

A model for release of fission products from a breached fuel plate under wet storage

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Abstract

MTR fuel elements burned-up inside the core of nuclear research reactors are stored worldwide mainly under the water of storage pools. When cladding breach is present in one or more fuel plates of such elements, radioactive fission products are released into the storage pool water. This work proposes a model to describe the release mechanism considering the diffusion of nuclides of a radioactive fission product either through a postulated small cylindrical breach or directly from a large circular hole in the cladding. In each case, an analytical expression is obtained for the activity released into the water as a function of the total storage time of a breached fuel plate. Regarding sipping tests already performed at the IEA-R1 research reactor on breached MTR fuel elements, the proposed model correlates successfully the specific activity of ^{137}Cs , measured as a function of time, with the evaluated size of the cladding breach.

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1. Introduction

Around the world, many research reactors use plate type, usually designated as Material Testing Reactor (MTR) fuel elements. A typical MTR fuel element has 18 plane parallel fuel plates, mounted mechanically between two lateral aluminum holders with grooves. Each fuel plate consists of an aluminum cladding and a meat, where the nuclear fuel is located (Terremoto et al., 2000).

Nowadays, most of the spent MTR fuel elements are stored under water in interim storage pools until reprocessed or stabilized for final disposition. However, due to legislation and other restrictions, many countries do not reprocess spent nuclear fuels. As a consequence, the long-term wet storage will remain for many years the prevalent destination of spent MTR fuel elements worldwide.

Under these circumstances, a very important phenomenon is the release of radioactive fission products into the storage pool water when cladding breach is present in one or more fuel plates of such stored elements.

In order to describe this phenomenon, the present work proposes a model that considers diffusion of nuclides (in the chemical form of atoms or ions) of a radioactive fission product through a postulated small cladding breach, which consists of a narrow cylindrical hole with known dimensions, directed perpendicularly to the cladding surface to reach the meat of the fuel plate, therefore exposing the irradiated nuclear fuel straightforwardly to the storage pool water.

By means of the model, an analytical expression is obtained for the fission product activity released into the water as a function of the storage time of the breached fuel plate.

As an extension of the model, it is also studied the case in which, instead of a small breach, there is a large circular hole in the cladding, emphasizing the quantitative difference of both situations concerning the release of radioactive fission products into the water.

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2. Hypothesis of the model

The model describes the release of a radioactive fission product during the wet storage of an irradiated fuel plate with a small cladding breach, based on the following fundamental hypothesis:

- (a) There is a narrow cylindrical hole with known dimensions, directed perpendicularly to the cladding surface to reach the meat of the fuel plate, therefore forming a breach.
- (b) At the beginning of the storage ($t = 0$), it is well known the total number of nuclides of a radioactive fission product contained inside the irradiated nuclear fuel.
- (c) Water penetrates inside the breach and comes directly in contact with the surface of the irradiated nuclear fuel.
- (d) Nuclides (in the chemical form of atoms or ions) of a radioactive fission product are released by means of diffusion and chemical reactions (mainly corrosion) from the surface of the irradiated nuclear fuel into the water that is inside the breach.
- (e) Once in water, the nuclides move by diffusion along the narrow cylindrical hole with a constant diffusion coefficient.
- (f) After diffusing along the whole narrow cylindrical hole, the nuclides leave the breach and are emitted into the storage pool water.
- (g) At the exit of the breach, the nuclides emitted into the storage pool water diffuse away instantaneously.
- (h) The emitted nuclides are not removed from the storage pool water.
- (i) In each instant of the storage period, the net number of nuclides in the water results from the difference between the number of nuclides emitted at the exit of the breach and the number of nuclides that undergone decay.
- (j) During the whole storage period, the water movement is negligible inside as well as outside the breach, so that convective effects can be ignored.

3. Model development

The diffusion of a radioactive fission product through a given medium is described by means of the following partial differential equation (Friskney and Speight, 1976):

$$D\nabla^2\rho - \lambda\rho + S = \frac{\partial\rho}{\partial t} \quad (1)$$

where ρ is the concentration of nuclides of the fission product (nuclides/cm³), D is the empirical diffusion coefficient of the fission product (cm²/s), λ is the radioactive decay constant of the fission product (s⁻¹) and S is the source term that describes the generation rate of the fission product (nuclides/cm³s). It is important to emphasize that the diffusion coefficient variation with temperature is fairly sharp (Lide, 1994).

The system to be studied consists of an irradiated, breached fuel plate under wet storage. The breach is a small cylindrical hole with radius r_0 and length a across the cladding of the fuel plate, reaching the meat of the plate perpendicularly. The length a of the breach is equal to the thickness of the aluminum cladding.

Under these circumstances, the diffusion of a radioactive fission product in the water inside the breach is described by means of the following partial differential equation:

$$D\nabla^2\rho - \lambda\rho = \frac{\partial\rho}{\partial t} \quad (2)$$

once the source term is not present in the hole, but in the meat of the fuel plate.

The radius r_0 of the breach is much smaller than the diffusion length $L = \sqrt{D/\lambda}$ of the radioactive fission product in water. This assumption, together with the angular symmetry of the problem, reduces the differential equation (2) only to the coordinate x along the hole axis (French, 1966), therefore:

$$D\frac{\partial^2\rho}{\partial x^2} - \lambda\rho = \frac{\partial\rho}{\partial t} \quad (3)$$

In this equation, the time variable t is accounted from the beginning of the wet storage.

