 for the whole drum can be obtained using Eq. 3 and activity ratio of each nuclide.

Co-60 6.67×10^{-3} x 65.14%

$4.34\text{x}10^{-3}$ Ci/drum×3.7×10^{10}Bq/Ci×10^{-12} TBq/Bq×5 drum/m^3

$8.04\text{x}10^{-4}$TBq/m^3 (as shown in our database, but not shown here, correct) (Eq. 4)

Similarly,

Cs-137 6.67×10^{-3}x 34.86%

$2.33\text{x}10^{-5}$Ci/drum×3.7×10^{10}Bq/Ci×10^{-12}TBq/Bq×5 drum/m^3

$4.30\text{x}10^{-4}$TBq/m^3 (as shown in our database, but not shown here, correct) (Eq. 5)

Decay correction method

Those D-to-C equations can be directly applied to surface dose rate for non-counted drums for repackaging in the same trench (Table II). Since decay correction is a function of assumed decay correction time, the radioisotopes need to be calculated with respect to different repackaging times. For instance, activity in the waste stream of 1167BSLUDG is primarily composed of Co-60 and Cs-137. Based on specific dose-to-curie conversion factors for a given waste drum configuration of 0.475 and 0.608 Ci per R/hr for Co-60 and Cs-137 and termed as k values fitted in the Fig. 1, each corrected fraction and combined conversion factors, termed k" can be plotted (Fig. 4). As can be observed, the fraction of Co-60 decreases as stored time, and Cs-137 increases as the stored time goes on. However, the combined dose conversion factor for both nuclides, k" does not change significantly.

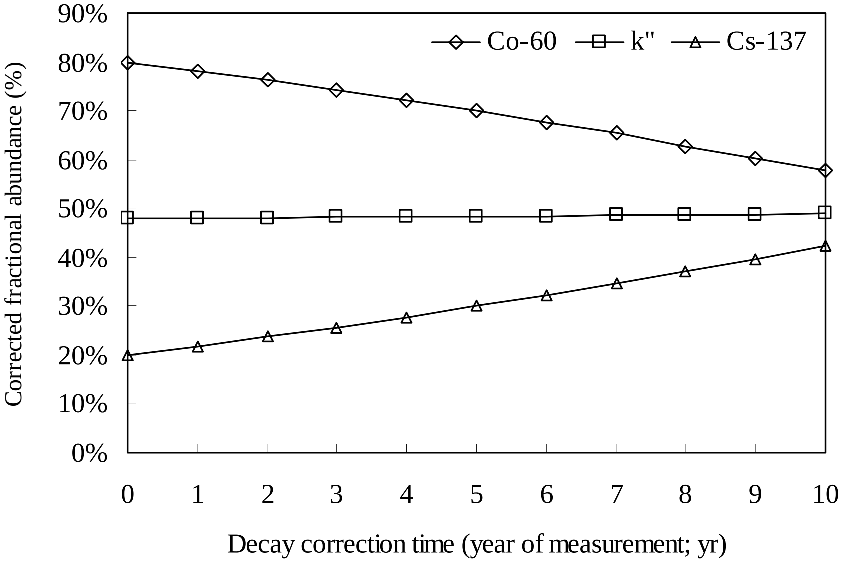

Fig. 4 Corrected fractional abundance of Co-60 and Cs-137 and variation of dose conversion factor as a function of time

Scaling factors and waste classification

Fifty-one samples of 47 waste streams showed that hard-to-measure (HTM) nuclides existed in each waste stream at the Lan-Yu temporary storage site, with Co-60 as predominant niclide. There were 13 scaling factor groups, with Co-60 and Cs-137 dominant nuclides to estimate activity of HTM nuclides such as H-3, C-14, Fe-55, Ni-63, Sr-90, Tc-99, I-129, and TRU isotopes for all drums in the trenches. More than 20 drums from 18 waste streams can be performed with calculation of scaling factors based on the D-to-C relationship.

Once D-to-C linearities are established and scaling factors are identified for each waste stream in the two trenches, it is possible to radiologically classify about 4500 waste drums generated since 1977, thereby complying with Taiwan FCMA regulations.

CONCLUSIONS

1. This study assessed the insignificant effect on drum density by simple hypothesis. In addition, we conclude that the decay correction process is essential to accurate activity evaluation when considering composition ratio of individual nuclide and should be used for reliable application of D-to-C methodology. On the other hand, the combined dose conversion factor for multi-nuclides does not change pronouncedly as the stored time increases.
2. Statistical data illustrated that 838 whole drums were counted using D-to-C approach to classify nuclide concentrations of 3,279 drums. The contact dose rate was detected for about 75% of drums to estimate gamma radioactivity of whole drums. The D-to-C technique reduces cost, time, and manpower.
3. The 4,508 drums were classified as A and 7 drums as C after repackaging was complete and the calculation model for scaling factors was established. The estimation of D-to-C relationship was

near 80% in the sorted containers. Since the functional relationship of D-to-C for the other 1,131 drums is insufficient, the database will be completed after chronologically re-packaging drums in the future.

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