

Designation: D5411 - 10

Standard Practice for

Calculation of Average Energy Per Disintegration $(\overline{E})(\overline{E})$ for a Mixture of Radionuclides in Reactor Coolant¹

This standard is issued under the fixed designation D5411; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

1. Scope

- 1.1 This practice applies to the calculation of the average energy per disintegration (E)(E) for a mixture of radionuclides in reactor coolant water.
- 1.2 The values stated in inch-pound units are to be regarded as<u>microcurie</u> (µCi) is the standard unit of measurement for this standard. The values given in parentheses are mathematical conversions to SI units, which are provided for information only and are not considered standard.
- 1.3 This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.

2. Referenced Documents

2.1 ASTM Standards:²

D1066 Practice for Sampling Steam

D1129 Terminology Relating to Water

D3370 Practices for Sampling Water from Closed Conduits

D3648 Practices for the Measurement of Radioactivity Practices for the Measurement of Radioactivity

D7282 Practice for Set-up, Calibration, and Quality Control of Instruments Used for Radioactivity Measurements

2.2 Code of Federal Regulations:

10CFR100 Reactor Cite Criteria

10 CFR 100 Reactor Site Criteria³

3. Terminology

ASTM D5/111 10

3.1 Definitions—For definitions of terms used in this practice, refer to Terminology D1129. 47490/astm-d5411_10

4. Summary of Practice

4.1 The average energy per disintegration, $\bar{E}\underline{\bar{E}}$ (pronounced E bar), for a mixture of radionuclides is calculated from the known composition of the mixture. $\bar{E}\underline{\bar{E}}$ is computed by calculating the total beta/gamma energy release rate, in MeV, and dividing it by the total disintegration rate. The resultant $\bar{E}\underline{\bar{E}}$ has units of MeV per disintegration.

5. Significance and Use

5.1 This practice is useful for the determination of the average energy per disintegration of the isotopic mixture found in the reactor-coolant system of a nuclear reactor (1). The resultant value is periodically reported upon, by the operators of nuclear power plants, in order to ensure that the 2-h radiation dose, measured at the plant boundary, will not exceed an appropriately small fraction of the Code of Federal Regulations, Title 10, part 100 dose guidelines.

5.2In calculating E, all the energy dissipated by charged particles and photons in each nuclear radioactive transformation is

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² For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

³ Available from Standardization Documents Order Desk, Bldg, 4 Section D, 700 Robbins Ave., Philadelphia, PA 19111-5094, Attn: NPODS.

⁴ The boldface numbers in parentheses refer to a list of references at the end of this practice.



included. This accounting includes the energy released in the form of beta particles and gamma rays as well as energy released from extra-nuclear transitions in the form of X-rays, Auger electrons, and conversion electrons. However, not all radionuclides present in a sample are included in the calculation of \bar{E} .

- 5.3Individual, nuclear reactor, technical specifications vary and each nuclear operator must be aware of limitations affecting their operation. Typically, radio-iodines, radionuclides with half lives of less than 10 min (except those in equilibrium with the parent), and those radionuclides, identified using gamma spectrometry, with less than a 95% confidence level, are not typically included in the calculation. However, the operator must account for at least 95% of the remaining activity. There are individual bases for each exclusion.
- 5.3.1Radio-iodines are typically excluded from the calculation of \bar{E} because many commercial nuclear reactors are required to operate under a more conservative restriction of 1 microCurie (37 kBq) per gram dose equivalent I-131 in the reactor coolant.
- 5.3.2Excluding radionuclides with half-lives less than 10 min, except those in equilibrium with the parent, has several bases.
- 5.3.2.1The first basis considers the nuclear characteristics of a typical reactor coolant. The radionuclides in a typical reactor coolant have half-lives of less than 4 min or have half-lives greater than 14 min. This natural separation provides a distinct window for choosing a 10 min half-life cutoff.
- 5.3.2.2The second consideration is the predictable time delay, approximately 30 min, which occurs between the release of the radioactivity from the reactor coolant to its release to the environment and transport to the site boundary. In this time, the short-lived radionuclides have undergone the decay associated with several half-lives and are no longer considered a significant contributor to E.
- 5.3.2.3A final practical basis is the difficulty associated with identifying short-lived radionuclides in a sample that requires some significant time, relative to 10 min, to collect, transport, and analyze.
- 5.3.3Radionuclides identified using less than a 95% confidence level are not typically included in the calculation to improve the accuracy of the calculation (2) The \overline{E} value is used to calculate a site-specific activity limit for the reactor coolant system, generally identified as

Alimiting = K/E

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where

 $\underline{K} =$ a power reactor site specific constant (usually in the range of 50 to 200).

