Policy for Conducting Radioactive Sample Experiments in APS Experiment Enclosures

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This policy reflects the basic ANL philosophy that in non-controlled areas, such as the APS experimental floor, there should be no release of radioactivity. Any significant release would pose a health hazard to APS users and a risk to APS operations. The policy described below is meant to prevent radioactive releases and to limit the consequences of a potential release.

There exists a very comprehensive protocol for all experiments at the Advanced Photon Source (APS). The required procedures are spelled out in the APS Experiment Hazard Classes (APS ESAF Experiment Hazard Class 8.1). The following guidelines supplement the above technical documents. The Investigators are advised to become familiar with these documents and the relevant sections of the ANL-E Environment, Safety & Health (ESH) Manual. In this document all references to "samples" mean "radioactive samples".

All proposed experiments involving radioactive samples will be reviewed by the APS Radioactive Sample Safety Review Committee (RSSRC). The review will be on a graded basis. Hence, the experimenters are strongly advised to send in the experiment proposal in detail at least 2 months before the expected scheduled date of the experiment. Previously approved containment, isotopes and weights can be submitted as late as 2 weeks in advance.

The following guidelines are to be followed for all experiments with radioactive materials at all the experimental hutches at APS. Although spelled out as guidelines, these are minimum requirements to be adhered to by all the experimenters. All solid samples must have at least one acceptable containment enclosure. In general, no credit shall be allowed for a sample holder as containment. This will be determined by the RSSRC after a review of the proposed experiment. Gaseous radioactive samples are prohibited. Powder and liquid radioactive samples will be allowed provided they have a minimum of two containment enclosures. If multiple samples are each individually contained within separate primary barriers, they can have a common secondary and tertiary barrier. In the case of multiple samples inside a primary containment, the sample masses for each isotope must be summed to give the total mass for each isotope within the primary. Fragile materials used as containment must be approved by the RSSRC. Upon request the experimenter must provide a physical example of the containment proposed and be available to meet with the RSSRC.

The investigator proposing to do experiments with radioactive materials at an experimental hutch must provide the following in addition to other items specified in the APS ESAF Experiment Hazard Class 8.1:

- sample matrix, weight, and dimensions
- detailed description of the sample containment
- weight of **each** radioactive isotope in the sample
- data on integrity of the sample, the sample holder, and containment under expected experiment conditions (e.g., heating, cooling, pressure, etc.)
- special training requirements, in reference to handling, accountability, transport, etc., of the samples
- exposure readings from the sample at contact and at 30 cm., with a description of the instruments used to take these readings

Work with radioactive materials involves some risk. If the material is "dispersible" the risk is certainly higher. To evaluate the maximum amount of activity of a given radioactive material that can be safely used in an experiment in a hutch, one must address the maximum risk one is willing to accept in the case of a leak or breach in a containment system. Then one must address the degree of containment necessary for that acceptable maximum risk.

For the purpose of establishing risk-based containment requirements, it is the policy of ANL-E that the maximum a person should inhale from a breach or leak in sample containment should not exceed 2% of the annual limit of intake (ALI) for a given dispersible radionuclide. This would result in a committed effective dosed equivalent of 100mrem. Hence, the containment goal must be such that the dose commitment from a person's annual intake will always be less than 100 mrem. This is 2% of 5 rem, the US-DOE whole body stochastic limit for annual total effective dose equivalent (10CFR835). . In cases of potential inhalation of multiple radionuclides, the total committed effective dose equivalent from all inhaled radionuclides should not exceed 100 mrem.

A Derived Air Concentration (DAC) is defined as that concentration of radionuclide in air which, if breathed by a Reference Man for a work-year (2000 hrs), would result in the intake of one Annual Limit of Intake, ALI. That is, the concentration of a radionuclide in air is limited by

$$\int C(t)Bdt \leq ALI$$

where C(t) is the concentration of the radionuclide in air per hour at time t, B is the volume of the air breathed by worker per unit time, and the integration is carried out over a 2000 hour work-year.

In the case of a constant air concentration, the DAC is related to the ALI through

$$\mathrm{DAC}(\ \mu\mathrm{Ci/cc})\ =\ \mathrm{ALI}(\ \mu\mathrm{Ci})/\ 2.4\mathrm{E09(cc)}$$

based on a normal breathing rate B of 2.0E+04 cc per minute. There are no derived guides for instantaneous or short term values of C(t).

For a given time t that a worker in a hutch could be exposed to a dispersed radionuclide resulting from a leaking or breached containment, one can use the ANL policy to calculate the maximum allowed sample activity q_i of a given i^{th} The rate of change of concentration in a hutch can be written as

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

- C is the concentration [μCi/(cc·h)] in air as a function of time,
- V is the volume of the hutch (cc),
- λr is the radioactive decay constant (1/h),
- λv is the effective air exchanges (1/h),
- q_i is the activity of the i^{th} radionuclide ((μCi),
- f_a is the fraction of material that could potentially become air borne, and
- f_r is the fraction of material that could escape a containment system in 2000 hours.

