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MCNPX Modelling of G-M Pancake Response to Radioiodine Charcoal Filter Used at OPG

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Abstract

The Monte Carlo N-Particle eXtended (MCNPX) radiation transport code was used to model the response of a G-M pancake detector to the charcoal-filled radioiodine cartridge air filter used at Ontario Power Generation (OPG). The MCNPX models used either a continuous beta spectrum, or the discrete photon energies from ^{131}I, exclusively. The electron and photon results were combined to determine the overall response of the G-M pancake probe. The model of the probe was benchmarked against ^{60}Co and ^{36}Cl certified button sources. The model of the charcoal filter was adjusted and benchmarked against a certified ^{131}I cartridge source. The results were in a very good agreement with the experimental data.

Keywords: MCNPX, experimental benchmark, radioiodine charcoal filter

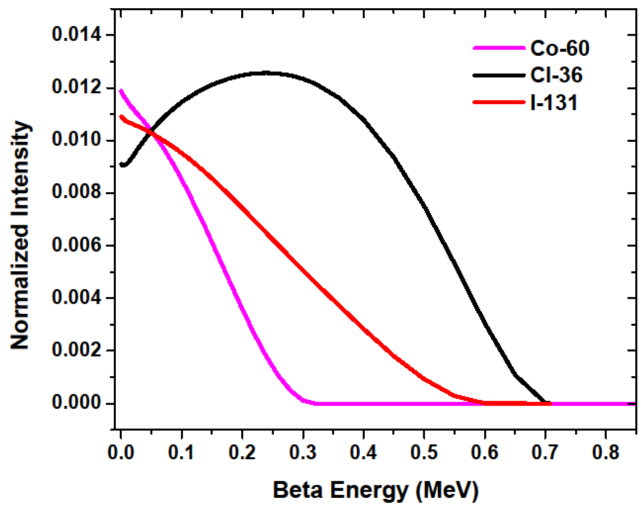
1. Introduction

Ontario Power Generation (OPG) determines the hazard from radio-iodine in air by passing 1m^3 of air through a filter paper (to filter radioactive airborne particulate) followed by a charcoal cartridge impregnated with tri ethyl diamine (TEDA). The Pickering Nuclear (PN) Fuel Handling (FH) work group experienced delays of several hours in obtaining the airborne radioiodine results, and requested that the Health Physics Department (HPD) develop a factor to convert a pancake reading in counts per minute (cpm) taken on the inlet surface of the charcoal cartridge, with the particulate filter paper removed, to radioiodine concentration in air (Bq/m^3). Since nominally 1m^3 of air is passed through the filter, with the total collection efficiency of about 88%, the overall air concentration of ^{131}I (in Bq/m^3) is expressed as calculated activity divided by 0.88. Monte Carlo N-Particle EXtended (MCNPX) **[1]** was used as a modelling tool in order to develop a realistic model of an iodine cartridge and other relevant geometry. The cartridge was modelled against G-M pancake probe. The G-M probe itself was benchmarked using ^{60}Co and ^{36}Cl certified standard sources, while the full model was benchmarked against commercially available iodine cartridge standards, uniformly filled with ^{131}I. Lastly, several other non-uniform distributions of ^{131}I in the charcoal filter were modelled as a tool to allow for evaluation of the best source distribution inside the cartridge used in the field (at CANDU stations). A study related to this work was performed by Kravchik, et al. [2]. The authors experimentally determined an absolute efficiency of HPGe detector for various geometries of detector - iodine cartridge system.

2. Methodology

MCNPX (version 2.70) was used for modelling radiation transport. Before a model of the complete geometry was put together, a model of the G-M pancake probe only, was benchmarked against the ^{60}Co beta and gamma and ^{36}Cl beta energy spectra. Both ^{60}Co and ^{36}Cl were button type certified disc sources, sandwiched between two layers of thin mylar material, sitting on a thin stainless steel backing material. Figure 1 depicts normalized beta energy spectra used in the model. The spectra were taken from ICRP 107 **[2].** In the case of ^{60}Co, gamma contribution to GM pancake probe was also modeled, by including ^{60}Co gamma lines at 1.173 and 1.332 MeV.

