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RECORD #250

TITLE: Monitoring at nuclear power plants for contamination by radionuclides that decay by electron capture



UNITED STATES
NUCLEAR REGULATORY COMMISSION
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May 28, 1992

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FROM: LeMoine J. Cunningham, Chief
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SUBJECT: MONITORING AT NUCLEAR POWER PLANTS FOR CONTAMINATION BY
RADIONUCLIDES THAT DECAY BY ELECTRON CAPTURE

In a June 18, 1991 memorandum, I provided some preliminary information concerning the subject of this memorandum and asked for regional feedback of additional information.

The enclosure to this memorandum provides information concerning monitoring for contamination by electron-capture emitters. This enclosure is a revision of the enclosure to my earlier memorandum.

The information you have provided does not indicate a generic health and safety problem with this type of monitoring. It does indicate (not surprisingly) a wide range, among nuclear power plants, in the fractions of contaminating activity that represent radionuclides that decay by electron capture. Many licensees appear to have recognized that conventional detectors used in hand frisking for contamination by beta emitters (particularly "pancake" GM detectors) have a very low counting efficiency for the x-rays and gamma-rays emitted by electron-capture nuclides. Some licensees have obtained, or are considering obtaining, more efficient detectors (such as proportional counters filled with argon-methane) for monitoring electron-capture nuclides.

Also, from the information provided, some licensees appear to be making improper applications of the numerical criteria in NRC (IE) Circular 81-07 to monitoring for electron-capture nuclides and to automated personnel contamination monitors. (See the section in the enclosure on "Previous NRC Guidance on Monitoring for Contamination").

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As indicated in the section, "NRC Staff Perspective," in the enclosure, the NRC staff continues to be concerned about the potential for unauthorized release of any detectable contamination by licensed material. Licensees should be aware of changes in contamination detection capabilities resulting from changes in the radionuclide composition of the contamination. Licensees need to continue to ensure that unauthorized release of licensed radioactive material in the form of contamination does not result from inadequate onsite monitoring procedures and detection capabilities before release of the material.

This memorandum, with enclosure, is being placed in the NRC Public Document Room so that it may be shared with licensees and others who are interested.

Original signed by
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Enclosure:
As stated

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Information Concerning Monitoring for Contamination
by Radionuclides that Decay by Electron Capture

Introduction

Questions have arisen as to the adequacy of monitoring at nuclear power plants for detection of radioactive contamination before release of materials from a radiologically controlled area, or before disposal of materials as uncontaminated waste, when radionuclides that decay by electron capture constitute a major fraction of the contaminating activity. This question first came to the attention of the NRC staff in relation to BWRs that use zinc injection. [See C.C. Messier and J.E. Laine, "Radiation Protection Considerations for Boiling Water Reactors Utilizing Zinc Injection," Radiation Protection Management B (No.6), pp. 44-56 (1991) for a discussion of how this question was addressed at one BWR.]

Radioactive Decay Data

Attachment 1 is a table of radioactive decay data for the longer-lived neutron activation products of chromium, manganese, iron, cobalt, nickel and zinc. This table shows the principal radiations emitted in the decay of these radionuclides and the intensity (as a percentage of the disintegrations) of these radiations. Of the nine radionuclides in the table, only two (Fe-59 and Co-60) emit both beta and gamma radiation. Two (Co-58 and Zn-65) emit positrons, x-rays and gamma rays. Two (Cr-51 and Mn-54) emit x-rays and gamma rays. Two (Fe-55 and Ni-59) emit x-rays only. One (Ni-63) emits only weak beta radiation. The six radionuclides that emit x-rays (Cr-51, Mn-54, Fe-55, Co-58, Ni-59 and Zn-65) decay by electron capture.

Detection of Electron-Capture Nuclides

Attachment 2 briefly summarizes information in four publications concerning detection of contamination by, and the relative external radiation hazard of, electron-capture nuclides.

The information in the first two publications indicates that gas-filled (GM and proportional counter) detectors that are customarily used for detection of beta radiation, and that are filled with a gas of low atomic number, are poor detectors for electron-capture nuclides that do not emit beta radiation. Electron-capture nuclides that emit relatively abundant gamma radiation (e.g., Zn-65) can best be detected using a gamma-sensitive detector such as NaI(Tl). Electron-capture nuclides that emit only characteristic x-rays can best be detected using gas-filled counters containing a high atomic number counting gas (e.g., argon). X-ray spectrometry can be performed in the laboratory using Si(Li) detectors.

