Designation: D5411 - 21

# Standard Practice for Calculation of Average Energy Per Disintegration (Ē) for a Mixture of Radionuclides in Reactor Coolant<sup>1</sup>

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  $(\bar{E})$  for a mixture of radionuclides in reactor coolant water.
- 1.2 The microcurie ( $\mu$ Ci) 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, health, and environmental practices and determine the applicability of regulatory limitations prior to use.
- 1.4 This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.

## 2. Referenced Documents catalog/standards/sist/0e

2.1 ASTM Standards:<sup>2</sup>

D1066 Practice for Sampling Steam

D1129 Terminology Relating to Water

D3370 Practices for Sampling Water from Flowing Process Streams

D3648 Practices for the Measurement of Radioactivity

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

D7902 Terminology for Radiochemical Analyses

2.2 Code of Federal Regulations: 10 CFR 100 Reactor Site Criteria<sup>3</sup>

# 3. Terminology

- 3.1 Definitions:
- 3.1.1 For definitions of terms used in this standard, refer to Terminologies D1129 and D7902. For terms not defined in this test method or in Terminologies D1129 and D7902, refer to other published glossaries.<sup>4</sup>

#### 4. Summary of Practice

4.1 The average energy per disintegration,  $\bar{E}$  (pronounced E bar), for a mixture of radionuclides is calculated from the known composition of the mixture.  $\bar{E}$  is computed by calculating the total beta/gamma energy release rate, in MeV, and dividing it by the total disintegration rate. The resultant  $\bar{E}$  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).<sup>5</sup> The  $\bar{E}$  value is used to calculate a site-specific activity limit for the reactor coolant system, generally identified as:

$$A_{\text{limiting}} = K/\bar{E} \tag{1}$$

where:

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  $A_{\text{limiting}}$ . If the reactor coolant activity value is less than  $A_{\text{limiting}}$  then the 2-h radiation dose, measured at the plant boundary, will not exceed an appropriately small fraction of the Code of Federal

<sup>&</sup>lt;sup>1</sup> This practice is under the jurisdiction of ASTM Committee D19 on Water and is the direct responsibility of Subcommittee D19.04 on Methods of Radiochemical Analysis.

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<sup>&</sup>lt;sup>2</sup> 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.

<sup>&</sup>lt;sup>3</sup> Available from DLA Document Services, Building 4/D, 700 Robbins Ave., Philadelphia, PA 19111-5094, http://quicksearch.dla.mil.

<sup>&</sup>lt;sup>4</sup> "American National Standard Glossary of Terms," *Nuclear Science and Technology (ANSI N1.1)*, American National Standards Institute, 1430 Broadway, New York, NY 10018.

<sup>&</sup>lt;sup>5</sup> The boldface numbers in parentheses refer to a list of references at the end of this practice.

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  $\bar{E}$  and subsequently  $A_{\text{limiting}}$  are only those that are measured using gamma spectrometry.

- 5.2 In calculating  $\bar{E}$ , the energy dissipated by beta particles (negatrons and positrons) and photons from nuclear decay of beta-gamma emitters includes the energy released in the form of extra-nuclear transitions such as X-rays, Auger electrons, and conversion electrons. However, not all radionuclides present in a sample are included in the calculation of  $\bar{E}$ .
- 5.3 Individual nuclear reactor technical specifications vary and each nuclear operator must be aware of limitations affecting plant operation. Typically, iodine 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 95 % confidence level are not included in the calculation. However, technical requirements specify 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  $\mu$ Ci (37 kBq) per gram dose equivalent <sup>131</sup>I (DEI) in the reactor coolant.
- 5.3.2 Beta-only-emitting radio isotopes (for example,  $^{90}$ Sr or  $^{63}$ Ni) and alpha emitting radioisotopes (for example,  $^{241}$ Am or  $^{239}$ Pu) which comprise a small fraction of the activity, are not 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  $A_{\text{limiting}}$ , 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  $\bar{E}$ .
- 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, any time 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  $\bar{E}$  value and affect  $A_{\text{limiting}}$ . The two possible causes to reassess the value of  $\bar{E}$  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  $\bar{E}$  such as  $^{239}\text{U}$  and  $^{239}\text{Np}$ .

## 6. Interferences

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, <sup>56</sup>Mn (t½ = 2.6 h) has a gamma-ray energy of 847 keV and <sup>134</sup>I (t½ = 53 min) also has a gamma ray energy of 847 keV. The 847 keV gamma ray is also the most abundant of each of these radionuclides. It would be inaccurate to use the 847 keV gamma ray for the determination for 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 a minimum of 20 days of power operation have elapsed since the reactor was last 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.