Figure 1: *Continuous beta energy spectra of ^{60}Co, ^{36}Cl and ^{131}I. The spectra are taken from ICRP 107 [2].*

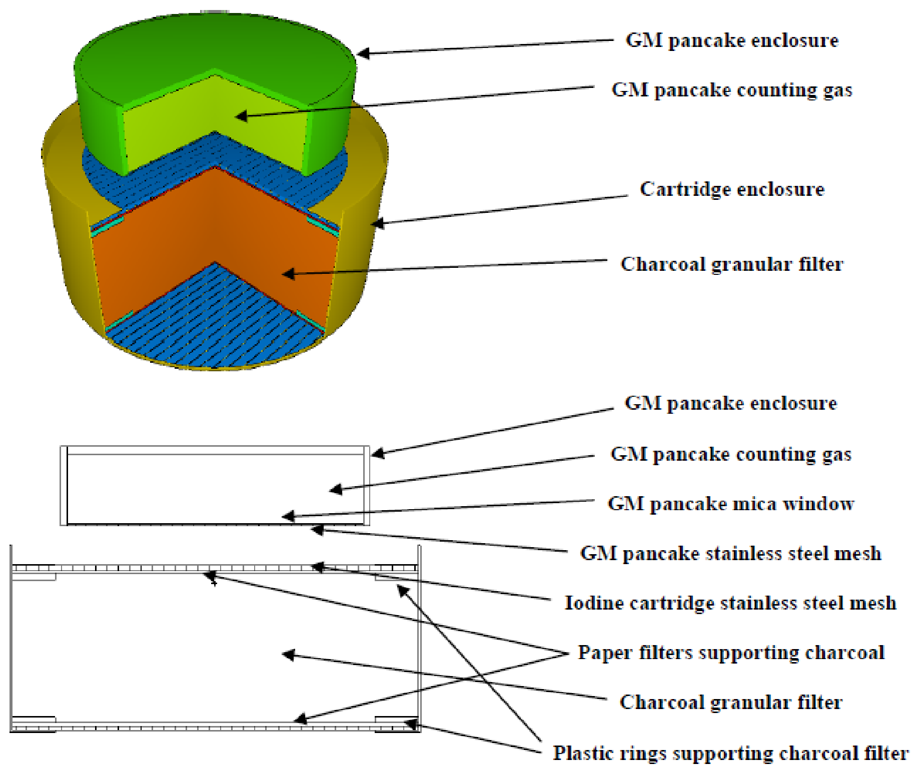


The next benchmarking step was modelling of the two certified standard reference ^{131}I sources, purchased from Eckert & Ziegler. One source was uniformly loaded with active charcoal, carrying ^{131}I, while the other source was face loaded with the same radio-nuclide, with the active charcoal having the depth of only 5 mm. The former is designated as “uniform distribution”, while the latter is designated as “uniform top distribution”, henceforth in the text. The rest of the charcoal was inactive. The nominal thickness of the charcoal filter was 22.5 mm. Both cartridges were physically counted with GM pancake probe several times. On the date of counting, both cartridges had a total of (29 ± 1) kBq of ^{131}I activity, according to source certificates **[3, 4]**. The MCNPX geometry of the GM pancake probe, placed above iodine charcoal cartridge was created using Visual Editor (Vised) tool **[5]**. This is given in Figure 2.

When counted, the probe was fixed and centred right above the cartridge. It was not possible to determine the exact distance from the mesh face of the probe to the mesh face of the cartridge. When probe is resting on the cartridge, this distance is about 6 mm, while nominal counting distance is known to be about 10 mm. Three two minute acquisitions were performed for both cartridges. The average number of counts for uniformly loaded source was 2752, while for face loaded source the average number of counts was 12099. The ten minute background was measured to be 281 counts.