For intercomparison of hand-held detectors (probes), a figure of merit for measuring a contaminated area less than the areas of the probes is $S^2/(S+B)$, where S is the net count rate caused by the contamination and B is the mean

background count rate. If the contaminated area to be measured is greater than the area of the probes, the figure of merit is $S^2A/(S+B)$, where A is the area of the probe window. (See the summary of the fourth publication in Attachment 2)

Hazard from External Sources of Radiation from Electron Capture Nuclides

The third publication in Attachment 2 indicates that external sources of K x-rays provide no serious problem in practical radiation protection because this type of radiation will be of importance in relatively few cases, and the resulting radiation dose or dose rate can be measured in ways similar to those used to measure low-energy beta doses and dose rates in most cases.

Hazard from Intakes of Electron-Capture Nuclides

Attachment 3 is a table that gives an indication of the relative significance of electron-capture nuclides as an internal radiation hazard. Data are presented for the electron-capture nuclides listed in Attachment 1 and for the beta-emitters Co-60 and Cs-137. This table lists the dose conversion factors (committed effective dose equivalent per unit radionuclide intake) on both an absolute basis (in units of Sv/Bq) and a relative basis (normalized to Co-60). These dose conversion factors indicate that, in general, intakes of the electron-capture nuclides are less hazardous, by an order of magnitude or more, than comparable intakes of Co-60 or Cs-137.

However, as recognized in ICRP Publication 60 (paragraphs 26 and B67), Auger electrons, which are emitted in the decay of electron-capture nuclides, may have values of the relative biological effectiveness (RBE) considerably higher than those for other electrons. In cases where the radionuclide does not penetrate the cell, Auger electron emitters are very inefficient in producing biological effects because of the short range of these low-energy electrons. In those Auger electron emitters that penetrate the cell, but are not incorporated into DNA, RBEs for a range of end points, including cell killing, were found between 1-5 and 8. For Auger emitters incorporated into DNA, much higher RBE values of 20-40 have been found for end points such as cell transformation. The effects of Auger electrons have to be assessed by the techniques of microdosimetry. Recognizing that Auger electrons emitted from nuclei bound to DNA present a special problem, ICRP Publication 60 excludes them from the class of "electrons and muons, all energies" to which ICRP assigns a radiation weighting factor of 1. Other than this limited guidance from ICRP, the possibly higher radiotoxicity of electron-capture emitters incorporated into DNA has not yet been reflected in radiation protection standards.

NRC Staff Perspective

From a general health and safety perspective (which does not consider the "Auger effect" discussed in the preceding paragraph), for a given level of contamination, an increase in the percentage of electron-capture nuclides in nuclear power plant surface contamination represents a decrease, or at least no increase, in radiological hazard. Nevertheless, from a regulatory

perspective, the NRC staff is concerned about the potential for unauthorized release of any detectable contamination by licensed radioactive material. The detection of such a release inevitably has an adverse effect on public relations and results in significant expenditures of staff time by both the NRC and licensees even when the radiological hazard is slight.

Previous NRC Guidance on Monitoring for Contamination

NRC (IE) Circular 81-07 provides guidance on monitoring for surface contamination by "beta-gamma" and alpha emitters. As indicated in that circular, and in NRC (IE) Information Notice 85-92, the numerical criteria included in that circular (e.g., a detection capability of 5000 dpm/100 cm² for total "beta-gamma" contamination) are based on considerations of hand frisking with portable survey instruments equipped with thin-window (relatively small area) "pancake" GM detectors that respond primarily to beta radiation and that are relatively insensitive to x-rays and gamma rays. Thus the numerical criteria were not intended for, and are not appropriate for, surveys for contamination by radionuclides (or a mixture of radionuclides) that emit photons but that emit little or no beta radiation. The staff does not plan to develop new numerical criteria for detection of photons, whether x-rays or gamma-rays, in contamination surveys. The qualitative guidance in Circular 81-07 and Information Notice 85-92 is applicable to all surveys for contamination of materials before release to unrestricted areas. However, the guidance in Circular 81-07 and Information Notice 85-92, for the detection of contamination of materials, is not intended to be applied to automated personnel contamination monitors, which are used for detection of contamination of workers. In any case, the numerical criteria of Circular 81-07, which are expressed in terms of activity per unit area, are not applicable to measurements of the total activity of the contamination on either materials or workers.