The initial and boundary conditions regarding the concentration of the radioactive fission product in the water inside the breach, described by the function $\rho(x, t)$, are:

- $\rho(x, 0) = 0 \Rightarrow$ there is no fission product inside the breach in the beginning of the storage;
- $\rho(a, t) = 0 \Rightarrow$ infinite dilution of the fission product at the exit of the breach;
- $\rho(0, t) = fS(t) \Rightarrow$ source term located at the bottom of the breach, where the interface between irradiated nuclear fuel and water is located.

In the source term of the bottom boundary condition, $S(t)$ is a function that gives the average concentration of the radioactive fission product in the irradiated nuclear fuel and f is the fraction of this average concentration released into the water inside the breach.

The fraction f depends on several factors: type of fission product, physical chemistry characteristics of the system that consists of water directly in contact with the irradiated nuclear fuel (temperature and water pH, among others), chemical composition and microstructure of the irradiated nuclear fuel. In the scope of the model, the fraction f is conceived as an empirical parameter whose average value has to be measured under steady-state storage conditions, for each gamma-emitting fission product, irradiated research reactor nuclear fuel and physical chemistry characteristics usually found in storage pools. The 24 h immediately after the breach is formed will be studied separately during the measurements, because the scarce evidences show that corrosion processes are more intense in this initial period (Durazzo and Ramanathan, 1987). Additionally, it is considered that the storage only begins at $t = 0$ when steady-state conditions are completely established.

During storage, the function that gives the average concentration of the radioactive fission product in the irradiated nuclear fuel is:

$$S(t) = S_0 e^{-\lambda t} \quad (4)$$

where λ is the decay constant of the radioactive fission product (s^{-1}) and S_0 is the initial average concentration of the radioactive fission product in the irradiated nuclear fuel (nuclides/cm³), measured at the very beginning of the storage period ($t = 0$).

Regarding the resolution of the partial differential equation (3), once the bottom boundary condition is non-homogeneous, the searched solution must have the form (Stephenson, 1970):

$$\rho(x, t) = v(x, t) + w(x, t) \quad (5)$$

with the following initial and boundary conditions (Stephenson, 1970):

$$w(x, 0) = -v(x, 0)$$

$$w(0, t) = fS(t) - v(0, t)$$

$$w(a, t) = v(a, t) = 0$$

The choice of function $v(x, t)$ must be made in order to get only homogeneous boundary conditions, which enable the employ of a usual method to obtain the function $w(x, t)$. In this case, the suitable choice is (Stephenson, 1970):

$$v(x, t) = fS_0 e^{-\lambda t} \left[1 - \frac{x}{a} \right] \quad (6)$$

An equation for $w(x, t)$ is obtained replacing initially expression (5) in the partial differential equation (3), with the result:

$$D \frac{\partial^2 w}{\partial x^2} - \lambda w - \frac{\partial w}{\partial t} = - \left[D \frac{\partial^2 v}{\partial x^2} - \lambda v - \frac{\partial v}{\partial t} \right] \quad (7)$$

to afterward replace function (6) on the right-hand side of Eq. (7) and find:

$$D \frac{\partial^2 w}{\partial x^2} - \lambda w - \frac{\partial w}{\partial t} = 0 \quad (8)$$

The partial differential equation (8) is homogeneous and can be written as follows:

$$D \frac{\partial^2 w}{\partial x^2} - \lambda w = \frac{\partial w}{\partial t} \quad (9)$$

The partial differential equation (9) can be solved employing the variables' separation method (Butkov, 1968), thus writing the function $w(x, t)$ in the form:

$$w(x, t) = X(x)T(t) \quad (10)$$

and replacing it in Eq. (9) to obtain:

$$DT(t) \frac{\partial^2 X(x)}{\partial x^2} - \lambda X(x)T(t) = X(x) \frac{\partial T(t)}{\partial t} \quad (11)$$

Dividing Eq. (11) by $X(x)T(t)$ and rearranging the terms, one finds:

$$D \frac{1}{X(x)} \frac{d^2 X(x)}{dx^2} - \lambda = \frac{1}{T(t)} \frac{dT(t)}{dt} \quad (12)$$

Both sides of expression (12) are non-constant functions of independent variables, which can be equalized to a constant:

$$D \frac{1}{X(x)} \frac{d^2 X(x)}{dx^2} - \lambda = -k^2$$

$$\frac{1}{T(t)} \frac{dT(t)}{dt} = -k^2 \quad (13)$$

in order to obtain two ordinary differential equations:

$$\frac{d^2 X(x)}{dx^2} + \left(\frac{k^2 - \lambda}{D} \right) X(x) = 0$$

$$\frac{dT(t)}{dt} + k^2 T(t) = 0 \quad (14)$$

The equation that involves the variable t has the solution:

$$T(t) = T_0 e^{-k^2 t} \quad (15)$$

whereas the equation that involves the variable x has the solution:

$$X(x) = A \cos \sqrt{\frac{k^2 - \lambda}{D}} x + B \sin \sqrt{\frac{k^2 - \lambda}{D}} x \quad (16)$$

and therefore the function $w(x, t)$ assumes the following general form:

$$w(x, t) = \left[A \cos \sqrt{\frac{k^2 - \lambda}{D}} x + B \sin \sqrt{\frac{k^2 - \lambda}{D}} x \right] e^{-k^2 t} \quad (17)$$

where A and B are arbitrary constants whose values must be determined applying the boundary conditions for the function $w(x, t)$. Consequently, one obtains:

$$w(0, t) = fS(t) - v(0, t) = 0 \Rightarrow A = 0$$

$$w(a, t) = 0 \Rightarrow B \sin \sqrt{\frac{k^2 - \lambda}{D}} a = 0 \Rightarrow \sqrt{\frac{k^2 - \lambda}{D}} = \frac{n\pi}{a} \Rightarrow k^2 = \frac{Dn^2\pi^2}{a^2} + \lambda$$

where n is an integer number ≥ 1 . As a result, the general form (17) becomes:

$$w(x, t) = \sum_{n=1}^{\infty} B_n \sin \frac{n\pi}{a} x e^{-\frac{Dn^2\pi^2}{a^2} t} e^{-\lambda t} \quad (18)$$

which is the general solution of the partial differential equation (9).

