Predicting Long-Lived, Neutron-Induced Activation of Concrete in a Cyclotron Vault

L. R. Carroll
Carroll & Ramsey Associates

Abstract

Many elements in concrete can become activated by neutrons in a cyclotron vault, but only a few of the activation products are long-lived. The most prominent of these are Eu-152, Eu-154, Co-60, and Cs-134 which build up over time from (n,γ) reactions in trace amounts of stable Europium, Cobalt, and Cesium that are normally present in concrete in concentrations of a few parts per million, or less, by weight.

A retrospective analysis of data taken in connection with a previous decommissioning of a cyclotron vault, coupled with independent published data, gives us an estimate of the concentrations of these elements in concrete. With that estimate as a benchmark, we then employ a Monte Carlo Radiation Transport Code to estimate the long-term activation profile in concrete for arbitrary irradiation conditions.

Introduction

Now that Positron Emission Tomography (PET) has been approved as a diagnostic imaging modality by the U.S. Food and Drug Administration, and now that reimbursement for clinical PET procedures by government health agencies and third-party insurers is generally available, a large number of cyclotrons are being installed and commissioned world-wide for commercial-scale production of short-lived positron-emitting isotopes, particularly ^{18}F.

Many of these cyclotrons are compact, low-energy (11-12 MeV proton energy) systems which come equipped with their own built-in radiation shielding. Others, however, are higher-energy (16-18 MeV proton-energy) accelerators which are intended for installation in a concrete vault for radiation protection.

Long-term activation of the concrete in the walls of the accelerator vault may become a liability when -- after many years of operation -- the time finally comes to decommission the facility.

There are many elements in normal concrete that become activated when irradiated by neutrons from a cyclotron target. Fortunately, only a few of the resulting radio-isotopes are long-lived. These are identified in Table 1 below[1]:

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[1]A.B. Phillips, et al; "Residual Radioactivity in a Cyclotron and its Surroundings", ***Health Physics*** Vol. 51, No. 3(September), pp. 337-342, 1986

These radioactive species build up over time from neutron capture (n,γ) reactions in trace amounts of stable Europium, Cobalt, and Cesium which are present in concrete in concentrations of a few parts per million or less by weight.

Isotope	**Reaction**	**Half-life**	**Principal γ's MeV (%)**
^{152}Eu	^{151}Eu(n,γ)^{152}Eu	13.4 y	0.122 (37%), 0.344 (27%) 0.779 (14%), 0.96 (15%) 1.087 (12%), 1.11 (14%) 1.408 (22%)
^{154}Eu	^{153}Eu(n,γ)^{154}Eu	8.5 y	0.12 (38%), 0.72 (21%) 1.00 (31%), 1.278 (37%)
^{60}Co	^{59}Co(n,γ)^{60}Co	5.27 y	1.17 (100%), 1.33 (100%)
^{134}Cs	^{133}Cs(n,γ)^{134}Cs	2.065 y	0.57 (23%), 0.605 (98%) 0.796 (99%)

Table 1:
Long-lived activation Products in Concrete

To calculate the build-up of activation in a cyclotron vault, we first need a benchmark -- an estimate of the concentrations of these trace elements. We then incorporate that information in

the formula for the concrete used in a neutron transport program. Using that formula, we can then compute the activation-reaction profiles in concrete for arbitrary irradiation conditions.

Initial Benchmark

In 1981 a Model CP-42 H^- ion cyclotron, designed and built by the Cyclotron Corporation of Berkeley, CA, underwent high-current endurance testing in the factory per customer contract specification. Beam conditions during the test irradiation were: 42 MeV protons on a Cu target; 200μA for ~500 hours; target located ~18 inches from surface of the vault wall. The test resulted in long-lasting thermal-neutron-induced activation of concrete in the region of the vault wall closest to the target. This volume of concrete was assayed in 1992 and finally removed and shipped to a rad-waste site for disposal in 1996. Data from the 1992 assay, reported in a 1997 article[4] is shown in Fig.1.

