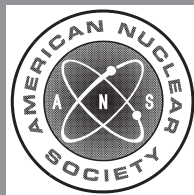


# American Nuclear Society

**method for calculating the  
fractional release of  
volatile fission products from oxide fuel**

**an American National Standard**



**published by the  
American Nuclear Society  
555 North Kensington Avenue  
La Grange Park, Illinois 60526 USA**

**American National Standard  
Method for Calculating the  
Fractional Release of  
Volatile Fission Products from Oxide Fuel**

Secretariat  
**American Nuclear Society**

Prepared by the  
**American Nuclear Society  
Standards Committee  
Working Group ANS-5.4**

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## Foreword

(This Foreword is not a part of American National Standard “Method for Calculating the Fractional Release of Volatile Fission Products from Oxide Fuel,” ANSI/ANS-5.4-2011.)

This standard provides a methodology for determining the radioactive fission product releases from  $\text{UO}_2$  fuel for use in assessing radiological consequences of postulated accidents that do not involve significant abrupt power transients such as in a reactivity-initiated accident (RIA). When used with isotopic yields, this method will give the so-called “gap activity,” which is the inventory of volatile fission products that could be available for release from the fuel rod if the cladding were breached. The standard as developed applies to steady-state conditions and, therefore, should not be applied to accidents where abrupt temperature increases are experienced resulting in what is sometimes referred to as “burst release.” The standard can be applied to Condition 1 and 2 transients that do not result in large fuel temperature increases, i.e.,  $\geq 300$  K temperature increase, where significant burst release is possible. The standard does not consider escape-rate coefficients or other transport of fission products from the fuel rod void space to the reactor coolant or other medium; the standard applies only to available fission products in the fuel rod void space available for release if failure is experienced. The standard assumes that no significant fuel oxidation will be present during the accident because fuel oxidation can significantly enhance the release. Therefore, the standard does not apply to accidents where significant fuel oxidation is present.

The volatile and gaseous fission products of primary significance are krypton, xenon, iodine, and cesium. The radioactive nuclide that contributes the largest to equivalent dose to individuals is generally  $^{131}\text{I}$  for accidents during in-reactor operation or shortly after reactor operation (e.g., a fuel-handling accident) because of its dose to the thyroid. These radioactive gaseous and volatile fission products can be divided into two categories: (a) short-lived radioactive nuclides (half-life  $< 1$  yr) and (b) long-lived radioactive nuclides (half-life  $> 1$  yr). This division is significant because the most important release mechanism involves thermally activated migration (diffusion) processes that proceed slowly such that the short-lived nuclides decay appreciably before they are released from the pellet. Consequently, release calculations for short-lived nuclides must include their decay rate, whereas for long-lived nuclides, decay does not have to be considered. The revised new standard is only applicable to the short-lived nuclides with half-lives  $< 1$  yr.

The ANS-5.4 standard was first implemented in 1982 with Dr. Stan Turner as chairman of the ANS-5.4 Working Group. The original methodology was based on the use of the Booth diffusion model [A. H. Booth, CRDC-721 and DCI-17, Atomic Energy of Canada (1957); A. H. Booth and G. T. Rymer, CRDC-720, Atomic Energy of Canada (1958); S. D. Beck, BMI-1433, Battelle Columbus (1960)]. The coefficients to the model were determined from the measured release data of stable nuclides of xenon and krypton. The coefficients were derived using the stable nuclide release data because very few release data were available for the radioactive nuclides. Also, because of the lack of data from the radioactive  $^{131}\text{I}$ , the diffusion of this nuclide was assumed to be a factor of 7 higher than for the xenon and krypton nuclides. The 1982 standard was not reaffirmed in 1992 because newer data were demonstrating that the assumptions used in the earlier standard were overly conservative. Many of the original working group members were not available to revise the standard at that time; therefore, a new ANS-5.4 Working Group was formed in 2000 to revise the standard. In the past this standard had primarily been used to estimate the dose to operators in the control room due to a fuel-handling accident but could be used

for any accidents where a large fuel temperature increase is not experienced and fuel failure has been determined or assumed.