The activity of the reactor coolant system is routinely measured, then compared to the value of $A_{limiting}$. If the reactor coolant activity value is less than $A_{limiting}$ then the 2-h radiation dose, measured at the plant boundary, will not exceed an appropriately small fraction of the Code of Federal Regulations, Title 10, part 100 dose guidelines. It is important to note that the measurement of the reactor coolant system radioactivity is determined at a set frequency by use of gamma spectrometry *only*. Thus the radionuclides that go into the calculation of \overline{E} and subsequently $A_{limiting}$ are only those that are calculated using gamma spectrometry.

- 5.2 In calculating \overline{E} , the energy dissipated by beta particles (negatrons and positrons) and photons from nuclear decay of beta-gamma emitters. This accounting includes the energy released in the form of energy released from extra-nuclear transitions in the form of X-rays, Auger electrons, and conversion electrons. However, not all radionuclides present in a sample are included in the calculation of \overline{E} .
- 5.3 Individual, nuclear reactor, technical specifications vary and each nuclear operator must be aware of limitations affecting their plant operation. Typically, radioiodines, radionuclides with half lives of less than 10 min (except those in equilibrium with the parent), and those radionuclides, identified using gamma spectrometry, with less than a 95 % confidence level, are not typically included in the calculation. However, the technical requirements are that the reported activity must account for at least 95 % of the activity after excluding radioiodines and short-lived radionuclides. There are individual bases for each exclusion.
- 5.3.1 Radioiodines are typically excluded from the calculation of \bar{E} because United States commercial nuclear reactors are required to operate under a more conservative restriction of 1 μ C (37 kBq) per gram dose equivalent ¹³¹I (DEI) in the reactor coolant.
- 5.3.2 Beta only emitting radio isotopes (for example, ⁹⁰Sr or ⁶³Ni) and alpha emitting radioisotopes (for example, ²⁴¹Am or ²³⁹Pu) which comprise a small fraction of the activity, should not be included in the E-bar calculation. These isotopes are not routinely analyzed for in the reactor coolant, and thus their inclusion in the E-bar calculation is not representative of what is used to assess the 10 CFR 100 dose limits. Tritium, also a beta only emitter, should not be included in the calculation. Tritium has the largest activity concentration in the reactor coolant system, but the lowest beta particle energy. Thus its dose contribution is always negligible. However its inclusion in the E-bar calculation would raise the value of A_{limiting}, yielding a non-conservative value for dose assessment.
 - 5.3.3 Excluding radionuclides with half-lives less than 10 min, except those in equilibrium with the parent, has several bases.
- 5.3.3.1 The first basis considers the nuclear characteristics of a typical reactor coolant. The radionuclides in a typical reactor coolant have half-lives of less than 4 min or have half-lives greater than 14 min. This natural separation provides a distinct window for choosing a 10-min half-life cutoff.



- 5.3.3.2 The second consideration is the predictable time delay, approximately 30 min, which occurs between the release of the radioactivity from the reactor coolant to its release to the environment and transport to the site boundary. In this time, the short-lived radionuclides have undergone the decay associated with several half-lives and are no longer considered a significant contributor to \overline{E} .
- 5.3.3.3 A final practical basis is the difficulty associated with identifying short-lived radionuclides in a sample that requires some significant time, relative to 10 min, to collect, transport, and analyze.
- 5.3.4 The value of E-bar is usually calculated once every 6 months. However, anytime a significant increase in the activity of the reactor coolant occurs, the value of E-bar should be reassessed to ensure compliance with 10 CFR 100. Such reassessment should be done any time there is a significant fuel defect that would alter the \overline{E} value and affect $A_{limiting}$. The two possible causes to reassess the value of \overline{E} would be:
 - (1) A significant fuel defect has occurred where the noble gas activity has increased.
 - (2) A significant corrosion product increase has occurred.