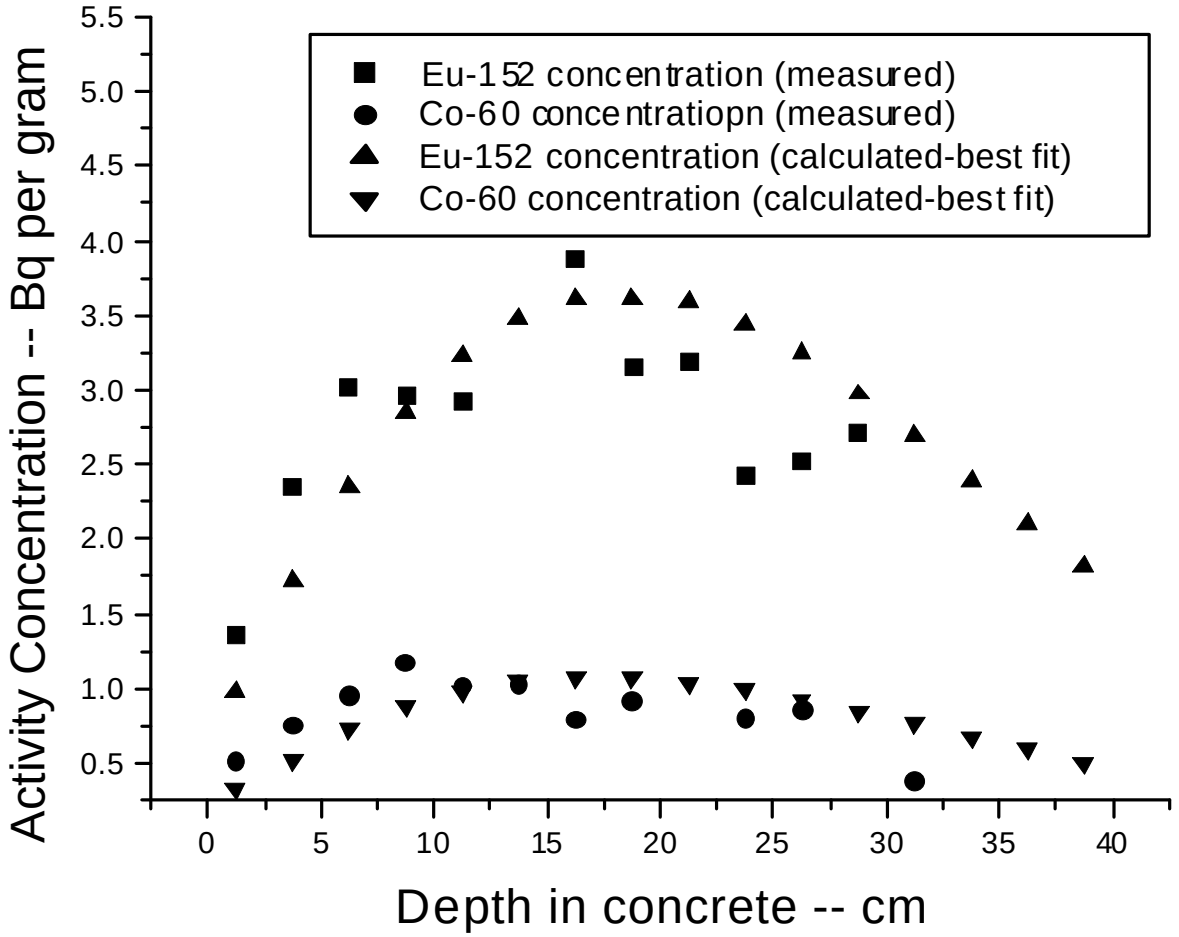

Fig. 1: ^{152}Eu and ^{60}Co concentrations as a function of depth in concrete 11 years post-irradiation.

The fluence and spectrum of forward-directed neutrons resulting from 42 MeV proton bombardment of copper are derived from experimental data by others[2]. The next step is to insert the (now known) fluence and spectrum of neutrons from the original target irradiation into our radiation transport code **TART98** [3] which, in turn, provides a tally of neutron-induced activation reactions.

