ANL/AOD/CP-103199

# Experiments With Radioactive Samples at the Advanced Photon Source

**V. R. Veluri, A. Justus, B. Glagola, A. Rauchas, & J. Vacca**
**(Argonne National Laboratory, Argonne, IL 60439)**

## Introduction:

The Advanced Photon Source (APS) at Argonne National Laboratory is a national synchrotron-radiation light source research facility. The 7 GeV electron Storage Ring is currently delivering intense high brilliance x-ray beams to a total of 34 beamlines with over 120 experiment stations to members of the international scientific community to carry out forefront basic and applied research in several scientific disciplines. Researchers come to the APS either as members of Collaborative Access Teams (CATs) or as Independent Investigators (IIs). Collaborative Access Teams comprise large number of investigators from universities, industry, and research laboratories with common research objectives. These teams are responsible for the design, construction, funding, and operation of beamlines. They are the owners of their experimental enclosures ("hutches") designed and built to meet their specific research needs. Fig. 1 gives a plan view of the location of the Collaborative Access Teams by Sector and Discipline.

In the past two years, over 2000 individual experiments were conducted at the APS facility. Of these, about 60 experiments involved the use of radioactive samples, which is less than 3% of the total. However, there is an increase in demand for experiment stations to accommodate the use of radioactive samples in different physical forms embedded in various matrices with activity levels ranging from trace amounts of naturally occurring radionuclides to MBq (mCi) quantities including transuranics. This paper discusses in some detail the steps in the safety review process for experiments involving radioactive samples and how ALARA philosophy is invoked at each step and implemented.

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**The Protocol**

A comprehensive protocol for handling all experiments at APS including the usage of experimental samples containing radioactive materials has been developed. The investigators after identifying the specific CAT and the respective beamline are directed to look into and follow the procedure spelled out in Standard Safety Envelopes for Experiment Activities at the Advanced Photon Source (APS Technical Documents, 2000). The Safety Envelope 8: Radioactive Materials gives in detail the steps an investigator is expected to go through to perform experiments with radioactive samples. These steps include among other things, the sample composition and size, specifics about sample containment planned during the experiment, any other data pertinent to the integrity and ability of the sample or the sample holder to withstand the beam conditions without any breach, special training requirements, handling, accountability, transport of the samples, etc. The APS reviews all safety aspects associated with the experiment prior to permitting the conduct of the experiment. For the purposes of this paper, we will concentrate on discussing how the radioactive sample safety issues are addressed.

**Experiment Assessment: Development Basics**

We will give a brief introduction in regard to the development of the experiment assessment criteria for radiological safety at the Argonne National Laboratory -East (ANL-E). About ten years ago, at ANL-E a draft policy on laboratory work place containment requirements and a draft implementation guide (Veluri,1991) for dispersible radionuclides were developed to answer the most common question a researcher handling radioactive materials would ask: "what is the amount of activity of a given material that I can safely work with for a 'bench top' operation in my radio-chemistry laboratory?" The policy and the guide addressed that question and over the years the guidelines developed have been reviewed by several peer groups, revised and applied at different laboratory facilities at ANL-E, and were finally fine tuned into a procedure of the Environment Safety and Health Manual (ESH Manual 5-18, 2000). A revised technical basis document (Veluri, 1998) that addresses risk-based requirements has been prepared. This technical basis document is used in conjunction with the ESH procedure 5-18 to assess the radiological safety of experiments at the APS.

Work with dispersible radioactive material involves risk. A decision must be made as to what minimal internal dose (committed effective dose equivalent) one is willing to accept for routine normal operations. Then most important question that should get a satisfactory answer is this: For a given quantity and form of a specific radionuclide what degree of containment is necessary for adequate protection for that minimal acceptable risk? To answer, let us look at some terms that are pertinent to DOE regulations.

The annual limit of intake (ALI) is that amount of a radionuclide that could be taken into the body each year without the 50-year committed dose exceeding 50 mSv (5 rem). To limit internal doses to less than 50 mSv the average radionuclide concentration in the work place would have to be such that a person would breathe in no more than one ALI during a full work-year (2000 hours) of exposure. This concentration is obtained by dividing the ALI with the volume of air breathed by a person in 2000 hours. This concentration is called the derived air concentration (DAC).

For the purpose of establishing risk-based containment requirements, it is the policy of the ANL-E that the committed effective dose equivalent from potential intakes of radionuclides resulting from normal work with dispersible radioactive materials shall not exceed 1 mSv (100 mrem). That is, the containment goal must be such that the dose commitment from a year's intake will always be less than 1 mSv. This is 2% of 50 mSv (5 rem), the DOE whole body stochastic limit for annual total effective dose equivalent.