In the last 25 yr, fuel experiments in test reactors have been performed that have measured the release fractions of the radioactive nuclides (including  $^{131}\text{I}$ ) up to 95 MWd/kg U (peak pellet average), allowing for a verification of the standard to higher burnups. These new data have demonstrated that the diffusion coefficients for the xenon, krypton, and iodine nuclides are essentially the same, such that the factor of 7 higher diffusion coefficient for iodine assumed in the 1982 standard was overly conservative. In addition, the use of the 1982 methodology also overpredicted the release fractions for the xenon and krypton stable nuclides when used in modern fuel performance codes that include a fuel thermal conductivity model that is a function of burnup. As a consequence, new empirical diffusion coefficients have been developed; however, the methodology still utilizes the simple phenomenological diffusion model that is often referred to as the Booth diffusion model with some modifications such as allowance for bubble interlinkage on grain boundaries that is temperature and burnup dependent. The interlinkage enhances the release with increasing burnup and temperature. The empirical coefficients have been developed from the radioactive xenon and krypton release data published in the last 20 yr. The new coefficients have been compared to the  $^{131}\text{I}$  release data and found to predict these release data well and, therefore, be applicable to radioactive iodine release that dominates most dose consequences for design-basis accidents.

The 1982 ANS-5.4 methodology and diffusion coefficients were considered to be conservative for application to  $\text{UO}_2$  fuel and bounding for light water reactor (LWR) applications. The revised coefficients for this revision to the standard are considered to provide more of a best-estimate prediction of release from  $\text{UO}_2$ ; a relatively small conservatism may exist but cannot be determined quantitatively because of the uncertainties in the current data. As a result the uncertainties of the predictions using the revised methodology are provided in the standard with recommendations on their application to determining radiological releases.

As noted earlier, the working group has chosen to retain the simple phenomenological Booth diffusion-type model proposed in the 1982 standard and has fitted the model coefficients empirically to selected data, whose characteristics will be described later. The “idealized” Booth model describes diffusion of fission product atoms in a spherical grain of fuel material. The governing differential equation is

$$\frac{\partial C}{\partial t} = B - \lambda C + \frac{D}{r} \frac{\partial^2 (rC)}{\partial r^2} , \quad (1)$$

where:

$C$  = nuclide concentration in the fuel ( $\text{atom} \cdot \text{cm}^{-3}$ );

$t$  = time (s);

$B$  = production rate ( $\text{atom} \cdot \text{cm}^{-3} \cdot \text{s}^{-1}$ );

$\lambda$  = decay constant ( $\text{s}^{-1}$ );

$r$  = radius at a point within the sphere (cm);

$D$  = diffusion coefficient ( $\text{cm}^2 \cdot \text{s}^{-1}$ ).

The latter term of Eq. (1) is the local mass flow of atoms at a radius  $r$  ( $\text{atom} \cdot \text{cm}^{-3} \cdot \text{s}^{-1}$ ).

The solution to Eq. (1) used in the original ANS-5.4 standard for the release fraction is sometimes referred to as  $R/B$ , the release-to-birth ratio, given by

$$R/B = 3 \left[ \frac{1}{\sqrt{\mu}} \coth(\sqrt{\mu}) - \frac{1}{\mu} \right], \quad (2)$$

where:

$R$  = number of atoms released per unit time (accounting for decay and temperature) at a point in time;  $R = -3(D/a)(\partial C/\partial r)_{r=a}$  (atom·cm<sup>-3</sup>·s<sup>-1</sup>);

$B$  = number of atoms produced per unit time at the same point in time;

$C$  = nuclide concentration in fuel;  $C = 0$  is assumed at the sphere surface, where  $r = a$ , with  $r$  = the radius at a point within the sphere and  $a$  = the idealized sphere radius;

$\mu = \lambda a^2/D$  (unitless).

Equation (2) is for equilibrium conditions; i.e., it assumes a period of constant power operation over several half-lives of the nuclide for the  $R/B$  in question (generally three half-lives is sufficient). If the temperature and power operation change within fewer than three half-lives, Eq. (2) provides a conservative prediction of  $R/B$  if the maximum power and temperature are used during the time period in question and the power change is not large enough to elicit large burst releases.

This diffusion equation assumes that a net flow of matter occurs because of the existence of a concentration gradient within the sphere and that the flux of atoms is proportional to that gradient. The production rate  $B$  and decay constant  $\lambda$  in Eq. (1) are known for the nuclides of interest, but the effective diffusion coefficients  $D$  and idealized sphere radius  $a$  in Eq. (2) are unknown and must be determined from experimental data.

