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CORRELATION OF CONTAINER EXPOSURE RATES WITH RADIOISOTOPIC INVENTORY OF WASTE

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ABSTRACT

The Radioactive Waste Management Department (GRR) at the Nuclear and Energy Research Institute (IPEN) develops methods for determining the radioactive inventory of waste packages and waste streams. These methods involve radioanalysis of waste samples, gamma scanning of waste drums and calculations. The determination of the radioisotopic content of nuclear waste is a basic step in the waste characterization process and is essential in the treatment, in the transportation, and in the disposal of the waste. While radiochemical analysis of the wastes yields the most accurate results, an expedite method based on the measurement of exposure rates around waste packages can provide good approximations when previous information on gamma emitters present are available. Results of exposure rates can be used as a quick check of content in waste drum consignment and as an initial estimate of activity of fresh waste packages. The point-kernel source method is used to correlate exposure rate measurement results with gamma emission rate. Exposure rates calculated with this method are compared with results of measurements of drums containing spent ion-exchange resin replaced from the IEA-R1 research reactor water treatment system.

1. INTRODUCTION

The mission of the Radioactive Waste Management Department (GRR) at the Institute of Energy and Nuclear Research (IPEN) is to 'Promote the safe management of radioactive wastes, observing the ethical principles of protecting the human health and the environment, today and in the future." One of its duties to fulfill this mission is to develop and apply methods of characterization of the wastes in such a way as to manage them within the safety limits and with the quality required by law and regulations and recommended by international organizations.

The characterization is carried out in two steps: the first step deals with raw waste; the second step deals with waste forms. The aim of the first step is to get all properties of the waste, necessary to guide the treatment and packing processes to produce a waste form ready for final disposal. The aim of the second step is to verify and confirm that the packages comply with the requirements for storage, transportation and disposal and to supply waste data for safety assessment of the repository.

‘Characterize’ here means to determine, by way of measurements, sampling, analysis and calculations, the relevant physical, chemical and radiological properties of the wastes. One of these properties is the radioisotopic inventory contained in each waste packaging.

The radioisotopic inventory can be obtained by mathematical modeling of the physical and chemical processes carried out in the waste generating plant or by conducting measurements and applying analytical techniques followed by statistical analyses of results to determine the values and their uncertainties, or a mix of all these techniques.

Evolving and more severe regulatory requirements for the quality of data about each waste packaging require more accurate and reliable methods of characterization, motivating the improvement of existing methods.

Spent ion-exchange resin removed from the water purifying system of the IEA-R1 research reactor is one of the waste streams that are the object of such work. This is a swimming-pool reactor, which operates between 2 and 5 MWt, with light water as shielding, moderator and cooling fluid [1].

The water polishing system is composed of two absorber beds: activated charcoal and ion-exchange resin with capacity to treat 75 L.min-1, to keep soluble impurities in the water below 2 ppm. When the absorbers are no more capable of maintaining the required quality of water in the system, the beds are replaced and are both packaged in separate 200 L drums and sent to the GRR for treatment.

Packages are characterized individually because water content varies in each one and some fluctuations in the concentrations of various radioisotopes are also detected not only between two different batches of absorber, but also among different drums of the same batch and even between two distinct points in the same package. Previous work on this subject were reported before [2] some of them involving lengthy radioanalytical procedures.

In this paper, the work undertaken to contribute to the development of the characterization methodology is presented. Gamma dose rate measurements of some waste drums containing ion-exchange resins were taken and compared with the results obtained by calculation, using the point-kernel source method.

2. MATERIALS AND METHODS

Measurements of dose rates at some fixed distances from lateral surface of packages were taken using a calibrated measurements of dose rates at some fixed distances from lateral surface of packages were taken using a calibrated Model 6150 AD Automess survey meter. The experimental setup is shown in figure 1. Two packages contained dry resin and two contained resin slurry with a large quantity of supernatant water.

Previous work showed that the supernatant water contained a negligible activity concentration and the height of the absorber inside each drum was measured and taken as the height of the volumetric radioactive source both for the effect of defining the geometry of the

source for the calculations as well for defining the geometry for the dose rate measurements. These were taken in the medial plane perpendicular to the longitudinal axis of the cylinder.