**General Containment Equation**

The rate of change of concentration in an experiment enclosure can be written as

$$\frac{dC}{dt} + C(\lambda_r + \lambda_v) = \frac{f_a}{V}\frac{f_r}{(2000)}q$$

where C is the concentration in kBq/cc (μCi/cc) in air as a function of time, V is the volume of the room or the experiment enclosure in the present case (cc), $\lambda_r$ is the radioactive decay constant ($h^{-1}$), $\lambda_v$ is the effective air exchanges per hour, q is the activity of the material in kBq (μCi), $f_r$ is the fraction of the material that could potentially become airborne over the 2000 hours indicated (i.e., the annual average) and $f_a$ is the fraction of airborne material that could escape from a properly operating containment system.

Solving the differential equation with the initial conditions that C = 0 at t = 0, one would obtain for the concentration in air C(t)

$$C(t) = \frac{f_a}{V}\frac{f_r}{(2000)}\frac{q}{(\lambda_r + \lambda_v)}\left[1 - e^{-(\lambda_r + \lambda_v)t}\right] . \qquad (1)$$

The average concentration is given by

$$\overline{C}(t) = C_0\left[1 - \frac{1 - e^{-(\lambda_r + \lambda_v)t}}{(\lambda_r + \lambda_v)t}\right], \qquad (2)$$

where

$$C_0 = \frac{f_a}{V} \frac{f_r}{(2000)} \frac{q}{(\lambda_r + \lambda_v)} . \qquad (3)$$

For radioactive materials with the decay constant $\lambda_r << \lambda_v$, the average air concentration is controlled by the effective air exchanges per hour. The effective number of air exchanges is obtained by multiplying the number of nominal air exchanges with a correction factor called the mixing factor. This is due to the fact that perfect mixing of air, even in small enclosures, is usually unattainable. This mixing factor could vary from 0.33 to 0.1 depending on the size of the room (Constance 72).

For almost all the radionuclides used in APS experiments, $\lambda_r << \lambda_v$ is always valid. Also one can simplify the product of $f_a$ $f_r$ as F, the assumed total release fraction, for a hypothetical breach of sample containment. In general, radioactive sample experiments at APS hutches do run for a maximum of eight hours. At the APS hutches we have assumed one air exchange every two hours, and the most conservative mixing factor of 0.1. Thus, the effective number of air exchanges per hour would become 0.05.

Expressing the average concentration in equation (2) to reflect the concentration that would deliver the acceptable dose of 1mSv in a full work-year (2000 hours) the maximum activity allowable for a given release fraction can be estimated.

**Release Fractions**

The literature is replete with a number of empirical release fractions. Brodsky (Brodsky, 1989), Hickey et al (Hickey, 1991), McGuire (McGuire, 1987), and DOE Standard 1027-92 (DOE, 1997) have all suggested release fractions for different physical forms of radionuclides. Simplified categories of physical forms and the respective release fractions are utilized for experiments conducted at the APS hutches. These release fractions are very conservative reflecting the need to use sample activity as small as possible. This reduces the potential dose to as low as reasonably achievable in case of a breach of the sample containment.. The release fractions generally used at the APS are as follows: solids, 0.001, powders 0.01, and liquids 0.01. The total release fraction. F, gets modified on an individual basis for each experiment depending on the matrix embedding the sample, nature of the x-ray beam, ( unfocussed or focussed), sample holder design known for its proven integrity by experience, additional containment provided to the sample such as kapton tape snugly glued surrounding the sample holder, etc.

**Example**

An investigator wants to conduct an experiment at Bending Magnet (BM) beamline. The beam is unfocussed. The radioactive material is Pu-239 in powder form. The radioactive material is placed in a sample holder and a layer kapton tape encapsulates the material. This sample holder sits snugly in a metallic box that will contain most of the sample within the box if there is a breach. The total release fraction is estimated to be (1E-05) as follows: Powder(0.01), Kapton (0.1), metal box (0.1) and unfocussed beam (0.1). From equation (2) one can see that a 0.175 equilibrium concentration is achievable for an eight-hour run of the experiment. The approximate volume of the hutch is estimated, for this example, to be 106 Cubic Meters. The volumes of hutches can vary from 100 Cubic Meters to 290 Cubic Meters.

Assuming that the above estimated equilibrium concentration can not exceed 2% of the DAC value, the maximum amount of radioactive material that would be allowed is for the experiment is 53.7 kBq (1.45 μCi) The DAC value are tabulated by Eckerman et al ( Eckerman, 1988).

