Characterization and D&D of PET Cyclotron and Radioisotope Production Facility – 17536

Paul Jones *, Jeff Slawson, CHP **, Christopher Brandjes *
* Ameriphysics, LLC
** University at Buffalo

ABSTRACT

The State University of New York at Buffalo is in the process of the decontamination and decommissioning (D&D) of a cyclotron and radioisotope production facility located in Parker Hall on the South Campus. Many unique characterization activities are needed for compliant waste packaging, transportation, and disposal.

In addition to the cyclotron used for successful production of radioisotopes, the facility features hot cells and glove boxes used for the purification and conditioning of the final product. Many challenges exist for the decommissioning of the cyclotron and the radioisotope production facility. These challenges include the proper characterization and management of the cyclotron, its associated equipment, and the remaining facility.

Slightly activated concrete constitutes the majority of the remaining radioactive materials to be disposed of during the decommissioning. Challenges for this project include;

- Regulatory interface between the New York Department of Health (NYDOH) and the New York Department of Environmental Conservation (DEC) for license closure

- Characterization of the cyclotron, its support equipment and the activated vault areas

- The most cost effective disposal options and the Data Quality Objectives needed for the various disposal options

The paper primarily focuses on challenges for characterization of the cyclotron and the remaining radioisotope production facility at the University at Buffalo.

INTRODUCTION

The decommissioning of the cyclotron and radioisotope production facility is in progress at this time. The Decommissioning Plan has been submitted to the regulator and provides the concise work instructions necessary to support the decommissioning of the University at Buffalo cyclotron and radioisotope production facility located in Parker Hall on the South Campus. Parker Hall is located on Hayes Road within the campus site. This paper provides the methodology of characterization of the major items associated with the decommissioning.

BACKGROUND

The University at Buffalo has contracted Ameriphysics, LLC to provide oversight for the decommissioning of the cyclotron and radioisotope production facility. The cyclotron and its associated components were sent for radioactive waste disposal in April 2016. The remaining impacted building surfaces remain and will require characterization, remediation, and performance of final status surveys. This will be accomplished using Ameriphysics' Nuclear Regulatory Commission (NRC) radioactive material license under reciprocity from the State of New York or a State of New York.

Activities necessary to accomplish decommissioning include decontamination where residual volumetric contamination exceeds release criterion, transportation of decommissioning wastes for disposal, and verification surveying to demonstrate compliance with the State of New York's release criterion.

The Decommissioning Plan was developed using the guidance provided in NUREG-1757, "Consolidated NMSS Decommissioning Guidance" and NUREG-1575, "Multi-Agency Radiation Survey and Site Investigation Manual" (MARSSIM) and provides the approach, methods, and techniques for the radiological decommissioning of impacted areas of the facility. These methods ensure technically defensible data are generated to aid in determining compliance with the release criteria for unrestricted use specified in the 10 NYCRR Part 16.113(g)(1), "Termination of a License and Release of a Site Without Restriction", of 25 millirem/year TEDE.

Facility description and radionuclide history

The University at Buffalo is decommissioning the cyclotron and radioisotope production facility located in Parker Hall on the South Campus. The IBA Cyclone 30 MeV cyclotron, produced traditional PET tracers and irradiated some solid targets. A separate target vault and beam line, permitted irradiation of targets at energies between 15 and 30 MeV at beam currents ranging from fractions of a microamp up to 500 microamps.

The operation of facility and possession of radioactive material is regulated by the New York State Department of Health (DOH) under the following licenses:

- License number 3033 is issued to the University for operation of the cyclotron.
- License number 2950 is issued to CRS, Inc. for operation of the on campus radiopharmacy.
- License number 1049 is issued to the University for the purpose of conducting research and providing education involving the use of radioactive materials.

A notification was made to the DOH on February 26, 2015 that operation of the cyclotron ceased on September 30, 2014. It is expected that License 3033 will be terminated upon completion of the decommissioning and final status surveys. The New York State Department of Environmental Conservation (DEC) Permit #9-1402-

00680/00029 is issued to the University for the discharge of airborne radioactive material from the facility. The University has formally requested termination of the effluent permit.

Radiological History

The site produced PET radionuclides including ^{11}C, ^{13}N, ^{15}O, ^{18}F, and ^{124}I as a result of normal operations. These radionuclides have short half-lives (110 minutes for predominant ^{18}F production), and since the site has not operated since September 2014, residual radioactivity has decayed. The reaction that produces ^{18}F, $^{18}O(p,n)^{18}F$, also produces neutrons that can cause building materials in the cyclotron area to become activated, primarily concrete and steel.