In order to determine the coefficients B_n , it is necessary to apply the initial condition:

$$w(x, 0) = -v(x, 0) \Rightarrow w(x, 0) = fS_0 \left[\frac{x}{a} - 1 \right] \quad (19)$$

with the result:

$$fS_0 \left[\frac{x}{a} - 1 \right] = \sum_{n=1}^{\infty} B_n \sin \frac{n\pi}{a} x \quad (20)$$

so that the following expansion according to the Fourier method (Stephenson, 1970) can be performed:

$$B_n = \frac{2}{a} \int_0^a fS_0 \left[\frac{x}{a} - 1 \right] \sin \frac{n\pi}{a} x \, dx \quad (21)$$

with the definite integral:

$$B_n = \frac{2fS_0}{a} \int_0^a \left[\frac{x}{a} - 1 \right] \sin \frac{n\pi}{a} x \, dx \quad (22)$$

solved by parts to give the result:

$$B_n = \frac{2fS_0}{a} \frac{(-a)}{n\pi} \Rightarrow B_n = -\frac{2fS_0}{n\pi} \quad (23)$$

and therefore the function $w(x, t)$ is:

$$w(x, t) = -\frac{2fS_0}{\pi} e^{-\lambda t} \sum_{n=1}^{\infty} \frac{1}{n} \sin \frac{n\pi}{a} x e^{-\frac{Dn^2\pi^2}{a^2}t} \quad (24)$$

The solution for $\rho(x, t)$ that satisfies the partial differential equation (3), with the initial and boundary conditions postulated for the system, is obtained, as shown in Eq. (5), by the sum of functions $v(x, t)$ and $w(x, t)$, which are given, respectively, by expressions (6) and (24). Rearranging the sum terms, one obtains the result:

$$\rho(x, t) = fS_0 e^{-\lambda t} \left[1 - \frac{x}{a} - \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{1}{n} \sin \frac{n\pi}{a} x e^{-\frac{Dn^2\pi^2}{a^2}t} \right] \quad (25)$$

The function $\rho(x, t)$ describes the concentration of the radioactive fission product, expressed in nuclides/cm³, inside the small, narrow cylindrical breach. Once the function $\rho(x, t)$ has been determined, it is possible to calculate two important quantities: the average concentration of the radioactive fission product inside the breach and the emission rate of this fission product into the storage pool water.

The average concentration $\bar{\rho}(t)$ of the radioactive fission product inside the breach, expressed in nuclides/cm³, is:

$$\bar{\rho}(t) = \frac{\int_v \rho(x, t) \, dv}{v} \quad (26)$$

where v is the total volume of the small cylindrical hole that forms the breach, therefore:

$$\bar{\rho}(t) = \frac{\int_0^{r_0} \int_0^{2\pi} \int_0^a \rho(x, t) r \, dr \, d\theta \, dx}{\pi r_0^2 a} \quad (27)$$

As a consequence of the angular and radial symmetries of the postulated breach, expression (27) becomes:

$$\bar{\rho}(t) = \frac{\int_0^a \rho(x, t) \, dx}{a} \quad (28)$$

with the result:

$$\bar{\rho}(t) = fS_0 e^{-\lambda t} \left[\frac{1}{2} - \frac{4}{\pi^2} \sum_{n=1,3,5,\dots} \frac{1}{n^2} e^{-\frac{Dn^2\pi^2}{a^2}t} \right] \quad (29)$$

The ratio between the average concentrations of the radioactive fission product inside the breach and inside the irradiated nuclear fuel is, therefore:

$$\frac{\bar{\rho}(t)}{S(t)} = f \left[\frac{1}{2} - \frac{4}{\pi^2} \sum_{n=1,3,5,\dots} \frac{1}{n^2} e^{-\frac{Dn^2\pi^2}{a^2}t} \right] \quad (30)$$

once $S(t) = S_0 e^{-\lambda t}$, as expression (4) shows.

The emission rate $E(t)$ of the radioactive fission product, expressed in nuclides/s, is proportional to the cross-sectional area of the breach, to the empirical diffusion coefficient of the radioactive fission product in water and to the concentration gradient of the radioactive fission product at the exit of the breach. Mathematically, this means (French, 1966):

$$E(t) = -\pi r_0^2 D \left(\frac{\partial \rho}{\partial x} \right)_{x=a} \quad (31)$$

with the result:

$$E(t) = \frac{\pi r_0^2 D f S_0 e^{-\lambda t}}{a} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-\frac{Dn^2\pi^2}{a^2}t} \right] \quad (32)$$

Finally, it is possible to determinate the total number N of nuclides of the radioactive fission product that will be present in the storage pool water, after a storage period with duration equal t and considering that these nuclides are not removed from the water.

