ANSI/ANS-5.4-1982

American Nuclear Society

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

WITHDRAWN
March 8, 1993
ANSI/ANS-5.4-1982

an American National Standard

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published by the
American Nuclear Society
555 North Kensington Avenue
La Grange Park, Illinois 60525 USA

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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

Published by the
American Nuclear Society
555 North Kensington Avenue
La Grange Park, Illinois 60525 USA

Approved November 10, 1982
by the
American National Standards Institute, Inc.

American National Standard

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Published by

American Nuclear Society
555 North Kensington Avenue, La Grange Park, Illinois 60525 USA

Price: $12.00

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Printed in the United States of America

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-5.4-1982.)

Working Group ANS-5.4 was established in 1974 to examine fission product releases from UO_2 fuel. The scope of ANS-5.4 was defined to include only the following:

(1) Review available experimental data on release of volatile fission products from UO_2 and mixed-oxide fuel.

(2) Survey existing analytical models currently being applied to light-water reactors.

(3) Develop a standard analytical model for volatile fission product release to the fuel rod void space. Place emphasis on obtaining a model for radioactive fission product releases to be used in assessing radiological consequences of postulated accidents.

The standard as developed applies to steady-state conditions. 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. This gap inventory of radioactive fission products can be used in accident analyses, but should not be applied if abrupt temperature changes are involved. The standard does not consider escape-rate coefficients or other descriptions of the transport of fission products after release from the fuel rod void space.

The volatile and gaseous fission products of primary significance are krypton, xenon, iodine, cesium, and tellurium. These gaseous and volatile fission products can be divided into two categories: (1) short-lived radioactive nuclides (halflife less than one year) and (2) long-lived radioactive nuclides (halflife greater than one year) and stable species. This division is convenient since the most important release mechanism involves thermally activated migration 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 and stable nuclides, decay does not have to be considered.

Most experimental measurements of released fission gas were preceded by a cooldown period of approximately a year, during which time all of the short-lived radioactive species decayed. As a result, insufficient data exist to directly determine a release correlation for short-lived nuclides. While gas-release correlations based on stable-nuclide data are useful for fuel-performance calculations, they are usually not capable of predicting the radioactive releases. However, it is possible to derive an analytical model that is based on mechanistic or phenomenological principles that will predict releases as a function of halflife and can be calibrated with stable-gas-release data. This is the approach taken by ANS-5.4.

The working group has chosen what is believed to be the simplest such phenomenological model, the Booth diffusion-type model(1-5)*, and has fitted the model empirically to selected data, whose characteristics will be described later.

The Booth model describes diffusion of fission-product atoms in a sphere of fuel material. The governing equation is:

$$\partial C/\partial t = B - \lambda C - \mathrm{div}\, J \qquad \text{(Eq. 1)}$$

where C is the isotope concentration (atoms/cm^3), B is the production or birth rate (atoms/cm^3 sec), λ is the decay constant (sec^{-1}), and J is the local mass flux (atoms/cm^2 sec). The rate of concentration change in a region is equal to the rate of

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*Numbers in parentheses refer to the Bibliography attached to this Foreword.

production minus the rate of decay minus the rate of loss by mass flow out of the region. Equation 1 implies nothing about the mechanism of mass flow. The apparent diffusion coefficient D is contained in the flux term, which is given by:

$$J = -D \, \mathrm{grad}\, C \qquad \text{(Eq. 2)}$$

This diffusion equation assumes that a net flow of matter occurs because of the existence of a concentration gradient and that the flux is proportional to that gradient. The production rate B and decay constant λ in Equation 1 are known for most isotopes, but the diffusion coefficient D in Equation 2 is unknown and must be determined from experimental data.

From a general knowledge of atomic migration(6), it is known that the diffusion coefficient of a species in a host material depends on the properties of that material and its interaction with the diffusing species. These interactions are primarily electronic in nature so that different atoms (elements) would have different diffusion coefficients. Because the valence and ionic properties of krypton and xenon are similar, their diffusion coefficients in UO_2 are similar. However, there is no reason to expect the noble gases to behave like iodine or other chemical species. Therefore, it must be presumed that different elements migrate and are released at different rates.

On the other hand, the diffusion behavior of a chemical species can be expected to be the same for all nuclides of that species. While, strictly speaking, there is a diffusion isotope effect that is dependent on isotopic mass (7), this effect is very small and has only been detected in a few precise experiments using nuclides with large mass differences. Small differences in diffusion behavior would be imperceptible in the context of fission gas release.

The Booth diffusion model is an oversimplification of the physical process. The effective diffusion parameters that are determined by empirically fitting the Booth model to gas release data are not the diffusion coefficients for atomic diffusion of inert gases and other chemical species in pure UO_2. Atomic diffusion, gas bubble nucleation, bubble migration, bubble coalescence, interaction of bubbles with structures, and irradiation resolution are all involved in fission gas release. Some of these processes, like bubble migration, are relatively well understood. The microscopic parameters that govern these mechanisms are, in turn, dependent on the materials properties, such as diffusion coefficient, heats of vaporization, etc., which are independent of isotopic makeup. Therefore, it seems appropriate to assume that the overall release kinetics are the same for all nuclides of the same chemical species regardless of the complicated nature of the release mechanisms. In some cases, precursor effects cause a small apparent difference in release kinetics.

The Booth equations describe a smooth continuous release process and should not be applied to discontinuous releases or bursts (i.e., abrupt releases observed during sudden temperature changes). It is considered beyond the state of the art to model burst releases in a quantitative manner. Nevertheless, gases released in bursts are included in the data and are, therefore, accounted for in the cumulative releases predicted by the empirical model.

Finally, temperature-independent mechanisms are expected to be important for gas releases at low temperatures. As with the temperature-dependent (high temperature) diffusion-type model, the release fraction for radioactive species will depend on the nuclide halflife. However, the low-temperature release mechanisms are thought to be controlled by knock-out and recoil and, therefore, all chemical species are treated alike.

At the time this standard was developed, Working Group ANS-5.4 of the Standards Committee of the American Nuclear Society had the following membership:

S. E. Turner, Chairman, *Southern Science Applications, Inc.*
C. E. Beyer, *Battelle Pacific Northwest Laboratories*
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At the time of its approval of this standard, Subcommittee ANS-5, Fission Product Release, had the following members:

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B. J. Buescher, *EG&G Idaho, Inc.*
T. R. England, *Los Alamos National Laboratory*
R. J. Klotz, *Combustion Engineering, Inc.*
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S. E. Turner, *Southern Science Applications*

The American Nuclear Society's Nuclear Power Plant Standards Committee (NUPPSCO) had the following membership at the time of its approval of this standard.

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E. R. Wiot	*NUS Corporation*

Bibliography

(1) A. H. BOOTH, Atomic Energy of Canada Report, CRDC-721 (1957).

(2) A. H. BOOTH, Atomic Energy of Canada Report, DCI-17 (1957).

(3) A. H. BOOTH, and G. T. RYMER, Atomic Energy of Canada Report, CRDC-720 (1958).

(4) S. D. BECK, Battelle Columbus Report, BMI-1433 (1960).

(5) B. LUSTMAN, in J. BELLE (ed.), *Uranium Dioxide: Properties and Nuclear Applications,* p. 431. U.S. Atomic Energy Commission, Washington, D.C. (1961).

(6) P. G. SHEWMON, *Diffusion in Solids.* McGraw-Hill, New York, N.Y. (1963).

(7) N. L. PETERSON, *Solid State Physics,* **22**, 409 (1968).

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