For the case of a fuel defect, the plant staff may need to include new radionuclides not normally used in the calculation of \overline{E} such as 239 U and 239 Np.

6. Interferences

- 6.1There are no true interferences to this practice. However, errors may result in the calculation of \bar{E} from incorrectly analyzing the sample mixture.
- 6.1 The analytical determination of the radionuclides used for this calculation is made by gamma ray spectrometry. Commercially available software is generally used to perform the spectrum analysis and data reduction. However there can be significant number of interferences from gamma ray emitters with multiple gamma ray emissions. The user must carefully select the appropriate interference free gamma ray energy for each radionuclide in order to determine accurately the activity of each radionuclide. As a specific example 56 Mn ($t_{1/2} = 2.6$ h) has a gamma ray energy of 847 keV and 134 I ($t_{1/2} = 53$ min) also has a gamma ray energy of 847 keV. The 847 keV gamma ray is also the most abundant for each of these radionuclides. It would be inaccurate to use the 847 keV gamma ray for the determination of either of these radionuclides.

7. Sampling

- 7.1 If samples are collected for analysis in support of this practice they should be representative of the matrix, be of sufficient volume to ensure adequate analysis, and be collected in accordance with Practices D1066, D3370, and D3648.
- 7.2 In addition to the requirements of 7.1, if samples of reactor coolant are required in support of this practice, they should typically be collected only after a minimum of 2 effective full-power days and 20 days of power operation have elapsed since the reactor was <u>last</u> subcritical for 48 h or longer. Individual nuclear operator technical specifications (or now for many plants called "technical requirements") vary and should be reviewed to determine specific requirements.

8. Calibration and Standardization

8.1 Any calibrations and standardizations required in support of this practice should be in accordance with the applicable sections of Practices D3648 -and D7282 and in accordance with the manufacturer's specifications for the gamma spectrometry system used.

9. Procedure

- 9.1 Conduct all analyses in support of this practice in accordance with the applicable sections of Practice D3648.
- 9.2Perform sufficient gamma isotopic analyses of the liquid, gaseous, and suspended fractions of the sample to ensure that at least 95% of the coolant activity due to gamma emitting isotopes has been quantified. Samples should be analyzed at approximately 2 h, 24 h, and 7 days following sample collection. Multiple sample analyses are required to ensure accurate quantification of the longer-lived isotopes because of masking caused by the high initial activity of the sample. If interferences continue to be a concern with the results of the analysis conducted on Day 7, it may be necessary to conduct additional gamma isotopic analyses of the sample at approximately 30 days after collection.
- 9.3Perform sufficient isotopic analyses of the liquid, gaseous, and suspended fractions of the sample to ensure that at least 95% of the coolant activity due to nongamma emitting isotopes has been quantified.
- 9.4Tabulate the concentrations, uniformly measured in μ Ci/cc (37kBq/cc) or μ Ci/g (37kBq/g), of all applicable gamma and nongamma emitting radioisotopes identified in the sample. Some examples of the radioisotopes or types of radioisotopes found in a typical sample are the radioactive noble gases, pure beta emiter such as tritium, carbon-14, strontium-89 and 90, and yttrium-90, beta/gamma emitters such as cobalt-60, electron capture isotopes such as iron-55, and reactor coolant suspended and particulate material (commonly referred to as *crud*).
- 9.2 Perform sufficient gamma isotopic analyses of the liquid, gaseous, and suspended fractions of the sample to ensure that at least 95 % of the coolant activity due to gamma emitting isotopes has been quantified. Samples should be analyzed at approximately 0.5 h, 2 h, 24 h, and 7 days following sample collection. Multiple sample analyses are required to ensure accurate quantification of the longer-lived isotopes because of masking caused by the high initial activity of short-lived radionuclides in the



sample. If interferences continue to be a concern with the results of the analysis conducted on Day 7, it may be necessary to conduct additional gamma isotopic analyses of the sample at approximately 30 days after collection.