Results of Cobalt-60 and Caesium-137 concentrations measured in dried samples; as well the fraction of the resin mass in the slurry samples reported earlier [3] were used to calculate the unitary activity concentration in each drum.

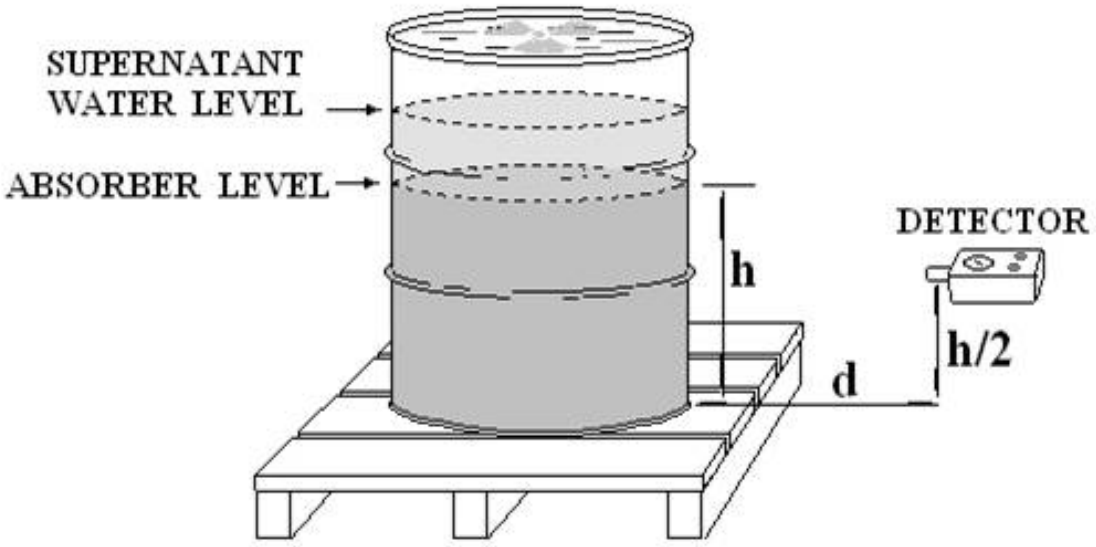

Figure 1. Experimental setup for dose rate measurements

The point-kernel source method described by Rockwell [4] consists of a macroscopic approach used for calculation of ionizing radiation exposure rate, where gamma radiation is assumed to propagate like a beam and the interactions of the photons within the traversed matter are described using macroscopic linear attenuation factors. The source is cut up in unitary cells, each one contributing to the photon flux in the measurement point. However, with this method, scattered photons that can contribute to the dose rate in the radiation detector position cannot be accounted for and so semi-empirical build-up factors are used to give consistent results.

The mathematical formulation is in Equations 1, 2 and 3, and the corresponding geometry describing this method is shown in Figure 2.