In each instant of the storage period, the net number of nuclides in the water results from the difference between the number of nuclides emitted at the exit of the breach and the number of nuclides that undergone decay. Mathematically, this balance is described by the following ordinary differential equation (Litz, 1954; Lewis et al., 1990):

$$\frac{dN}{dt} = E(t) - \lambda N \quad (33)$$

that can be written in the form:

$$\frac{dN}{dt} + \lambda N = E(t) \quad (34)$$

where $E(t)$ is given by expression (32).

The general solution of the ordinary differential equation (34) is obtained by means of the formula (Machado, 1999):

$$N(t) = e^{-\lambda t} \left[\int e^{\lambda t} E(t) dt + C \right] \tag{35}$$

where C is a constant that depends on the initial conditions.

Replacing expression (32) for $E(t)$ in formula (35), one obtains the result:

$$N(t) = \frac{\pi r_0^2 D f S_0}{a} \left[t e^{-\lambda t} - \frac{2a^2 e^{-\lambda t}}{D \pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} e^{-\frac{D n^2 \pi^2 t}{a^2}} + C e^{-\lambda t} \right] \tag{36}$$

In order to determinate the value of the constant C , the initial condition $N(0) = 0$ is employed, which means that, in the beginning ($t = 0$) of the storage period, there were no nuclides of the radioactive fission product in the storage pool water. The obtained value:

$$C = \frac{2a^2 e^{-\lambda t}}{D \pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \tag{37}$$

is replaced in expression (36) to give the following result:

$$N(t) = \frac{\pi r_0^2 D f S_0}{a} \left[t e^{-\lambda t} + \frac{2a^2 e^{-\lambda t}}{D \pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \left(1 - e^{-\frac{D n^2 \pi^2 t}{a^2}} \right) \right] \tag{38}$$

Using the result (Jolley, 1961):

$$\sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} = -\frac{\pi^2}{12} \tag{39}$$

and rearranging the terms in a convenient way, one obtains:

$$N(t) = \frac{\pi r_0^2 D f S_0 e^{-\lambda t}}{a} \left\{ t - \frac{a^2}{D} \left[\frac{1}{6} + \frac{2}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} e^{-\frac{D n^2 \pi^2 t}{a^2}} \right] \right\} \tag{40}$$

and the activity in the storage pool water is $A(t) = \lambda N(t)$.

A summary of the differential equations with the corresponding initial and boundary conditions employed in the model is presented in Fig. 1, illustrating their validity region in the system.

When the release of a long-lived radioactive fission product is analyzed, many practical situations involve time intervals that are much shorter than the fission product half-life ($t \ll T_{1/2}$). Under these circumstances, it is possible to consider $e^{-\lambda t} \cong 1$ in Eq. (40) and outline the graph (Crank, 1975) of the obtained function, which in this case describes the total number $N(t)$ of nuclides of the radioactive fission product in the storage pool water as a function of the storage time t , with the result shown in Fig. 2.

The general behavior of the function exhibits two markedly distinct regions. Whereas the first region is transient, the second one is of steady-state diffusion characterized by a linear increase.

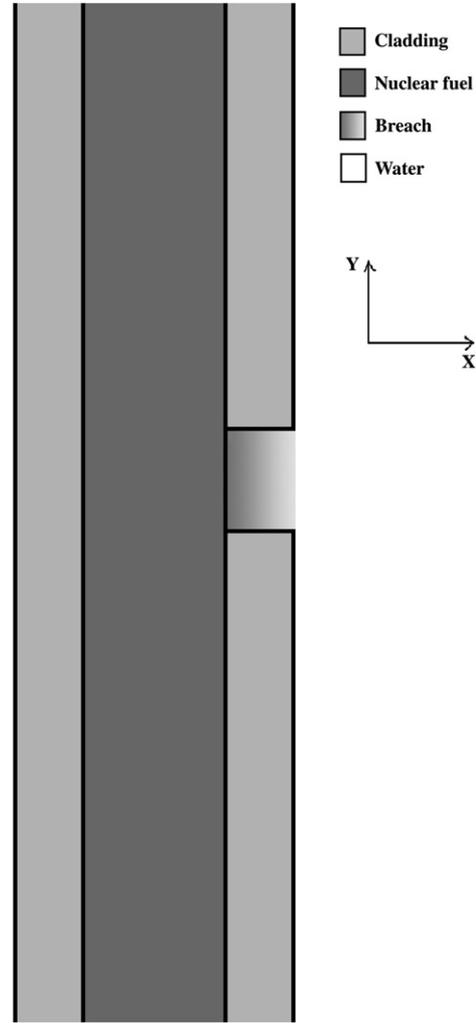


Fig. 1. Cross-sectional diagram of an irradiated, breached fuel plate, enlarged 25 times in order to illustrate the region of validity for the differential equations corresponding to the hypothesis of the model:

- Nuclear fuel: $dS/dt = -\lambda S$ with initial condition $S(0) = S_0$.
- Breach: $D(\partial^2 \rho / \partial x^2) - \lambda \rho = \partial \rho / \partial t$ with initial condition $\rho(x, 0) = 0$ and boundary conditions $\rho(a, t) = 0$; $\rho(0, t) = fS(t)$.
- Water: $dN/dt + \lambda N = -\pi r_0^2 D (\partial \rho / \partial x)_{x=a}$ with initial condition $N(0) = 0$.