It is then a simple matter to vary the concentrations of trace elements in the concrete formula used in our computer model, and repeat the calculations until the average (within the first 30 cm of concrete depth) in the computed result is forced to match the average (after correcting for isotope decay) which was actually measured in 1992 [4] . See Fig. 1.

The measured data are made to match the TART98 simulation results when concentrations of 0.294 parts per million by weight for natural (stable) Europium and 2.54 parts per million by weight for natural Cobalt[5] are chosen.

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[2] Thomas Marshall Amos Jr., **"Neutron Yields from Proton Bombardment of Thick Targets"**, Ph.D. Thesis in Nuclear Physics, Michigan State University, 1972. University Microfilms International, Ann Arbor, MI, #73-12,658.

[3] **"TART98 – A Coupled Neutron-Photon Monte Carlo Transport Code"** by D. E. Cullen, University of California Lawrence Livermore National Laboratory. Available from RSICC, Oak Ridge National Laboratories, Oak Ridge, TN.

[4] J. E. Cehn and L. R. Carroll; "Fast-Neutron Activation and Ultimate Decommissioning of a Cyclotron Vault: 1981-1996" in **Health Physics of Radiation-Generating Machines; Proceedings of the 30th Midyear Topical Meeting of the Health Physics Society**, San Jose, CA, January, 1997.

[5] For reference, the concentration by weight of these elements averaged over the entire earth's crust, are 2.1 ppm by weight for Europium and 29 ppm by weight for Cobalt. See N.N. Greenwood and A. Earnshaw, **Chemistry of the Elements**, Pergamon Press, 1990. ISBN 0-08-022057-6

Element	Symbol	Atomic Fraction
Hydrogen	1001	0.1047
Oxygen	8016	0.584
Magnesium	12000	0.0157
Aluminum	13027	0.0317
Silicon	14000	0.2115
Calcium	20000	0.0479
Iron	26000	0.01
Europium	63000	3.7×10^{-8}
Cobalt	27059	0.826×10^{-6}

Table 2: Material definition for "normal concrete" used in TART98 simulations.

We entered these parameters (concentrations are expressed as "atomic fractions" per TART98 input data format) into the concrete recipe in the simulation program (See table 2), and repeated the TART98 calculations for three different irradiation conditions, as shown in Fig. 2 below:

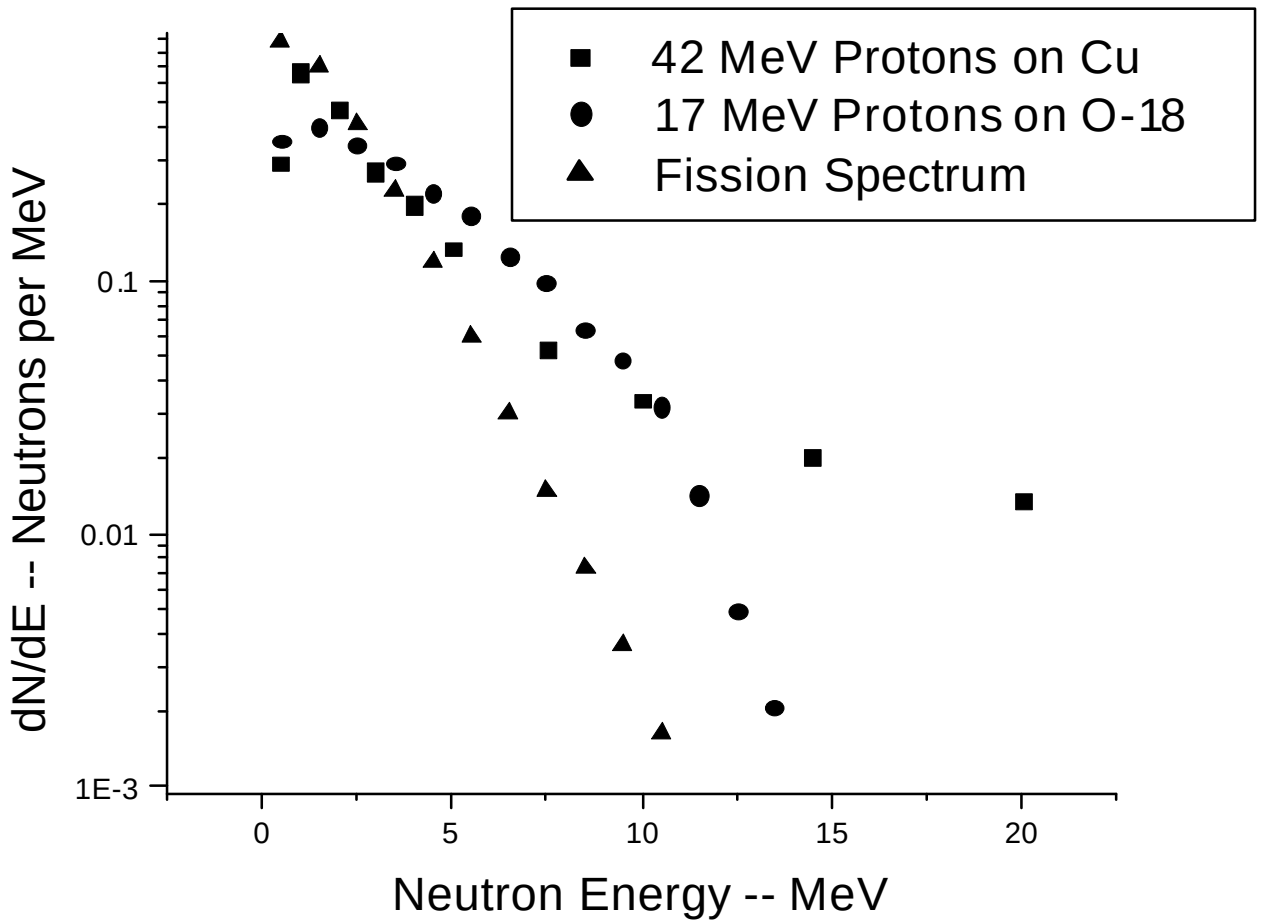

Fig. 2: TART98 Neutron Spectra

The three target irradiation conditions are:
1) neutrons from 42 MeV protons on copper,
2) neutrons from 17 MeV protons on $H_2{}^{18}O$, and
3) a fission neutron spectrum

The spectrum for 42 MeV protons on Copper, obtained from ref.[2] cited earlier, is truncated at 20 MeV since TART98, like other commonly-used neutron transport codes, currently accepts neutron energies up to 20 MeV. This introduces only a slight error, however, since the great majority of source neutrons are less than 10 MeV, and only a very small percentage of the total exceeds 20 MeV.

The "thick target" neutron spectrum for 17 MeV protons on $H_2{}^{18}O$ is constructed from a weighted sum of "thin target" spectra which, in turn, are computed using **ALICE-91** an "Evaporation Model" nuclear-reaction computer code.[6]

The fission spectrum is approximated by the expression[7]:

$$S_f(E) = E^{1/2}\exp^{-E\,/\,1.29}$$

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[6] M Blann, **PSR-146 / ALICE-91** Code Package, Available from RSICC, Oak Ridge National Laboratories, Oak Ridge, TN.

[7] Section 82 in **Neutrons and Related Gamma Ray Problems; Volume XXXVIII/2** in **Encyclopedia of Physics**, edited by S. Flügge, Springer-Verlag, Berlin, Göttingen, Heidelberg, 1959

All spectra are normalized so that the area under their respective curves = 1.0. Somewhat surprisingly, there is very little difference in the shape of the neutron spectrum for 42 MeV protons on Copper, versus 17 MeV protons on ^{18}O-water. This is reflected, in turn, by the "per-neutron" activation profiles shown in Fig 3, in which the data for Ep = 17 MeV are almost indistinguishable from the data for Ep = 42 MeV.

The activation-reaction profile from fission-neutrons, which have a substantially lower mean energy, does peak more toward the surface of the concrete, since lower-energy neutrons do not penetrate as far before thermalizing and finally being absorbed.