9.3 Sample fractions that are going to be stored for recounting (at 24 h, 7 days, or 30 days) should be preserved with at least 2 mL of concentrated nitric acid per litre of sample immediately after the sample is taken to preserve the sample geometry. This mitigates the precipitation of radionuclides or adhesion of radionuclides onto container walls.

9.4 Tabulate the concentrations, uniformly measured in μ Ci/cc (37kBq/cc) or μ Ci/g (37kBq/g), of all applicable gamma radioisotopes identified in the sample. Examples of the most significant contributing radioisotopes to \overline{E} are:

(1) Noble gas fission products: ^{131m}Xe, ¹³¹Xe, ^{133m}Xe.

¹³³Xe, ⁸⁷Kr (others),

(2) Soluble fission products: ¹³⁷Cs, ¹³⁴Cs, ¹⁴¹Ce.

140Ba, 140La, 92Sr (others),

(3) Corrosion activation products: ⁵⁸Co, ⁵⁶Mn, ⁵⁴Mn,

⁶⁰Co, ⁵¹Cr, ⁵⁹Fe, ⁹⁵Zr, ⁹⁵Nb (others),

(4) Miscellaneous radionuclides: ⁴¹Ar, ²⁴Na, ¹⁸F, ⁷Be (others), and

(5) Reactor coolant suspended and particulate material (commonly referred to as crud) will also have the activated products in them and must be included in the calculation

of \bar{E} .

10. Calculation

10.1 Calculate the average energy per disintegration, $\bar{E}_{\tau}E_{\tau}$, in MeV according to the following equation:

(1)
$$E = i = \ln(A i * Ei)i = \ln Ai$$

(1)
$$E = i = \ln (A i * Ei)i = \ln Ai$$

where: **Ē**

= average energy per disintegration, MeV/disintegration,

 A_i = activity of the *i*th radionuclide uniformly measured, μ Ci/cc or μ Ci/g, and

 E_i = isotopic energy emission for the ith radionuclide, MeV/disintegration.

10.2 The values for A; are simply the measured activity levels, uniformly measured in μCi/cc (37 kBq/cc) or μCi/g (37 kBq/g), for each appropriate radionuclide identified in the sample (for example, Co-60, Sr-90, Xe-133, etc.). I are the measured activity levels of a representative sample in µCi/cc (37 kBq/cc) or µCi/g (37 kBq/g), for each appropriate radionuclide identified in the sample (for example, ⁶⁰Co, ¹³³Xe, ¹³⁷Cs, etc.).

10.3 The values for E_i are constant for each radionuclide and depend upon the decay scheme for that radioisotope. E_i is calculated from the following equation:

where:

= the average, abundance weighted, beta energy per disintegration, MeV/disintegration, E_i (beta)

 $E_i(CE)$ = the average, abundance weighted, conversion electron energy per disintegration, MeV/disintegration,

= the average, abundance weighted, Auger electron energy per disintegration, MeV/disintegration, $E_i(A)$

 E_i (gamma) = the average, abundance weighted, gamma energy per disintegration, MeV/disintegration, and

= the average, abundance weighted, X-ray energy per disintegration, MeV/disintegration. $E_i(X)$

10.4 An example for the calculation of E_i for the disintegration of $\frac{133}{2}$ (E_{Xe-133}) follows. 10.4.1 The decay scheme for $\frac{133}{2}$ (3) is given in Fig. 1.

10.4.2 First, calculate E_{Xe-133} (beta).

10.4.2.1 To determine each E_i(beta), multiply the average energy per disintegration for each beta emitted by its abundance and sum the products. The average beta energies for each isotope may be found in the literature (4-6(3, 4). Or, it may be approximated

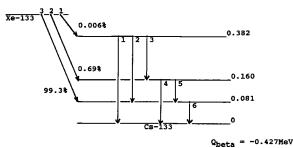


FIG. 1 Decay Scheme for Xe- 133Xe