4. Large circular hole in the cladding

The existence of a large circular hole in the cladding of a fuel plate is defined when the breach, although with the same depth a as the cladding thickness, has a radius R_0 that:

$$R_0 \gg a$$

$$R_0 \geq 0.01 \sqrt{D_0 / \lambda}$$

where D_0 is the diffusion coefficient for the infinite dilution of the fission product in water (cm^2/s), λ is the radioactive decay constant of the fission product (s^{-1}) and $L_0 = \sqrt{D_0 / \lambda}$ is the diffusion length of the fission product in water (cm).

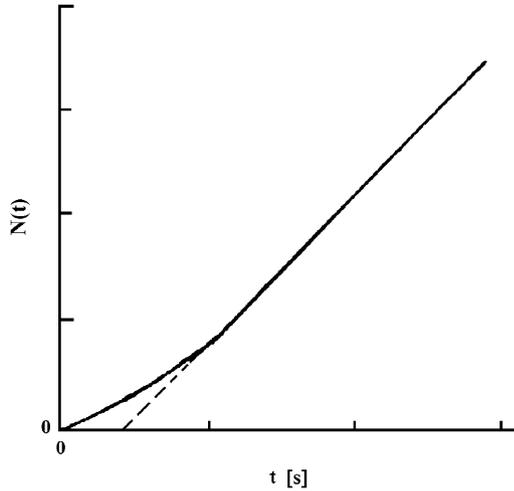


Fig. 2. Total number of nuclides of a radioactive fission product in the storage pool water as a function of the storage time of an irradiated fuel plate, considering a small cylindrical breach in the cladding and time intervals much shorter than the fission product half-life.

If there is a large hole in the cladding, the hypothesis regarding diffusion through a small breach, described in Section 2 of the present work, lose their validity, because in this case nuclides of the fission product are released so quickly to the water that their concentration inside the hole remains permanently equal to zero, in such way that the releasing mechanism can be attributed only to diffusion (Rae, 1985). Additionally, it is not valid the approximation that reduces the partial differential equation for diffusion inside the breach only to the coordinate along its axis, because in this case the radial coordinate must be also considered (Rae, 1985).

Under these circumstances, the emission rate $E(t)$, expressed in nuclides/s, assumes the form given by (Kim et al., 2002):

$$E(t) = 4 \frac{D_i D_0}{D_i + D_0} S_0 e^{-\lambda t} R_0 \left[1 + 2R_0 \left(\frac{1}{\pi D_0 t} \right)^{1/2} \right] \quad (41)$$

where D_i is the diffusion coefficient of the fission product inside the irradiated nuclear fuel (cm^2/s), D_0 is the diffusion coefficient for the infinite dilution of the fission product in water (cm^2/s), λ is the radioactive decay constant of the fission product (s^{-1}), R_0 is the large hole radius (cm) and S_0 is the initial average concentration of nuclides of the fission product inside the irradiated nuclear fuel (nuclides/ cm^3), measured in the beginning of the storage period. It is important to emphasize that a retardation factor, included in the original expression of the emission rate (Kim et al., 2002), is absent in Eq. (41) because there is no porous medium surrounding the large hole in the cladding, so that the nuclides of the fission product are supposed to diffuse from the meat of the irradiated fuel plate to the water of the storage pool.

Based on Eq. (41), it is possible to determinate the total number N of nuclides of the radioactive fission product that will be present in the storage pool water, after a storage period

with duration equal t and considering that these nuclides are not removed from the water.

In each instant of the storage period, the net number of nuclides in the water results from the difference between the number of nuclides emitted from the large circular hole and the number of nuclides that undergone decay. As previously mentioned, this balance is mathematically described by the ordinary differential equation (34) whose general solution is obtained by means of formula (35).

Replacing $E(t)$ by expression (41) in formula (35), one obtains the result:

$$N(t) = 4 \frac{D_i D_0}{D_i + D_0} S_0 R_0 e^{-\lambda t} \left[t + 4R_0 \left(\frac{t}{\pi D_0} \right)^{1/2} \right] + C e^{-\lambda t} \quad (42)$$

In order to determinate the value of the constant C , the initial condition $N(0) = 0$ is employed, which means that, in the beginning ($t=0$) of the storage period, there were no nuclides of the radioactive fission product in the storage pool water. In this case, the obtained result is $C = 0$, therefore:

$$N(t) = 4 \frac{D_i D_0}{D_i + D_0} S_0 R_0 e^{-\lambda t} \left[t + 4R_0 \left(\frac{t}{\pi D_0} \right)^{1/2} \right] \quad (43)$$

and the activity in the storage pool water is $A(t) = \lambda N(t)$.

When the release of a long-lived radioactive fission product is analyzed, many practical situations involve time intervals that are much shorter than the fission product half-life ($t \ll T_{1/2}$). Under these circumstances, it is possible to consider $e^{-\lambda t} \cong 1$ in Eq. (43) and outline the graph of the obtained function, which in this case describes the total number $N(t)$ of nuclides of the radioactive fission product in the storage pool water as a function of the storage time t , with the result shown in Fig. 3.

A comparison between the graphs outlined in Fig. 2 and in Fig. 3 shows quite different behaviors regarding the release of nuclides of a fission product into the storage pool water. The most important difference is that, if a large hole is present in the cladding of the fuel plate, no linear region is observed in the graph. A similar correlation has been obtained from measurements on the migration of radionuclides through samples of various sedimentary rocks (Meier et al., 1988).

5. Application of the model to the sipping tests

Sipping test is a non-destructive technique employed to evaluate the structural integrity of the cladding of irradiated nuclear fuels, which is based on the detection of radioactive fission products leakage to the water, usually by means of gamma-ray spectroscopy (Perrotta et al., 1998; Terremoto et al., 2002; Zeituni et al., 2004).