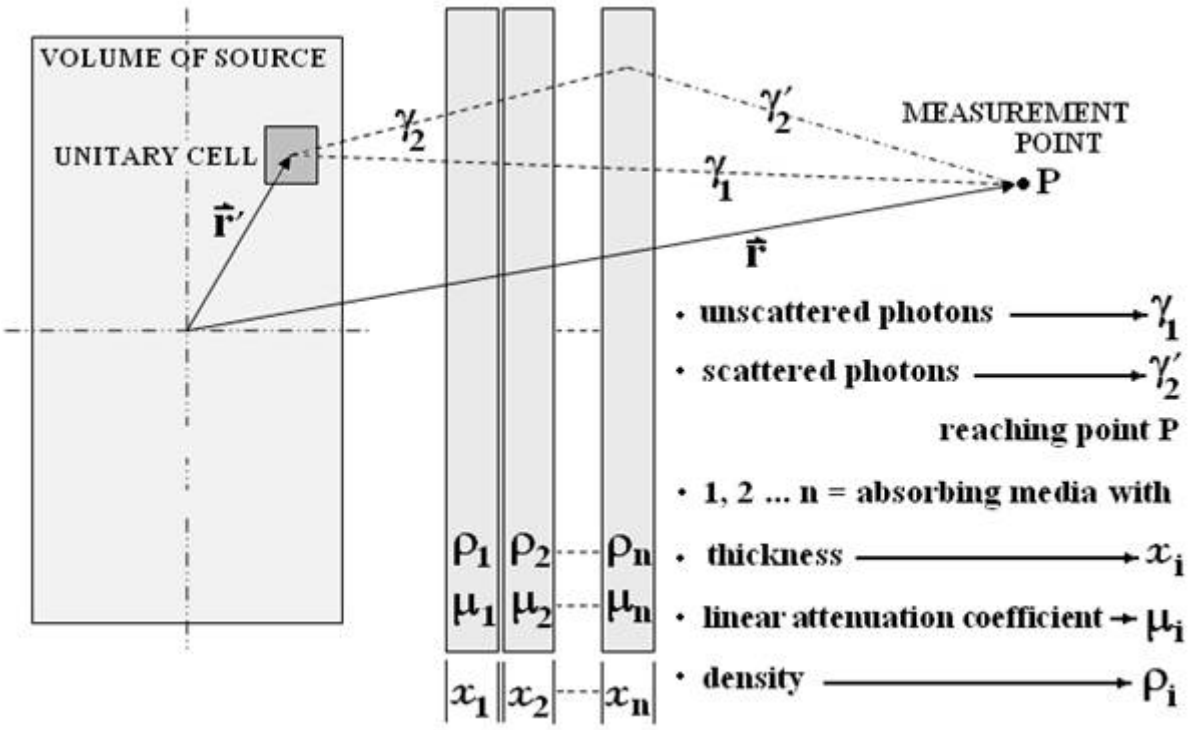

Figure 2. Geometry source-detector for application of the Point-kernel method

$$\Phi_P(\vec{r},\vec{r}',E) = S_V \cdot B(E,t) \cdot \frac{\exp[-t(E)]}{4\pi(\vec{r}-\vec{r}')^2} \quad (1)$$

with t(E) given by:

$$t(E) = \sum_{i=1}^{n} \mu_i(E) \cdot X_i \quad (2)$$

and B(E,t) given by:

$$B(E,t) = A \cdot \exp[-\alpha_1 \cdot t(E)] + (1-A) \cdot \exp[-\alpha_2 \cdot t(E)] \quad (3)$$

Where:

$\Phi_P(\vec{r},\vec{r}',E)$ is the flux, in photons of initial energy E, per second per square centimeter, at point P;

$\vec{r},\vec{r}'$ are the coordinates of the point P and of the unitary cell of the source, respectively;

S_V is the unitary cell source intensity, in photons per second per cubic centimeter,

B (E,t) is the build-up factor for scattered photons of initial energy E reaching point P
t(E) is the distance $\vec{r} - \vec{r}'$ measured as the number of mean free paths of the photon;
μ_i's are the attenuation coefficients of each shielding i for photon energy E, per centimeter;
X_i's are the thickness of each medium, in centimeter; and
A, α_1, α_2 are empirical parameters that depend on photon energy.

The total flux at point P is obtained by integrating Eq. 1 over the entire source volume and by summing up the fluxes of all photon energies emitted by the radioisotopes present in the waste.

A simplified expression of the resulting integrated flux at point P is given by Eq. 4, whose parameters are shown in Figure 3, for measurements of dose rates without any shielding between the package and the detector.