Lastly, after modelling uniformly loaded cartridges and comparing the results with the actual experiments, a few other models were implemented. In particular an exponential model of the source with respect to the main axis of the source volume (cylinder) was implemented. This is done by applying an appropriate *si* and *sp* variables in MCNPX source definition (*sdef*) card [1]. Six exponential (including uniform) models were studied. Finally, ^{131}I source was placed inside the top paper filter that supports the charcoal (see Figure 2) and also a model was investigated where the disc source was placed just below the upper cartridge stainless steel mesh (see Figure 2). These models were investigated because of a suspicion that during the air sampling in the field, not all the radio-iodine accumulates in the charcoal filter, but some of it ends up in the filter paper and/or on the stainless steel mesh.

Figure 2: *3D MCNP geometry Visual Editor (Vised) output of the ^{131}I counting system; GM-pancake probe placed above ^{131}I charcoal cartridge.*



3. Results and Discussion

3.1 Modelling of Cobalt and Chlorine Source

Monte Carlo models of ^{60}Co and ^{36}Cl button sources counted using GM pancake detector yielded a very good results. The agreement with the experimental counting at 10 mm for both sources was within 3%. This ensured that the model of the GM pancake counter and approach taken to tally beta and gamma efficiencies was correct. This approach is thoroughly described in the following section.

3.2 Modelling of ^{131}I Charcoal Cartridge Loaded With Standard Reference Source

Because of the uncertainty in distance between the GM pancake probe and the iodine cartridge, during the acquisition, Monte Carlo runs were performed both at 6 and 10 mm distances. Furthermore, the density of the charcoal granules, inside the cartridge, was estimated to be 0.55 g/cm^3, using total mass of the charcoal granules and a nominal volume it occupies. However, because of irregular geometry of the granules and the fact that this was not a single, solid piece of a charcoal, but rather granules mixed with air, another set of Monte Carlo runs was performed where the density of 0.45 g/cm^3 was used. For each case, both beta and photon efficiencies of the GM pancake probe were calculated using MCNPX code. The calculations were performed separately for beta and photon cases. The discrete beta spectrum from ICRP 107 was used [**2**]. The spectra are given in Figure 2.

For photon efficiency calculations, 5 most prominent ^{131}I gamma lines were modelled: 80, 284, 364, 637 and 723 keV, having the gamma yields of 2.6, 6.1, 81.7, 7.2 and 1.8%, respectively [ICRP107]. MCNP tally *f1* was used for scoring efficiencies of beta particles. This tally determines the particles' current across the tallied surface regardless of their directions, or angles of incidence. Therefore, in order to account for particles that ENTER the active volume of detector a cosine binning of tally *f1* was used, where angles from zero to 90 degrees (with respect to the surface normal at the point of entry) were allowed only. This method assures that backscattered particles, that cross the detector window twice (when entered and when exited), are not double counted. Also, a method of surface segmentation of the tally was used in order to assure that only particles that cross the physical detector window are counted. Lastly, every particle that satisfies both described criteria was counted. The total beta efficiency (ε_β) was the actual output of the tally *f1*, since the source emission default in MCNPX is 1 particle per second per nuclear transformation, which is indeed the case with physical ^{131}I radio-nuclide. This approach could not be applied for calculation of photon (gamma) efficiency (ε_Υ), because most of the gammas that enter the active volume of the gas pass through without interacting. For that reason a tally *f8* had to be applied. This tally represents the detector response function and is constructed using a special feature of the MCNPX, called pulse height light (phl) tally treatment. The phl tally treatment allows for the conversion of the energy deposition (*f6* tallies) of secondary particles into detector pulse height (*f8* tally).