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

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

The time-averaged concentration is given by

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

where

$$C_0 = \left(\frac{f_a}{V}\right)\left(\frac{f_v}{2000}\right)\frac{q_i}{(\lambda_r + \lambda_v)}$$

Now, if we set

$$\overline{C(t)} = C_0 \left[1 - \frac{1 - e^{-(\lambda_r + \lambda_v)t}}{(\lambda_r + \lambda_v)t} \right] = 0.02$$

for a time t that a worker in a hutch could be exposed to a dispersed radionuclide, then one can determine the maximum allowed quantity $(q_{max})_i$.

$$(q_{\max})_i = \frac{0.02(DAC)V(2000)(\lambda_r + \lambda_v)}{f_a f_v \left[1 - \frac{1 - \exp\{-(\lambda_r + \lambda_v)t\}}{(\lambda_r + \lambda_v)t} \right]}$$

where,

q is the maximum allowable activity of the solid radioactive material (μCi)
V is the volume of the experiment enclosure (cc)
λ_r is the radioactive decay constant (1/h)

λ_v is the "effective "air exchanges in the enclosure (1/h)

f_a is the fraction of the material that could escape a properly operating containment system

f_r is the fraction of the material that could potentially become airborne

DAC is the Derived Air Concentration of the radioactive material in μCi/cc.

2000 is the number of hours in a work-year.

The basis and assumptions are given in detail in section 18 of the ANL-E ES&H manual, in Veluri et al (2001), and in addition in an unpublished technical basis document in Veluri (1992).

Some simplifications to the above equation can be made without any loss of generality. For radioactive materials with $\lambda_r << \lambda_v$, the average air concentration is controlled by the effective number of air exchanges per hour. This effective number of air exchanges is obtained by multiplying the number of nominal air exchanges with a correction factor called the "mixing factor." (Constance, 1972). It should be understood that perfect mixing of air even in small enclosures is usually unattainable. The mixing factor could vary from 0.33 to 0.1 depending on the size of the enclosure. Customarily, for small enclosures 0.33 is used and for large enclosures like the hutches, 0.1 is used. At the APS hutches, it is assumed that one nominal air exchange takes place every two hours. With the most conservative mixing factor of 0.1, the effective number of air exchanges computes to 0.05 (1/h).

Similarly the product $f_a\ f_r$ can be simplified as F, the assumed total release fraction. The literature is replete with a number of empirical release fractions for different physical forms of radionuclides. (Brodsky, 1989, 10 CFR 30.72, 1991, NUREG- 1400, 1991). Conservative release fractions are used for experiments at APS, reflecting the need to use a sample activity as small as possible to reduce the potential dose to as low as reasonably achievable in case of leakage or breach.

At APS, 0.001 is typically used as the release fraction for solids, 0.01 for powders and 0.01 for liquids. If better release fractions are provided with some technical basis to back up, APS would be willing to use those release fractions.

In Table 1, the maximum allowed sample activity $(q_{max})_i$ calculated from Eqn(1) for a solid sample of selected radionuclides is provided.

In the case of multiple radioisotopes, the q_i for each radioisotope must obey

$$\sum_i q_i /(q_{\max})_i \leq 1.0$$

which ensures that the committed effective dose does not exceed 100 mrem.

Nuclide Activity	Specific Solid Sample Activity * (Ci/g)	Derived Air Concentration (μCi/cc)	Maximum Allowed Solid Sample Activity (μCi)	Maximum Allowed Solid Sample Weight (g)
Th-229	2.12E-01	4.0E-13	3	1.415E-05
Th-230	2.05E-02	3.0E-12	24	1.17E-03
Th-232	1.09E-07	5.0E-13	4	3.67E+01
U-235	2.15E-06	2.0E-11	163	7.58E+01
U-238	3.35E-07	2.0E-11	163	4.87E+02
Nat-U	6.85E-07	2.0E-11	163	2.38E+02
Dep-U	3.35E-07	2.0E-11	163	4.87E+02
Np-237	6.99E-04	2.0E-12	16	2.29E-02
Pu-238	17.0	3.0E-12	24	1.41E-06
Pu-239	6.19E-02	3.0E-12	24	3.88E-04
Pu-240	2.27E-01	3.0E-12	24	1.06E-04
Pu-242	3.91E-03	3.0E-12	24	6.14E-03
Am-241	3.42	3.0E-12	24	7.02E-06
Am-243	0.198	3.0E-12	24	1.21E-04
Cm-248	4.23E-03	7.0E-13	6	1.42E-03
Cf-248	1.58E03	3.0E-11	244	1.5E-07
Cf-252	5.35E02	8.0E-12	65	1.22E-07
Bk-249	1.63E03	7.0E-10	5691	3.49E-06
Es- 253	2.51E04	6.0E-10	4878	1.9E-07
Sr-90	13.7	8.0E-09	6.5E04	4.74E-03
Tc-99	1.69E-02	3.0E-07	2.4E06	1.42E+02
Tc-99m	5.24E06	6.0E-05	8.13E07	1.55E-05

Table 1: Maximum Allowed Solid Sample Activity of Selected Radionuclides

*The Maximum Allowed Activity is evaluated for one hour duration of stay in the hutch, i.e., t=1hr, with an assumed volume of 1.0E08 cc, with an assumed effective number of air exchanges, λ_v =0.05 (1/h), and the total release fraction F=0.001. The total release fraction could be modified on a case by case basis depending on the sample matrix, the nature of the x-ray beam, (unfocused or focused), the proven integrity of the sample holder design and any additional containment provided to the sample. For powder and liquid samples, the maximum allowed activity is reduced by a factor of 10.

References:

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