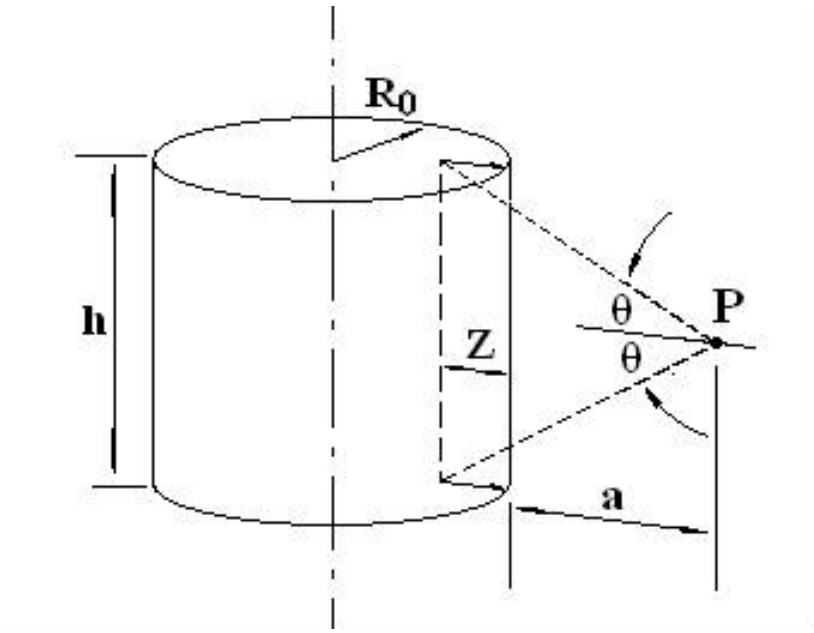

Figure 3. Geometry of source-detector setup for Eq. 4.

$$\Phi(E) = \frac{B \cdot S_V \cdot R_o^2}{2(a + Z)} \cdot F(\theta, \mu_S Z) \tag{4}$$

With **$F(\theta,\mu_S Z)$** given by

$$F(\theta, \mu_S Z) = \int_0^{\theta} e^{-\mu_S Z} \cdot \sec\theta' d\theta' \tag{5}$$

Where
R_0 is the source radius, (cm);
a is the distance of the point of measurement to the package surface, (cm);
Z is called effective self-attenuation distance, (cm);
μ_S is the linear attenuation coefficient of the slurry for each photon energy E (cm^{-1}); and
θ is the arctg [h/2 ÷ (a+Z)], (degrees) – see figure 3;

Finally, the dose rate at the measurement point P, was calculated by multiplying the flux of photons of each energy E, in photon.s^{-1}.cm^{-2} by the gamma constant for energy E, in μSv/h per unity of flux.

3. RESULTS

The values of S_V, the concentration of activity per cubic centimeter of waste were obtained from data on the concentration of activity per unity of mass of the dried resin and data of water content in resin slurries reported earlier [5]

The linear attenuation coefficients μ_S of the resin slurry for each energy E, was calculated by multiplying the density of the slurry by the attenuation coefficients, in cm^2/g, for each energy E obtained by interpolation of data from table "Partial interaction coefficients and total attenuation coefficients, for liquid water", of NIST (2011) [6]. The density of the slurry was assumed as 1 $g.cm^{-3}$.

The parameters for calculation of build-up factors were obtained by interpolation of the graph "Dose build-up factor for water, point isotropic source", of Rockwell (1956), page 419. The source radius, R_0, is half the diameter measured in the waste drums. The values of **a**, the distances between the detector and the drum surface were set at 50 cm and 100 cm. The values of Z, the self-attenuation distance, were calculated based on interpolations in graphs "Self-absorption distance, Z, of a cylinder as a function of cylinder diameter R_0, for $a/R_0 <$ 10" of pages 362 and 363 of Rockwell.

The values of F(θ,The values of θ were calculated as indicated above, using the measured values of **h**, the height of the resin absorber in each drum. The gamma constant for each photon energy was obtained by interpolation of the graph "Gamma dose rate due to 1 photon/cm^2 – sec." of page 19 of Rockwell, given in roentgens per hour, and corrected for the SI unit μSv/h, by multiplying the values by 10^4 μSv/R.

All input values and intermediate parameters used in the calculations are presented in table 1. Table 2 shows the calculated and measured dose rates.