For deriving the total cartridge activity from Monte Carlo calculated efficiencies, one can start with a simple expression:

$$\boldsymbol{Activity = \frac{CR_\gamma}{\varepsilon_\gamma} = \frac{CR_\beta}{\varepsilon_\beta}} \quad \textbf{(1)}$$

where CR_Υ and CR_β are measured count rates due to gamma and beta radiations, respectively, while ε_Υ and ε_β are Monte Carlo computed efficiencies of the GM pancake counter for gamma and beta radiations, respectively. During the real measurement it is not possible to distinguish between beta and gamma count rates. It is rather a single count rate (*CR)* of the GM pancake counter that is observed. The *CR* is the sum of two count rates:

$$\boldsymbol{CR_\gamma + CR_\beta = CR} \quad \textbf{(2)}$$

From Equation 1, both CR_Υ and CR_β can be expressed:

$$\boldsymbol{CR_\gamma = \varepsilon_\gamma \times Activity} \ \textbf{(3)}$$

$$\boldsymbol{CR_\beta = \varepsilon_\beta \times Activity} \ \textbf{(4)}$$

Finally, by adding Equations 3 and 4, and substituting Equation 2 into the expression, one can obtain the activity of the charcoal cartridge due to ^{131}I:

$$\boldsymbol{Activity = \frac{CR}{\varepsilon_\gamma + \varepsilon_\beta}} \ \textbf{(5)}$$

Using Poission counting statistics, combined with Monte Carlo uncertainties, for the MCNPX cases described above and measured count rates, the ^{131}I activities were computed and presented in Table 1. The standard uncertainties at coverage factor k = 2 (95% confidence interval) are also included in the table.

	$\rho_{charcoal}$ = 0.55 g/cm^3		$\rho_{charcoal}$ = 0.45 g/cm^3	
Source	**6 mm**	**10 mm**	**6 mm**	**10 mm**
Uniform Distribution (kBq)	35 ± 2	41 ± 2	30 ± 2	35 ± 2
Uniform Top Distribution (kBq)	39 ± 1	45 ± 1	33 ± 1	38 ± 1

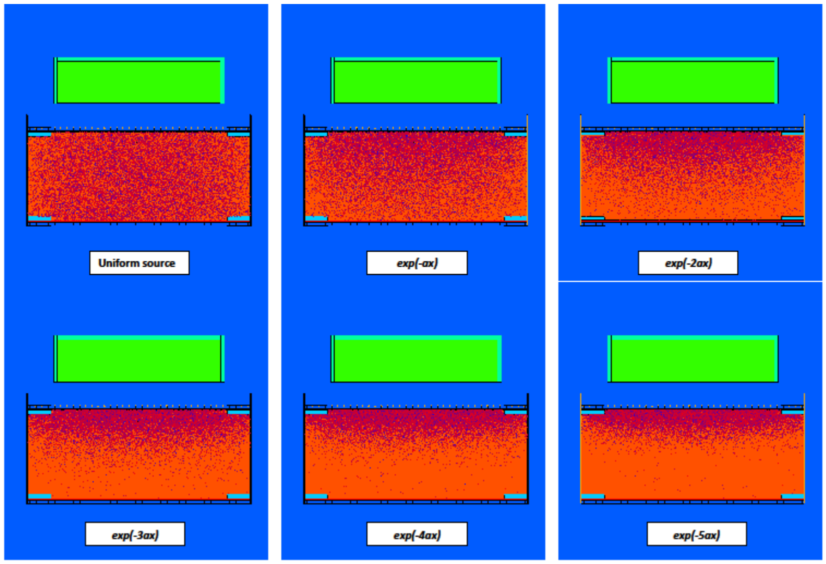
Table1: *Calculated ^{131}I activities of uniformly loaded charcoal cartridges, using measured count rates and Monte Carlo computed efficiencies of the GM pancake counter. The standard uncertainties were propagated from Poisson uncertainties, corresponding to counting statistics, and relative uncertainties computed by MCNPX code. The coverage factor of k=2 was applied. According to source certificates, true activities of both uniformly distributed charcoal cartridges were (29 ± 1) kBq for coverage factor of k=2.*

From Table 1, it is evident that Monte Carlo calculations yielded a conservative overestimates of the real activity of ^{131}I. The closest match to the certified activity of (29 ± 1) kBq was at the 6 mm geometry and charcoal density of 0.45 g/cm^3, both for uniform and uniform top source distributions. This result is an assurance that Monte Carlo model of the ^{131}I charcoal cartridge counting system is a robust and conservative with average response between 1.25 and 1.3, yielding an average over-estimate between 25 and 30%. The response value was obtained by averaging responses derived from Table 1 results (calculated activity divided by real activity of 29 kBq).