Radioactive Decay Data for Neutron Activation Products
of
Cr, Mn, Fe, Co, Ni and Zn
(from D.C. Kocher, Radioactive Decay Data Tables
DOE/TIC 11026, 1981)

<u>Radionuclide</u>	<u>Half-Life</u>	<u>Type of Radiation*</u>	<u>Energy (keV)</u>	<u>Intensity (%)</u>
Cr-51	27.7d	K x-rays gamma-ray	4.9 320.	22. 9.8
Mn-54	313.d	K x-rays gamma-ray	5.4 834.	25. 100.
Fe-55	2.7y	K x-rays	5.9	28.
Fe-59	44.6d	betas gamma-ray	118.(avg.) 1099. 1292.	100. 56. 43.
Co-58	70.8d	positrons K x-rays gamma-ray	201.(avg) 6.4 811.	15. 26. 99.
Co-60	5.27y	betas gamma-ray	96.(avg.) 1173. 1332.	100. 100. 100.
Ni-59	7.5x10 ⁴ y	K x-rays	6.9	34.
Ni-63	100y	betas	17.(avg.)	100.
Zn-65	244d	positrons K x-rays gamma-ray	143.(avg.) 8.0 1116.	1.4 39. 50.8

*Note: Auger electrons and L x-rays, which are listed in Kocher's tables, are not listed here because their energies are so low that they are not detectable with any portable contamination survey instruments.

Four Publications Concerning
Surface Contamination by Radionuclides
That Decay Primarily by Electron Capture

1. D. Bush and R. C. Hundal, "Difficulties in measuring the maximum permissible levels of surface contamination for radionuclides that decay primarily by electron capture," Health Physics 21, 651-655 (Nov. 1971).

The authors of this paper were at the University of Birmingham, in England. They measured the response of a number of detectors in common use at that university to radiations from Cr-51, Mn-54, Zn-65, and I-125. All of these radionuclides emit both characteristic x-rays and gamma radiation. The detectors included thin-window GM tubes, a thin plastic scintillator, and a 1½ in. x 1 in. NaI(Tl) detector. The authors found that detectors "commonly used in contamination monitors, such as GM tubes and thin plastic scintillators," cannot reliably detect surface contaminants such as Cr-51, Mn-54, Zn-65 and I-125 at levels of uniform contamination of 10^{-4} microcuries per cm^2 (222 dpm/ cm^2 or 22,200 dpm/100 cm^2). 10^{-4} microcuries per cm^2 was a United Kingdom limit on surface contamination at that time. They stated that "only by selecting appropriately sized scintillators such as thallium-activated sodium iodide crystals, with careful choice of operating conditions, can such levels be reliably detected in the presence of quite low backgrounds."

2. M. Heinzelman and H. Schuren, "Studies on the detection of surface contamination by electron capture emitters," Radiation Protection Dosimetry 29 (No. 3), 183-188 (1989).

The authors of this relatively recent paper are at the Nuclear Research Center, Julich, Germany. They note that contamination in German nuclear power plants often contains Cr-51, Mn-54, Fe-55, Co-58 and Zn-65. They show that large area (330 mm x 220 mm) proportional counters of a type commonly used (in Germany) for detection of surface contamination can detect surface contamination of less than 5 Bq/ cm^2 (300 dpm/ cm^2 or 30,000 dpm/100 cm^2) of electron-capture emitters if argon-methane (90% argon) is used as the counter gas. Argon-methane was selected because of the higher atomic number of argon in comparison to methane; the higher atomic number gives a highly effective cross-section (or absorption coefficient) for absorption of the characteristic K x-rays of the electron-capture nuclides. The response to Cl-36, a pure beta emitter, is said to be about ten times as great as the response to Fe-55. The authors report a 4% counting efficiency for Fe-55 with this detector. Under German regulations, the limits for surface contamination by beta emitters are ten times the limits for K x-ray (from electron capture) emitters. Thus the count rate with the large-area proportional counter filled with argon-methane is said to be proportional to the contamination hazard of either beta emitters or K X-ray emitters.