During the last 10 years, sipping tests had to be performed at the IEA-R1 research reactor only three times: one embracing irradiated fuel elements stored for many years inside the spent fuel pool and two regarding fuel elements in use inside the reactor core. Leakage of radioactive fission products has been confirmed in two of these occasions.

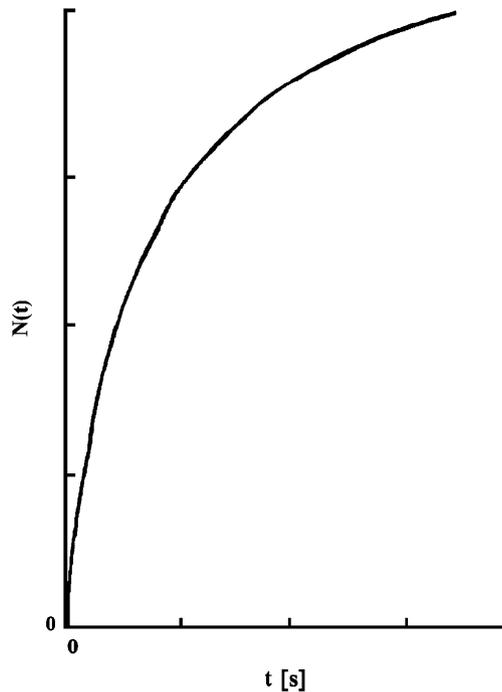


Fig. 3. Total number of nuclides of a radioactive fission product in the storage pool water as a function of the storage time of an irradiated fuel plate, considering a large circular hole in the cladding and time intervals much shorter than the fission product half-life.

Along this Section of the work, experimental data obtained from sipping tests performed in these two occasions are presented concisely and analyzed directly by means of the proposed model.

5.1. Fuel element IEA-53

Manufactured in the 1950s by Babcock & Wilcox Co. (USA), the MTR fuel element IEA-53 had 19 slightly curved fuel plates with aluminum cladding. Its nuclear fuel was a U–Al alloy with density 1.8 gU/cm^3 and uranium enrichment in ^{235}U equal to 20%.

The fuel element IEA-53 attained a burnup of 16.7% prior to removal from the IEA-R1 research reactor and thereafter remained continuously under wet storage during almost 34 years in a stainless steel rack inside the spent fuel pool.

Galvanic corrosion occurred as a consequence of the long storage period under water inside a stainless steel rack (Perrotta et al., 1998; Kain et al., 2000), originating many small pits in the aluminum cladding of the two most external plates of the fuel element IEA-53, which were easily observed during visual inspections (Perrotta et al., 1998).

These pits were removed, collected and sent to the laboratory, where gamma-ray spectroscopy measurements revealed the presence of fission products, showing that the corrosion had perforated the whole thickness $a = 0.038 \text{ cm}$ of the aluminum cladding and reached the meat of the fuel plates. Such evidence favored the decision to perform sipping tests on fuel element IEA-53 and other spent fuel elements that were under wet storage (Perrotta et al., 1998).

Concerning sipping tests on irradiated fuel elements stored for many years inside spent fuel pools, the most suitable gamma-emitting fission product for use as leakage monitor is ^{137}Cs , due to its long half-life ($T_{1/2} = 30.14$ years), large fission yields and high solubility in water.

The fuel element IEA-53 exhibited the highest average leaking rate of ^{137}Cs among all the 60 spent fuel elements submitted to sipping tests (Perrotta et al., 1998). In the date the tests were performed, the total ^{137}Cs activity of the fuel element IEA-53 was equal to $1.406 \times 10^{12} \text{ Bq}$ (Perrotta et al., 1998) distributed in 440.33 cm^3 of irradiated nuclear fuel. Therefore, regarding ^{137}Cs in this fuel element, $S_0 = 4.379 \times 10^{18} \text{ nuclides/cm}^3$.

During a sipping test, the time intervals involved (Perrotta et al., 1998; Terremoto et al., 2002; Zeituni et al., 2004) are such that:

$$\frac{a^2}{D\pi^2} \ll t \ll T_{1/2} \quad (44)$$

and consequently:

$$e^{-\lambda t} \cong 1, \quad e^{-\frac{D\pi^2 t}{a^2}} \cong 0$$

so that Eq. (40) becomes:

$$N(t) = \frac{\pi r_0^2 D f S_0}{a} \left\{ t - \frac{a^2}{6D} \right\} \quad (45)$$

with the specific activity of the collected water sample given by:

$$A(t) = -\frac{\pi r_0^2 a f \lambda S_0}{6\text{Vol}} + \frac{\pi r_0^2 D f \lambda S_0}{a\text{Vol}} t \quad (46)$$

where $\text{Vol} = 28 \text{ l}$ is the total volume of water contained inside the aluminum sipping tube used in the test.

This expression for $A(t)$, deduced from the model, reproduces the general form of the function fitted to the experimental data obtained in the sipping test of the breached fuel element IEA-53, which presented a linear increase of ^{137}Cs specific activity with sipping time (Perrotta et al., 1998) as shown in Fig. 4.

Qualitatively, this behavior concerning the release of radioactive fission products into the water is compatible with steady-state diffusion in case of a small breach in the cladding of one or more fuel plates.