In addition to the restriction on allowable quantity of the radioactive material that could be used at the facility, the investigators have to follow several other ANL-E/APS mandated safety requirements. These requirements include the use gloves to exchange samples and at each sample exchange the area and the sample holder are smeared for possible loose contamination. The area is posted appropriately and a Hand & Shoe monitor is placed outside the hutch. The accessibility into the hutch is administratively controlled.

**Conclusion**

Evidently, one could see that ALARA principles are strictly adhered to in the design and the execution of the experiments involving radioactive materials at the APS. During the past two years, several experiments involving, samples of transuranics such as Neptunium, Americium, were run safely in addition to samples of Cs-137, natural Uranium, etc.

**Acknowledgements**

The authors wish to thank M. J. Robinet of ANL-E for his constant encouragement and the APS user community for their cooperation. This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract W-31-109-ENG-38.

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## APS Collaborative Access Teams by Sector & Discipline

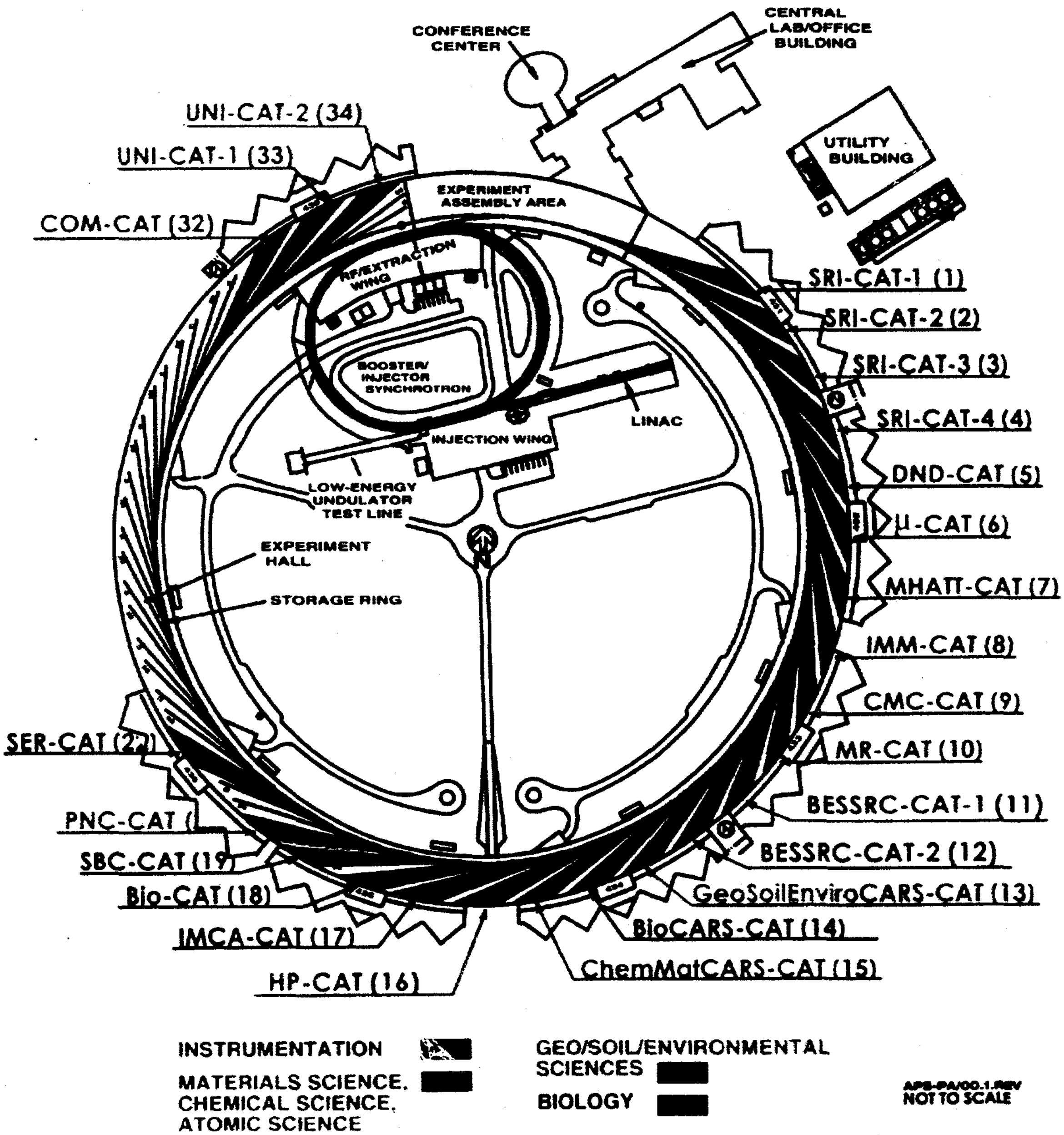


**Fig. 1**