The cyclotron has been shut down since September 30, 2014. This cool down period has allowed very short-lived radionuclides to decay; however, the radionuclides in Table 1 may be present in the inner concrete vault areas. The radionuclide concentration varies according to material, proximity to the beam, and interferences that cause losses.

Table 1, Potential Radiological Contaminants

Atomic Number	Element	Radionuclide(s)
3	Hydrogen	^{3}H
11	Sodium	^{22}Na
21	Scandium	^{46}Sc
24	Chromium	^{51}Cr
25	Manganese	^{54}Mn
26	Iron	^{59}Fe, ^{55}Fe
27	Cobalt	^{56}Co, ^{57}Co, ^{58}Co, ^{60}Co
30	Zinc	^{65}Zn
41	Niobium	^{95}Nb
47	Silver	^{108m}Ag, ^{110m}Ag
48	Cadmium	^{109}Cd
51	Antimony	^{124}Sb
55	Cesium	^{134}Cs, ^{137}Cs
63	Europium	^{152}Eu, ^{154}Eu, ^{155}Eu

Three of the radionuclides listed in Table 1, (^{51}Cr, ^{56}Co, and ^{108m}Ag) are not present in the concrete samples and are typically associated with the cyclotron magnet materials such as copper and various metal alloys. Since the cyclotron and components have been removed from the site, the only potentially activated materials are the inner vault concrete and rebar.

CYCLOTRON CHARACTERIZATION

Radionuclide and Activity based on Process Knowledge

The cyclotron was characterized using a model predicting the neutron and proton induced radioactivity. The model was developed using the MCNP 4C2 and ORIGEN-S computer codes. The induced radioactivity was calculated for the Cyclone 30 based on 20 years of operation, a 20% utilization rate, production with a 40 μA current of 30 MeV protons, and a one-month decay time.

Fig. 1: The IBA 30 Cyclotron within the vault

For the purposes of characterizing the University of Buffalo Cyclone 30 machine, University at Buffalo, the owner and operator of the machine, provided the operational history consisting of the duration of time in which the machine was in use (22 years), percent utilization (18%), decay period (24 months), and production

current (33 μA). Using these parameters, the activities from Tables 5 and 6, were adjusted accordingly. The isotopes and activities associated with the cyclotron are identified in Table 1.

As a means of demonstrating that the manufacturer's activities are representative, Ameriphysics developed a Microshield® (Version 8.0.3) model for comparison. A cylindrical-volume geometry and the radionuclides and activities from Table 1 were used. The code was asked to return dose points on contact (1") and at 1-foot, 2-foot, 3-foot, and 4-foot distances. The results were compared against radiation surveys. Microshield® predicted 0.27 mR/hr on contact, whereas observed survey results were 0.3 mR/hr. Consequently there is very good agreement between the source term reflected by Table 1 and the observed measurements.

Table 1: Isotopic Distribution and

Activity for University of Buffalo Cyclotron.

Isotope	Half-life	Activity (Bq) 20 y - 1 mo decay	Corrected Activity (Bq) Based on Use Factors	Decay Corrected (Bq)	Decay Corrected (Ci)
Ag-108m	418 y	7.90E+05	4.35E+05	4.34E+05	1.17E-05
Ag-110m	249.9 d	7.20E+07	3.96E+07	1.57E+07	4.23E-04
Al-26	717,000 y	1.30E+03	7.15E+02	7.15E+02	1.93E-08
Co-58	70.86 d	1.10E+07	6.05E+06	2.29E+05	6.20E-06
Co-60	5.27 y	9.80E+07	5.39E+07	4.78E+07	1.29E-03
Cr-51	27.7 d	2.50E+07	1.38E+07	3.18E+03	8.61E-08
Fe-55	2.73 y	5.40E+08	2.97E+08	2.35E+08	6.36E-03
Fe-59	45.1 d	1.30E+07	7.15E+06	4.18E+04	1.13E-06
Mn-54	312.3 d	4.80E+07	2.64E+07	1.26E+07	3.40E-04
Ni-59	76,000 y	1.60E+04	8.80E+03	8.80E+03	2.38E-07
Ni-63	100.1 y	3.70E+06	2.04E+06	2.02E+06	5.47E-05
Ta-179	1.82 y	4.70E+05	2.59E+05	1.82E+05	4.93E-06
W181	121.2 d	1.70E+10	9.35E+09	1.38E+09	3.73E-02
Zn-65	244.3 d	3.40E+08	1.87E+08	7.24E+07	1.96E-03

The decommissioning of the cyclotron and radioisotope production facility is in progress at this time