The shape of the spectrum governs the depth-profile of activation but -- more importantly – it is ***the number of neutrons*** in concrete which determines the overall average level of activation. A simple -- albeit overly optimistic -- estimate of the number of neutrons produced per second by a cyclotron target is just the saturation yield of the isotope being produced. However, the number of possible reaction channels increases very rapidly with increasing energy. For example, in the reaction ^{18}O(p,n)^{18}F at Ep = 17 MeV ALICE-91predicts almost four times as many neutrons produced per second as ^{18}F atoms per second. Experimentally-measured neutron production data are not always available; when in doubt, one is well-advised to use a code such as ALICE-91 to obtain as accurate an estimate as possible of total neutron yield.

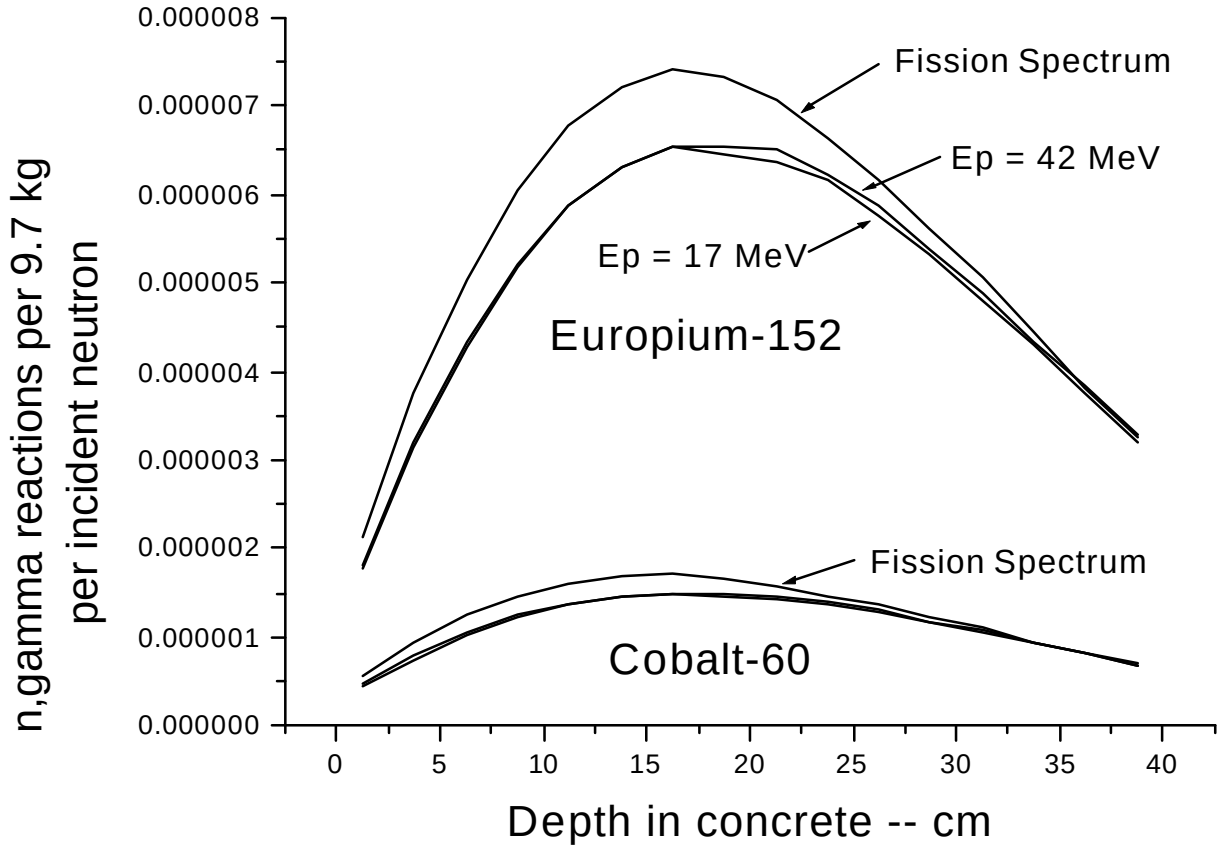

Fig. 3: TART98 Activation Profile per incident neutron per "standard" mass (9.7 kg) of concrete for various neutron spectra.