The release fraction for the current standard uses the same equation but defines the radius of the sphere  $a$  in terms of a surface area-to-volume ratio  $S/V$ , which for a sphere is  $a = 3/(S/V)$ . Substituting  $3/(S/V)$  in place of coefficient  $a$  in Eq. (2) results in the following relationship:

$$R/B = S/V \frac{1}{\sqrt{\frac{\lambda}{\alpha D}}} \left[ \coth \sqrt{\mu} - \frac{1}{\mu} \right], \quad (3)$$

where:

$$\mu = \frac{9\lambda}{(S/V)^2 \alpha D}.$$

Note that  $\alpha$  is a unitless term for precursor effects that were generally ignored in the 1982 standard with the exception of the nuclides <sup>133</sup>Xe and <sup>135</sup>Xe. However, the current standard will include precursor effects for additional nuclides. Equation (3) can be further simplified with only a small overprediction (by  $<5 \times 10^{-3}$  relative) for release values  $<0.02$  with the following relationship:

$$R/B = S/V \sqrt{\frac{\alpha D}{\lambda}}. \quad (4)$$

The maximum  $R/B$  for  $^{131}\text{I}$  (the longest lived nuclide of interest) in a peak rod in today's LWRs is determined to be  $<0.02$  from a best-estimate prediction with the new standard. Also, the error introduced in Eq. (4) is several orders of magnitude less than the  $2\sigma$  uncertainty in the revised standard (between a factor of 2.5 to 5). Therefore, Eq. (4) provides a reasonably accurate prediction of release for nuclides considering the uncertainties in the analysis, and this is the form of the model recommended for determining the radiological release of the volatile nuclides.

The Booth diffusion model is an oversimplification of the physical processes involved in the release of fission products, and as such, it cannot correctly calculate the release for all fuel rod accident scenarios. One particular accident is the RIA that involves a very large increase in rod power over a very short time period ( $<10^{-1}$  s) where atom diffusion is negligible but large burst releases have been observed experimentally from in-reactor fuel tests that have simulated this accident [T. Fuketa et al., "NSRR/RIA Experiments with High Burnup PWR Fuels," *Proc. Int. Topl. Mtg. Light Water Fuel Performance*, Portland, Oregon, March 2–6, 1997; T. Fuketa et al., "NSRR RIA-Simulating Experiments on High Burnup LWR Fuels," *Proc. Water Reactor Fuel Performance Mtg.*, Kyoto, Japan, October 2–6, 2005, pp. 633–645; F. Lemoine et al., "The Role of Grain Boundary Fission Gases in High Burn-Up Fuel Under Reactivity Initiated Accident Conditions," *Proc. Seminar Fission Gas Behavior in Water Reactor Fuels*, Cadarache France, September 26–29, 2000; T. Nakamura et al., "Boiling Water Reactor Fuel Behavior Under Reactivity-Initiated-Accident Conditions at Burnup of 41 to 45 GWd/tonne U," *Nucl. Technol.*, **129**, 141 (2000); T. Nakamura et al., "High-Burnup BWR Fuel Behavior Under Simulated Reactivity-Initiated Accident Conditions," *Nucl. Technol.*, **138**, 246 (2002); L. Yegorova et al., "Experimental Study of Narrow Pulse Effects on the Behavior of High Burnup Fuel Rods with Zr-1%Nb Cladding and  $\text{UO}_2$  Fuel (VVER Type) Under Reactivity-Initiated Accident Conditions: Program Approach and Analysis of Results," NUREG/IA-0213, Vol. 1, U.S. Nuclear Regulatory Commission (May 2006)]. The hypothesized release mechanism for this release during a RIA is grain boundary fracturing where much of the fission gas is stored in high-burnup fuel. Therefore, the proposed model is not applicable to fuel accidents where very large sudden fuel temperature changes are experienced, i.e.,  $\geq 300$  K temperature increase, where significant burst release is possible. The proposed model can reasonably predict release from power changes that last several hours or days (typical of normal LWR operation), and the data used to develop and verify the model include these types of power changes. There may be small burst releases at high burnups for the milder temperature transients ( $<300$  K increase), but these will be covered by the conservatisms in the methodology such as assuming equilibrium conditions and the large uncertainty assigned to  $R/B$ .

This standard might reference documents and other standards that have been superseded or withdrawn at the time the standard is applied. A statement has been included in the references section that provides guidance on the use of references.

This standard does not incorporate the concepts of generating risk-informed insights, performance-based requirements, or a graded approach to quality assurance. The user is advised that one or more of these techniques could enhance the application of this standard.

The current working group acknowledges and appreciates the past efforts of the earlier working group that established the simple diffusional model that was also used for the revised standard with revisions to the coefficients.



Working Group ANS-5.4 of the American Nuclear Society (ANS) Standards Committee had the following membership at the time of the approval of the 1982 version of this standard:

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