ON-SITE CHARACTERIZATION OF CYCLOTRON ANCILLARY SUPPORT EQUIPMENT

Ameriphysics performed gamma spectroscopy via non-destructive assay (NDA) of ancillary support equipment (i.e. pumps, electrical components, etc.) that were removed from the cyclotron and packaged separately. Each package containing ancillary support equipment was analyzed using the Canberra InSpector 1000 equipped with an IPROL-1 LaBr 1.5”x1.5” probe and analyzed using the Genie 2000 and ISOCS™ (In-situ Object Counting System) software. ISOCS™ by Canberra is a software package and is factory developed and calibrated to the IPROL-1 LaBr gamma system. The factory calibration includes NIST traceable sources and modeling code for specified geometry sets. The geometry and parameter settings used for the ISOCS™ counts were noted for each package analyzed and maintained in a characterization file. Only radionuclides identified during the analysis were reported on the shipping paperwork used to transfer possession of these items.

If no radionuclides were identified but residual radioactivity is suspected based on field measurements, package activity was assigned using dose-to-Ci inference and the distributions identified in Table 1.

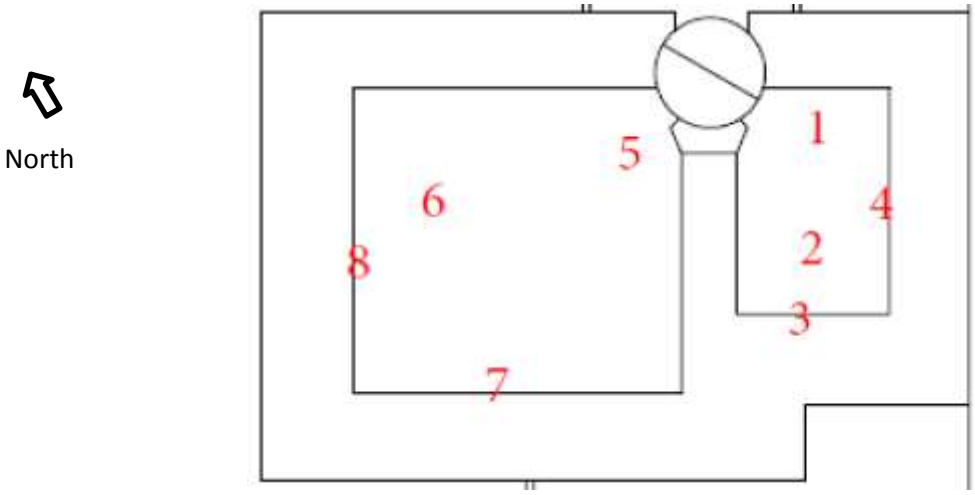
Cyclotron Vault Characterization

The extent to which the cyclotron vaults are impacted by long-lived gamma-emitting radionuclides was evaluated in May 2016 following cyclotron removal. A shielded 2-inch x 2-inch (2” x 2”) NaI gamma survey and offsite gamma spectroscopy were used to assess the site.

First, the NaI instrument background was determined by a static count in the north hallway to be 3,527 counts per minute (cpm). The acceptable background range of +/- 20% was determined. Then, the cyclotron and target vaults were scanned with the NaI detector. While all areas of the inner cyclotron and target vault demonstrated activity slightly above the background range, the target vault walls at a height of approximately 4 ft above the floor demonstrated the highest activity above background.

Following NaI scan surveys, eight sample locations were chosen. The locations of the concrete samples are provided in Figure 2.

Figure 2, Concrete Sample Areas



The above locations were selected because they represented lower count-rate activity, intermediate count-rate activity, and high count-rate activity within the expected activated area of the vault.

The samples were analyzed off site via gamma spectroscopy by General Engineering Laboratories, Charleston, SC. The laboratory results are summarized in Table 2.

Table 2, Summary of Cyclotron Vault Sample Results

Nuclide	Sample Results (pCi/g)							
	Loc. 1	Loc. 2	Loc.3	Loc.4	Loc.5	Loc.6	Loc.7	Loc.8
Antimony-124	ND	ND	ND	ND	ND	ND	ND	ND
Cadmium-109	ND	ND	ND	ND	ND	ND	ND	ND
Cesium-134	ND	0.185	0.402	0.205	0.177	0.269	0.115	0.317
Chromium-51	ND	ND	ND	ND	ND	ND	ND	ND
Cobalt-56	ND	ND	ND	ND	ND	ND	ND	ND
Cobalt-57	ND	ND	ND	ND	ND	ND	ND	ND
Cobalt-58	ND	ND	ND	ND	ND	ND	ND	ND
Cobalt-60	2.24	2.73	5.09	3.76	1.96	3.86	1.03	2.75
Europium-152	8.92	11.6	21.2	17.5	8.48	16.8	3.10	11.6
Europium-154	0.758	1.22	2.20	1.55	0.524	1.57	0.369	1.2
Europium-155	ND	ND	ND	ND	ND	ND	ND	ND
Iron-59	ND	ND	ND	ND	ND	ND	ND	ND