Also according to the model, the empirical diffusion coefficient D of ^{137}Cs in water can be obtained by means of the ratio of the angular coefficient \mathbf{B} and the linear coefficient \mathbf{A} of the straight line fitted to the experimental data, with the result:

$$\frac{6D}{a^2} = \frac{\mathbf{B}}{60\mathbf{A}} \Rightarrow D = (6 \pm 1) \times 10^{-8} \text{ cm}^2/\text{s}$$

This value is much lower than the diffusion coefficient D_0 for the infinite dilution of ^{137}Cs in water, which measured (Lide, 1994; Sato et al., 1996) at the temperature of 25°C yields

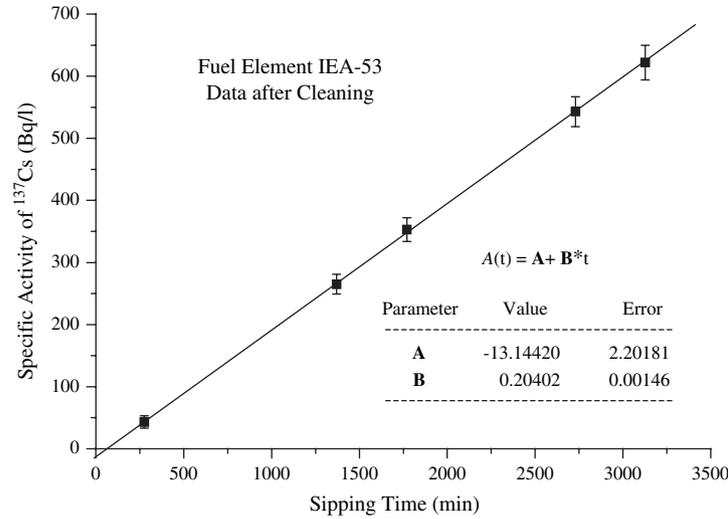


Fig. 4. Specific activity of ¹³⁷Cs measured as a function of sipping time for the breached fuel element IEA-53, presenting a linear increase (Perrotta et al., 1998).

$D_0 = 2.056 \times 10^{-5} \text{ cm}^2/\text{s}$. This difference can be explained as follows.

At a given temperature, the diffusion coefficient of a particle in a liquid medium is inversely proportional to the particle radius (Jost, 1960). Once the radius of the ion Cs^+ is $r_0 = 1.67 \times 10^{-8} \text{ cm}$ (Ruben, 1985), it is, therefore, possible to evaluate the average radius r of the particles that diffused in water during the sipping test carrying the ions Cs^+ :

$$r = \frac{D_0 r_0}{D} \Rightarrow r = (5.5 \pm 0.9) \times 10^{-6} \text{ cm}$$

Solid particles with such dimensions in aqueous medium can be either a colloid or a suspension. This result is in agreement with the observation that, in nuclear fuels of UAl_x dispersed in aluminum, aqueous corrosion products assume the form of colloidal gel (Kaminski and Goldberg, 2002; Kaminski, 2003; Kaminski et al., 2005). It also indicates that, in this case, aqueous corrosion is the predominant mechanism whereby radioactive fission products from the irradiated nuclear fuel are released into the water.

Before the sipping test, a pit was completely removed from one of the external fuel plates of the fuel element IEA-53 and sent to the laboratory in order to measure its mass, volume and specific activity of fission products. These analyses demonstrated that the pit material was impregnated with ¹³⁷Cs (Perrotta et al., 1998).

Under these circumstances, it is possible to evaluate the fraction f of the ¹³⁷Cs average concentration in the irradiated nuclear fuel that is released into the water inside the breach. Measuring the ¹³⁷Cs specific activity A_p of the pit material (Bq/cm^3), determining the ¹³⁷Cs specific activity λS_0 of the irradiated nuclear fuel (Bq/cm^3) and calculating the ratio of both, the result is, according to the model, expressed by Eq. (30) considering time intervals defined in (44), which gives:

$$\frac{f}{2} = \frac{A_p}{\lambda S_0} \Rightarrow f = 2 \frac{1.9 \times 10^4}{3.2 \times 10^9} \Rightarrow f = 1.2 \times 10^{-5}$$

Finally, once all parameters of Eq. (46) are known, the total area of the breach in the cladding of fuel element IEA-53 can be evaluated by means of the angular coefficient **B** of the straight line fitted to the experimental data, with the result:

$$\frac{\text{Area} D f \lambda S_0}{a \text{Vol}} = \frac{\mathbf{B}}{60} \Rightarrow \text{Area} = (1.5 \pm 0.2) \text{ cm}^2$$

This result is half of the estimated value of approximately 3 cm^2 for the largest breach extension in the cladding of a spent MTR fuel element that can be received by the Savannah River National Laboratory (SRNL, USA) regarding the Foreign Research Reactor Spent Nuclear Fuel Acceptance Program (Sindelar and Howell, 1997; Vinson et al., 2002).

The fuel element IEA-53 has been accepted for deposition and, together with other 126 spent MTR fuel elements, transported from IPEN/CNEN-SP to SRNL in 1999.

5.2. Fuel element IEA-156

On July 30th 2001, during a routine operation of the IEA-R1 research reactor at 2 MW, the presence of gaseous radioactive fission products was detected in the hall of the reactor pool. Detailed analysis performed shortly after the incident showed that these fission products were released from one of the 24 MTR fuel elements in use inside the reactor core. Sipping tests were then carried out in order to identify the breached fuel element and measure its leaking rate (Terremoto et al., 2002; Zeituni et al., 2004).