3. F. Rohloff and M. Heinzelmann, "Absorbed dose rate due to K radiation," Radiation Protection Dosimetry 27 (No. 3), 193-196 (1989).

The dose rates from K x-rays emitted in the decay of Cr-51, Mn-54, Fe-55, Co-58 and Zn-65 (as external radiation sources) were calculated and compared to the dose rates from beta and gamma radiation. The results of these calculations are provided. For example, measurements were made of a radiation source consisting of 97% of nuclides decaying by electron capture (the highest percentage of electron-capture nuclides found in measurements of contamination at German nuclear power plants) and containing 92% Cr-51, 1% Mn-54, 2% Co-58, and 2% Zn-65 and 3% Co-60. The dose rate from beta radiation was found to be greater than that from K x-rays up to distances of 60 cm from the source; at this distance the dose rate from K x-rays was only 50% of that from gamma radiation. The authors conclude that K x-radiation from external sources will provide no serious problem with regard to health physics because this kind of radiation will only be of importance in a few cases and it can be measured like low-energy beta radiation, except for the use of non-tissue-equivalent TLDs.

4. D. R. McClure, C. E. Hill, et al., Evaluation of Some Hand-Held Instruments for Measuring Surface Contamination, NRPB-R232, (National Radiological Protection Board, United Kingdom)

This National Radiological Protection Board (NRPB) a report giving the results of an evaluation of 22 hand-held radiation monitors for measuring surface contamination. The report can be used as a guide to the most suitable instrument for a user's particular needs.

The responses of instruments to a wide range of large- and small-area alpha, beta and x-ray sources were measured, and ratings are given in the report to indicate the level of compliance with the requirements for monitoring contamination under the United Kingdom (UK) Ionizing Radiations Regulations 1985. However, the instrument responses were not measured for x-ray sources with x-ray energies in the range of 4.9 to 8 keV (see Attachment 1); the only x-ray source used was I-125, which emits K x-rays of 27-31 keV.

The report derives figures of merit for intercomparison of hand-held probe (detector) performance in terms of the mean background count rate, B , and the net count rate, S , caused by contamination under the probe window. If the contaminated area to be measured is less than the area of the probe, the figure of merit is $S^2/(S+B)$. If the contaminated area to be measured is greater than the areas of the probes, the figure of merit is $S^2A/(S+B)$, where A is the area of the probe window. The contamination monitors were subjected to a wide range of tests of their capabilities. The radiation, electrical and environmental characteristics were considered, together with those aspects of construction that make an

instrument most convenient for routine use. The range of tests performed were based broadly on the appropriate recommendations of the International Electrotechnical Commission (IEC).

Committed Effective Dose Equivalent Factors (Sv/Bq)

(from Federal Guidance Report No. 11,
 "Limiting Values of the Radionuclide Intake and Air Concentration
 and Conversion Factors for Inhalation, Submersion, and Ingestion.")

Radionuclide	Principal Radiations	Committed Effective Dose Equivalent per Unit Intake			
		Inhalation		Ingestion	
		Sv/Bq	Relative(%)*	Sv/Bq	Relative(%)*
Co-60	β^- , γ	5.91×10^{-8}	100.	7.28×10^{-9}	100.
Cs-137	β^- , γ	8.63×10^{-9}	14.6	1.35×10^{-8}	185.
Fe-59	β^- , γ	4.00×10^{-9}	6.8	1.81×10^{-9}	24.9
Co-58	β^+ , x, γ	2.94×10^{-9}	5.0	9.68×10^{-10}	13.3
Zn-65	β^+ , x, γ	5.51×10^{-9}	9.3	3.90×10^{-9}	53.6
Cr-51	x, γ	9.03×10^{-11}	0.15	3.98×10^{-11}	0.5
Mn-54	x, γ	1.81×10^{-9}	3.1	7.48×10^{-10}	10.3
Ni-63	β^-	8.39×10^{-10}	1.4	1.56×10^{-10}	2.1
Fe-55	x	7.26×10^{-10}	1.2	1.64×10^{-10}	2.3
Ni-59	x	3.58×10^{-10}	0.6	5.67×10^{-11}	0.8

*"Relative (%)" is the dose factor normalized to Co-60, expressed as a percentage of the Co-60 value.