Nuclide	Sample Results (pCi/g)							
	Loc. 1	Loc. 2	Loc.3	Loc.4	Loc.5	Loc.6	Loc.7	Loc.8
Manganese-54	ND	ND	0.303	0.239	0.182	0.514	ND	0.286
Niobium-95	ND	ND	ND	ND	ND	ND	ND	ND
Scandium-46	ND	ND	ND	ND	ND	ND	ND	ND
Silver-108m	ND	ND	ND	ND	ND	ND	ND	ND
Silver-110m	ND	ND	ND	ND	ND	ND	ND	ND
Sodium-22	0.207	11.1	0.775	0.434	ND	ND	ND	ND
Zinc-65	ND	ND	ND	ND	ND	ND	ND	ND
TOTAL	12.1	26.8	30.0	23.7	11.3	23.0	4.6	16.2

ND = None Detected

Solid Target Cell #4 Characterization

Approximately 1 ft^2 on the horizontal surface in the back of the solid target cell (a hot cell in room 47) was found to have elevated activity up to 10,000 dpm/cm^2.

The ventilation was surveyed on the cell outlet and thoroughly at the filter boxes. Minor elevated activity was detected on the pre-filters and samples were sent off for gamma spectroscopy.

The samples were analyzed off site via gamma spectroscopy by General Engineering Laboratories, Charleston, SC. The laboratory results are attached in their entirety in Attachment 1 and are summarized in Table 3.

Table 3, Summary of Samples from Ventilation System and Sump

Nuclide	Sample Results (pCi/g)			
	Cyclotron Ventilation (NF1)	Hot Cell Ventilation (NF2)	Hot Labs Ventilation (NF3)	Sump Location (SUMP)
Antimony-124	ND	ND	ND	ND
Cadmium-109	ND	ND	ND	ND
Cesium-134	ND	26.0	ND	ND
Cesium-137	ND	61.6	20.4	ND
Chromium-51	ND	ND	ND	ND
Cobalt-56	ND	ND	ND	ND
Cobalt-57	ND	53.8	ND	ND
Cobalt-58	ND	ND	ND	ND
Cobalt-60	ND	27.4	ND	ND
Europium-	ND	ND	ND	ND

Nuclide	Sample Results (pCi/g)			
	Cyclotron Ventilation (NF1)	Hot Cell Ventilation (NF2)	Hot Labs Ventilation (NF3)	Sump Location (SUMP)
152				
Europium-154	ND	ND	ND	ND
Europium-155	ND	ND	ND	ND
Iron-59	ND	ND	ND	ND
Manganese-54	ND	32.4	ND	ND
Niobium-95	ND	ND	ND	ND
Scandium-46	ND	ND	ND	ND
Silver-108m	ND	ND	ND	ND
Silver-110m	ND	ND	ND	ND
Sodium-22	ND	389	ND	ND
Zinc-65	ND	130	ND	ND

ND = None Detected

CONCLUSIONS

The cyclotron (55-ton) and radioisotope production facility have been fully characterized. The cyclotron has been removed and sent for disposal. The Decommissioning Plan has been accepted by the NY DOH.

The two main challenges that still remain are, the removal of the activated concrete (contractor selection still in progress) and the radiological release for unrestricted use from the New York State Department of Health

REFERENCES

[1] State of New York, Department of Health 10 NYCRR Part 16.113(g), "Termination of a License and Release of a Site Without Restriction"
[2] Title 49 of the Code of Federal Regulations (CFR) Part 172, Subpart H
[3] NUREG-1575, "Multi-Agency Radiation Survey and Site Investigation Manual" (MARSSIM)
[4] NUREG-1505, "A Nonparametric Statistical Methodology for the Design and Analysis of Final Decommissioning Surveys"
[5] NUREG-1757, Volume 1 "Consolidated NMSS Decommissioning Guidance," Appendix B
[6] NUREG-1757, Volume 2 "Consolidated NMSS Decommissioning Guidance"
[7] CRP Publication 107, "Nuclear Decay Data for Dosimetric Calculations"
[8] 10 CFR 20 Subpart E, "Radiological Criteria for License Termination"
[9] Handbook of Health Physics and Radiological Health, Third Edition.