- 9.2 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 d 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 d, or 30 d) 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  $\mu$ Ci/mL (37 kBq/mL) or  $\mu$ Ci/g (37 kBq/g), of all applicable gamma radioisotopes identified in the sample. Examples of the most significant contributing radioisotopes to  $\bar{E}$  are:

(1) Noble gas fission products: <sup>131m</sup>Xe, <sup>131</sup>Xe, <sup>133m</sup>Xe, <sup>133X</sup>E, <sup>133x</sup>Xe, <sup>130</sup>Xe, <sup>130</sup>

(2) Soluble fission products: <sup>137</sup>Cs, <sup>134</sup>Cs, <sup>141</sup>Ce, <sup>140</sup>Ba, <sup>140</sup>La, <sup>92</sup>Sr (others),

(3) Corrosion activation products: <sup>58</sup>Co, <sup>56</sup>Mn, <sup>54</sup>Mn, <sup>60</sup>Co, <sup>51</sup>Cr, <sup>59</sup>Fe, <sup>95</sup>Zr, <sup>95</sup>Nb (others),

(4) Miscellaneous radionuclides: <sup>41</sup>Ar, <sup>24</sup>Na, <sup>18</sup>F, <sup>7</sup>Be (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  $\overline{E}$ .

# 10. Calculation

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

$$\bar{E} = \frac{\sum_{i=1}^{n} (A_i * E_i)}{\sum_{i=1}^{n} A_i}$$
 (2)

where:

 $\bar{E}$  = average energy per disintegration, MeV/disintegration,

 $A_i$  = activity of the  $i^{th}$  radionuclide uniformly measured,  $\mu$ Ci/cc or  $\mu$ Ci/g, and

 $E_i$  = isotopic energy emission for the  $i^{th}$  radionuclide, MeV/disintegration.

10.2 The values for  $A_{\rm i}$  are the measured activity levels of a representative sample in  $\mu {\rm Ci/cc}$  (37 kBq/cc) or  $\mu {\rm Ci/g}$  (37 kBq/g), for each appropriate radionuclide identified in the sample (for example,  $^{60}{\rm Co}$ ,  $^{133}{\rm Xe}$ ,  $^{137}{\rm Cs}$ , 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:

$$E_i = E_i(\text{beta}) + E_i(CE) + E_i(A) + E_i(\text{gamma}) + E_i(X)$$
 (3)

where:

 $E_i(A)$ 

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

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

disintegration, = the average, abundance weighted, Auger elec-

tron energy per disintegration, MeV/disintegration,

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

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

10.4 An example for the calculation of  $E_i$  for the disintegration of  $^{133}\mathrm{Xe}~(E_{\mathrm{Xe-133}})$  follows.

10.4.1 The decay scheme for  $^{133}$ Xe (2) is given in Fig. 1. 10.4.2 First, calculate  $E_{\text{Xe-}133}$  (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 energy for each isotope may be found in the literature (2, 3) or, it may be approximated by multiplying the maximum beta particle energy per transformation by a factor of one-third. Only one-third of the maximum beta energy is included in the calculation because the remaining two-thirds of the radionuclide decay energy is dissipated by neutrino emission (4). Neutrinos are very high-energy, chargeless particles that do not undergo interaction with matter like the human body. Therefore, their contribution is ignored when considering the total energy available for absorption by a person at the site boundary of the nuclear facility.

10.4.2.2 The average energies and abundances of the major beta emissions for the decay of  $^{133}$ Xe are (2):

| beta # | Average Energy | Abundance |
|--------|----------------|-----------|
| 2      | 0.0751 MeV     | 0.69 %    |
| 3      | 0.101 MeV      | 99.3 %    |

10.4.2.3 Therefore,  $E_{\text{Xe-}133}$  (beta) is:

 $E_{Xe-133}$  (beta) = (beta #2 average energy) × (beta 2 abundance) + (beta #3 average energy) × (beta 3 abundance)

 $E_{\text{Xe-}133}$  (beta) = 0.0751 × 0.0069 + 0.101 × 0.993,  $E_{\text{Xe-}133}$  (beta) = 0.101 MeV/disintegration.

10.4.3 Next, calculate  $E_i(CE)$ .

10.4.3.1 Unlike beta-particle emissions, conversion electrons are monoenergetic emissions and are not accompanied by

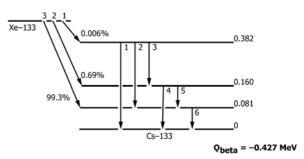


FIG. 1 Decay Scheme for <sup>133</sup>Xe