3.3 Modeling of Arbitrary Source Distributions

Figure 3 depicts the source distribution inside the cartridge for various MCNPX built-in exponential functions with respect to the main axis of a cylinder. To achieve these source distributions, an appropriate *si* and *sp* variables, describing the axial source distribution inside the cylinder, were used [1]. As mentioned earlier, a stainless steel mesh and paper filter sources were modelled also. The results for 6 and 10 mm counting distances, for an arbitrary net count rate of 2500 cpm are given in Table 2.

Figure 3: *MCNPX Vised source representation of ^{131}I source in the charcoal cartridge. Source distribution varies from uniform throughout the cartridge to different degrees of exponential distributions towards the upper part of the cartridge.*



	$\rho_{charcoal}$ = 0.55 g/cm³	
Source Distribution	**6 mm**	**10 mm**
Uniform (kBq)	32	38
Uniform Top (kBq)	8.1	9.4
exp(-*a*x) (kBq)	15.2	17.6
exp(-2*a*x) (kBq)	8.9	10.2
exp(-3*a*x) (kBq)	6.2	7.2
exp(-4*a*x) (kBq)	4.8	5.6
exp(-5*a*x) (kBq)	4.0	4.6
S. Steel Mesh (kBq)	0.27	0.33
Paper Filter (kBq)	0.52	0.61

Table 2: *Calculated ^{131}I activities for several source distributions for arbitrary net count rate of 2500 counts per minute and Monte Carlo computed efficiencies of the GM pancake counter for these source distributions. The total uncertainties are not quoted, but they are on the order of 5% for coverage factor of k=2.*

In reality ^{131}I source distribution inside the charcoal cartridge (that has been in the field), should be a linear combination of two or more source distributions presented in Table 2. By the means of gamma spectroscopy measurements, one can deduce a relative contribution of the radio-iodine source attached to a cartridge stainless steel mesh and paper filter. This could be achieved by dismantling a cartridge and separately counting all these features. Once these values are established, the portion of the source distribution in the charcoal can be obtained by linearly combining two or more distributions from Table 2. For example, if one found that:

- 20% of radio-iodine accumulates in the paper filter
- 10% accumulates on the stainless steel mesh,
- 30% corresponds to uniform source distribution
- 20% corresponds to *exp(-ax)* distribution
- 20% corresponds to *exp(-2ax)* distribution

the air concentration for net count rate of 2500 counts per minute at 10 mm counting distance would be:

$$\frac{0.2 \times 0.61kBq + 0.1 \times 0.33kBq + 0.3 \times 38kBq + 0.2 \times 17.6kBq + 0.2 \times 10.2kBq}{0.88\ m^3} = 19.4\ {}^{kBq}/_{m^3}$$

This method of calculating the airborne activity due to ^{131}I should be applied in the field, providing that a relative source distribution in the filter is known *a-priori*.

4. Conclusion

This paper thoroughly describes a Monte Carlo model of the ^{131}I counting system used in the field at Ontario Power Generation. The model consists of GM pancake counter and a charcoal filter that collects the airborne radio-iodine. The model was successfully benchmarked with ^{60}Co and ^{36}Cl certified button sources, as well as commercially available certified ^{131}I charcoal sources. As a result, a method of assessing the concentration of airborne radio-iodine, from obtained count rate, is laid out in this paper, providing that the relative source distribution inside the charcoal filter is known *a-priori*.

5. References

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