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Table 1 – Relationship of the data used to calculate the dose rate for Co-60

Nº Drum	a (cm)	a/Ro (cm)	μs water 0,66 Mev(cm-1)	m	μs (a+Ro) mean free path	μsz	z (cm)	A	α1	α2	Buildup	Sv Bq/cm3	ϴ (Degree)	F	Φ (photons/cm2/s)
1	50	1,79	0,06	0,88	4,9E+00	1,09	17,27	8,70	-0,10	0,06	2,44	4,4E+02	2,1E+01	1,4E-01	8,8E+02
	100	3,57	0,06	1,10	8,1E+00	1,11	17,58	8,70	-0,10	0,06	2,46	4,4E+02	1,2E+01	6,5E-02	2,4E+02
	150	5,36	0,06	1,65	1,1E+01	1,47	23,24	8,70	-0,10	0,06	2,95	4,4E+02	8,4E+00	3,5E-02	1,0E+02
3	50	1,79	0,06	0,88	4,9E+00	1,09	17,27	8,70	-0,10	0,06	2,44	3,0E+02	1,7E+01	9,0E-02	3,8E+02
	100	3,57	0,06	1,10	8,1E+00	1,11	17,58	8,70	-0,10	0,06	2,46	3,0E+02	1,0E+01	6,0E-02	1,5E+02
	150	5,36	0,06	1,65	1,1E+01	1,47	23,24	8,70	-0,10	0,06	2,95	3,0E+02	6,9E+00	3,3E-02	6,5E+01
16	50	1,79	0,06	0,88	4,9E+00	1,09	17,27	8,70	-0,10	0,06	2,44	1,1E+04	2,0E+01	1,4E-01	2,2E+04
	100	3,57	0,06	1,10	8,1E+00	1,11	17,58	8,70	-0,10	0,06	2,46	1,1E+04	1,2E+01	7,0E-02	6,4E+03
	150	5,36	0,06	1,65	1,1E+01	1,47	23,24	8,70	-0,10	0,06	2,95	1,1E+04	8,2E+00	3,5E-02	2,6E+03

Table 2 – Relationship of the data used to calculate the dose rate for Cs-137

Nº Drum	a (cm)	a/Ro (cm)	μs water 0,66 Mev(cm-1)	m	μs (a+Ro) mean free path	μsz	z (cm)	A	α1	α2	Buildup	Sv Bq/cm3	ϴ (Degree)	F	Φ (photons/cm2/s)
1	50	1,79	0,086	1,13	7,0E+00	1,40	16,29	21,50	-0,13	0,00	5,12	2,5E+01	2,1E+01	1,1E-01	6,7E+00
	100	3,57	0,086	1,54	1,1E+01	1,56	18,09	21,50	-0,13	0,00	5,61	2,5E+01	1,2E+01	4,0E-02	1,2E+00
	150	5,36	0,086	1,96	1,5E+01	1,74	20,28	21,50	-0,13	0,00	6,24	2,5E+01	8,4E+00	3,2E-02	6,4E-01
3	50	1,79	0,086	1,13	7,0E+00	1,40	16,29	21,50	-0,13	0,00	5,12	1,6E+01	1,7E+01	7,0E-02	2,7E+00
	100	3,57	0,086	1,54	1,1E+01	1,56	18,09	21,50	-0,13	0,00	5,61	1,6E+01	1,0E+01	3,5E-02	6,8E-01
	150	5,36	0,086	1,96	1,5E+01	1,74	20,28	21,50	-0,13	0,00	6,24	1,6E+01	6,9E+00	2,7E-02	3,5E-01
16	50	1,79	0,086	1,13	7,0E+00	1,40	16,29	21,50	-0,13	0,00	5,12	2,1E+02	2,0E+01	1,0E-01	5,1E+01
	100	3,57	0,086	1,54	1,1E+01	1,56	18,09	21,50	-0,13	0,00	5,61	2,1E+02	1,2E+01	4,0E-02	1,0E+01
	150	5,36	0,086	1,96	1,5E+01	1,74	20,28	21,50	-0,13	0,00	6,24	2,1E+02	8,2E+00	3,2E-02	5,3E+00

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Table 3 – relationship between the dose rates for Co-60 and Cs-137 calculated and experimental, respectively