Comparison with Other Data

The data in Table 3 below, excerpted from [1], shows the results of an assay of concrete in an accelerator vault in 1985 as reported by Phillips, et al, at the University of Colorado. The data were recorded ~one year post-shut-down, after operating a cyclotron there for 21 years[8] .

When back-corrected for isotope decay, our levels of ^{152}Eu and ^{60}Co activation are almost 2 orders of magnitude higher than the corresponding data in Table 3, due to a much higher average neutron flux in the samples tested. Further, after back-correcting to 1-year post-irradiation to better match the Colorado conditions[9], our ratio of concentration of ^{152}Eu / ^{60}Co is ~1.51.

Our data in Fig. 1 were obtained from relatively small (~10 gm) samples of concrete dust recovered at various depths from small-diameter (3/4") bore-holes. In our assay, (taken 11 years post-irradiation) activity concentrations of ^{134}Cs and ^{154}Eu were too low for reliable quantitation. The data in Table 3, in contrast, were taken only 1 year post-shut-down using much larger (10 cm x 20 cm x 40 cm) concrete slabs.

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[8] The listing in Table 3 for naturally-occurring ^{40}K ($t_{1/2}$ = 1.3 x 10^9 years) is not an activation product, but was included for comparison by the authors of ref. 1.

[9] Moreover, the 42 MeV activation occurred almost instantaneously (relative to the half-lives of the various products) while the activation in the University of Colorado vault built up gradually. Had both irradiations been equivalent (i.e., gradual, over 21 years) our corrected ^{152}Eu / ^{60}Co ratio would be higher still; ~ 2.4.

Isotope	Activity
^{60}Co	110 Bq kg^{-1}
^{134}Cs	37 Bq kg^{-1}
^{152}Eu	89 Bq kg^{-1}
^{154}Eu	11 Bq kg^{-1}
^{40}K [8]	300 Bq kg^{-1}

Table 3: Long-term activation products in concrete in the walls of another cyclotron vault.

It shouldn't be too surprising that trace elements which are present in concentrations of fractional parts -- to a few parts -- per million in concrete should vary by 50% to 100% or more from sample to sample.

Notwithstanding these potential sources of uncertainty, we shall endeavor to estimate the build-up of activation in concrete which is in close proximity to a PET cyclotron target during many years of commercial service. Back-projecting our data from Fig. 1 for ^{152}Eu and ^{60}Co, and scaling in proportion to the data from the University of Colorado (Table 3) for ^{154}Eu and ^{134}Cs, we present Fig. 4:

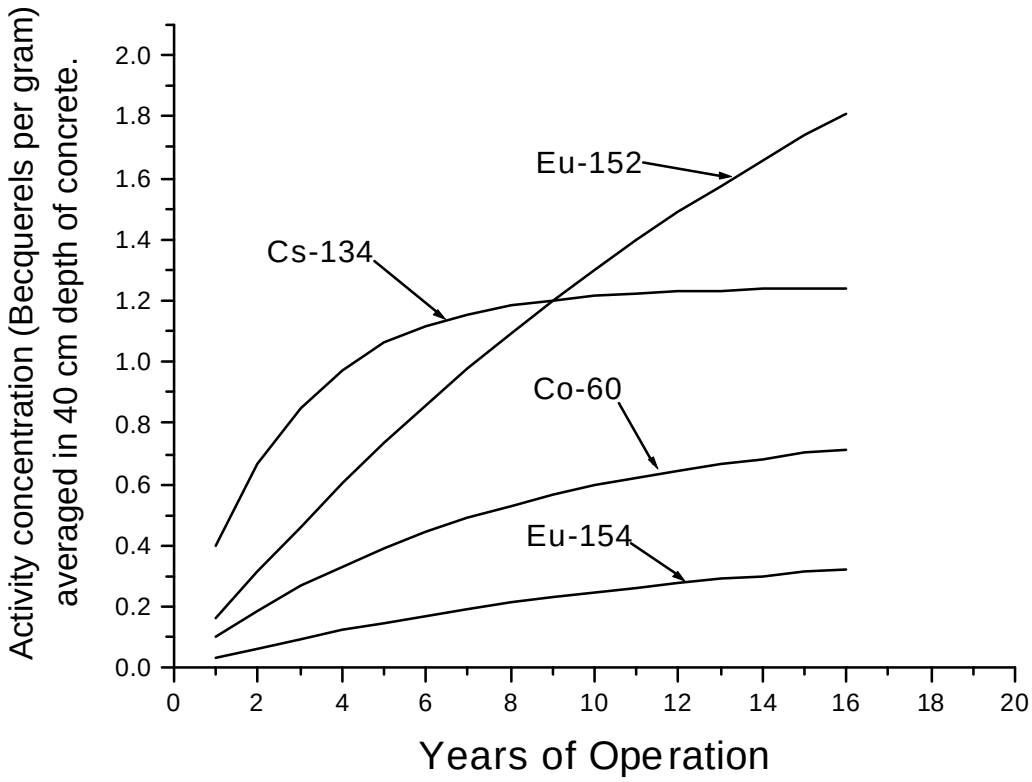

Fig. 4: Build-up of long-term, neutron-induced activity in concrete close to the target.

Assumptions underlying the plots in Fig. 4 are:

1) 80,000 µA-hours per year proton bombardment of ^{18}O water target at Ep = 17 MeV.
2) 18" - 30" distance from target to the surface of a concrete wall or floor.
3) No additional shielding between target and concrete.
4) The concrete in our sample is reasonably (within the limitations discussed earlier) representative of all concrete.

If the same bombardment conditions are maintained for many years (several half-lives of the longest-lived product) the average activity concentrations in the concrete nearest the target eventually level off at the "saturation activity concentrations", which are as shown in table 4:

Isotope	**Average Conc.**	**Total in 6 ft. dia. x 1.5 ft. deep plug**.
^{152}Eu	3.22 Bq gm^{-1}	~ 0.24 mCi
^{154}Eu	0.447	~ 0.032 mCi
^{60}Co	0.814	~ 0.06 mCi
^{134}Cs	1.24	~ 0.0925 mCi

Table 4: Saturation Activities close to the target

The build-up of radioactivity averaged over the interior of the vault as a whole -- away from the target -- is obviously much lower than that in the hottest region near the target. Moreover, as shown in Table 3, concrete already contains radioactive (but naturally-occurring) ^{40}K ($t_{1/2}$ = 1.3 x 10^9 years) at a concentration of ~0.3 Becquerel per gram. Thus, areas of concrete which are far from the target should maintain concentrations of induced activity at or below that of naturally-occurring radioactivity.

Conclusions and recommendations

Cyclotron-based production of radioisotopes on a commercial scale can result in significant activation of concrete which is near the target if operation is conducted over many years. Unless preventive measures are taken to reduce the neutron flux which is incident on the wall or floor nearest the target, there will be a gradual build-up of radioactivity which may ultimately require remediation by removal of concrete (and disposal as rad-waste) up to a depth of 18 inches and approximately 2 - 3 ft. radius around the target(s). Beyond that radius, the activity concentration should be low enough to qualify for disposal as rubble in an ordinary landfill – if that is necessary to reduce the ambient dose-rate in the room to a normal background level.

To reduce this build-up of radioactivity in the concrete, the area immediately surrounding the target should -- at the outset -- be stacked with neutron-absorbing material such as borated polyethylene (2 - 5% boron by weight) or equivalent.

For example, for a medium-energy (Ep = 16 to 18 MeV) PET cyclotron, one foot of borated polyethylene will reduce the number of neutrons reaching the concrete wall by a factor of ~30,

so that the “hot spot” activity concentration will be maintained at or below the level of naturally-occurring ^{40}K, and the induced activity away from the hottest spot will be 1-2 orders of magnitude lower.