Calculated Dose Rates(μSv/h)		Total Dose Rate (μSv/h)	
Co-60	**Cs-137**	**calculated**	**experimental**
2,02E+01	9,04E-02	2,03E+01	4,25E+00
5,41E+00	1,62E-02	5,43E+00	3,20E+00
2,37E+00	8,58E-03	2,38E+00	1,45E+00
8,68E+00	3,71E-02	8,72E+00	4,30E+00
3,35E+00	9,17E-03	3,36E+00	2,65E+00
1,50E+00	4,67E-03	1,50E+00	1,00E+00
5,10E+02	6,91E-01	5,11E+02	5,45E+02
1,48E+02	1,37E-01	1,48E+02	1,85E+02
6,00E+01	7,22E-02	6,00E+01	7,00E+01

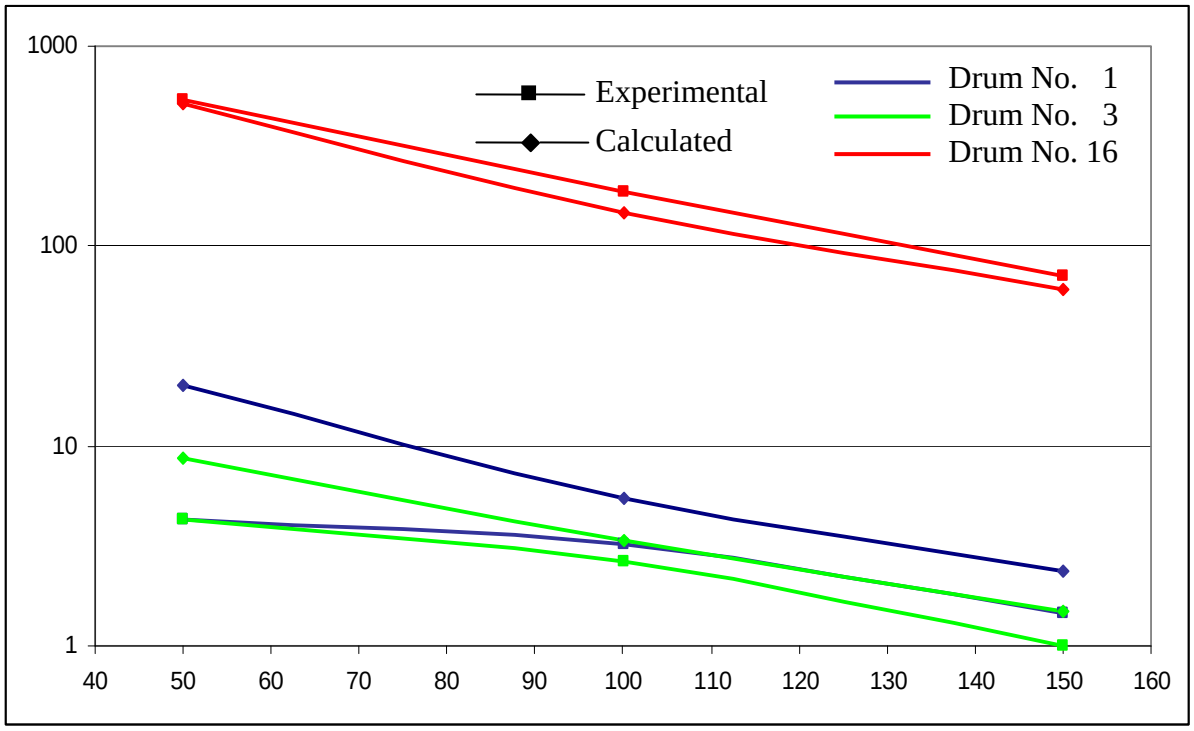

Figure 4. Correlation between the dose rates obtained

4. CONCLUSIONS

Results of measurements and calculations are in good agreement for the analyzed waste packages. Many more waste packages will be assessed next, to confirm that the method can be applied with confidence. The uncertainties in the results, both empirical and theoretical,

were not yet calculated and will require some effort because of the complexity of the method. In parallel, a commercially available computer code, that is under procurement, will be use to perform this comparison and then used as a complementary characterization tool for wastes that can be sampled and analyzed by radiochemical methods or as the only characterization tool for dry, difficult to sample, solid wastes, in connection with more sophisticated methods as gamma spectrometry and gamma scanning of waste packages.

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