The fuel element IEA-156 was identified as breached. This fuel element, manufactured at IPEN/CNEN-SP, had 18 plane parallel fuel plates with aluminum cladding. Its nuclear fuel was U_3O_8 dispersed in aluminum with a density of $2.3 \text{ gU}/\text{cm}^3$ and uranium enrichment in ²³⁵U equal to 19.88%. This fuel element was in use inside the reactor core since September 8th 1997 and attained a burnup of 21.95%.

In the date the sipping tests were performed, the total ¹³⁷Cs activity of the fuel element IEA-156 was equal to $5.350 \times 10^{12} \text{ Bq}$

(Terremoto et al., 2002; Zeituni et al., 2004) distributed in 487.10 cm^3 of irradiated nuclear fuel. Therefore, regarding ^{137}Cs in this fuel element, $S_0 = 1.506 \times 10^{19}$ nuclides/ cm^3 . The total volume of water contained inside the aluminum sipping tube used in the test was $\text{Vol} = 28 \text{ l}$.

Detailed visual inspections, performed with a radiation-resistant underwater camera, were unable to provide any information concerning the location and extension of the breach in the cladding, because it was situated in internal plates of the fuel element (Terremoto et al., 2002; Zeituni et al., 2004).

The specific activity of ^{137}Cs measured as a function of sipping time (Terremoto et al., 2002; Zeituni et al., 2004) for the breached fuel element IEA-156 is shown in Fig. 5.

Although only two measurements were carried out during the sipping test, a sharp difference is observed when compared to the previous case, concerning not only the activity level reached but also the general distribution of the data.

Using expression (43) and the experimental data (Terremoto et al., 2002; Zeituni et al., 2004) of the specific activity of ^{137}Cs measured as function of sipping time, one obtains a system of two algebraic equations with the parameters R_0 and D_i as unknowns. The resolution of the system gives the following values:

$$R_0 = -(3.7 \pm 0.1) \text{ cm}, \quad R_0 = (3.2 \pm 0.1) \text{ cm},$$

$$D_i = (1.24 \pm 0.03) \times 10^{-12} \text{ cm}^2/\text{s}$$

Mathematically, there are two possible values for R_0 because the equations are quadratic in this parameter. However, only the positive value has physical meaning.

The value $R_0 = (3.2 \pm 0.1) \text{ cm}$ is compatible with the existence of a large hole in the cladding of one or more fuel plates of the fuel element IEA-156, whereas the value

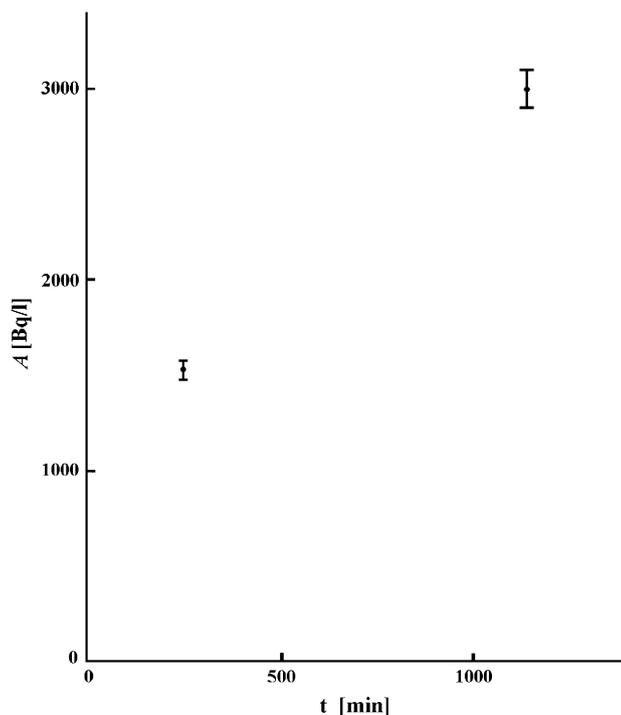


Fig. 5. Specific activity of ^{137}Cs measured as a function of sipping time for the breached fuel element IEA-156 (Terremoto et al., 2002; Zeituni et al., 2004).

$D_i = (1.24 \pm 0.03) \times 10^{-12} \text{ cm}^2/\text{s}$ indicates an expected low mobility of ^{137}Cs inside the irradiated nuclear fuel at room temperature ($25 \text{ }^\circ\text{C}$). Though the hole is probably not circular in shape at all, the value of R_0 obtained by means of the model provides an indication of the breach extent in the cladding.

6. Discussion

The proposed model may be used to evaluate the breach extent in the cladding of irradiated MTR fuel elements as well as the activity released into the storage pool water where these elements are deposited.

However, it is necessary first to measure very accurately the parameters f and D_i at usual wet storage temperatures (around $25 \text{ }^\circ\text{C}$), because there is a serious lack of experimental data regarding this subject.

Measurement of these parameters can be carried out using irradiated fuel miniplates in controlled experiments where:

- holes of precise sizes are drilled in the cladding of miniplates whose dimensions, burnup, microstructure and fission product inventory are well known;
- each miniplate contains in the meat a different kind of nuclear fuel usually employed in MTR fuel elements (U–Al alloy, UAl_x dispersed in aluminum, U_3O_8 dispersed in aluminum, U_3Si_2 dispersed in aluminum, U–Mo dispersed in aluminum);
- each miniplate is vertically immersed in a recipient containing deionized water;
- temperature and pH of the recipient water are monitored continuously;
- each miniplate remains immersed during 24 h in order to simulate the period immediately after the breach is formed, during which hydrogen evolution has been observed as a consequence of corrosion in meats containing unirradiated U_3O_8 dispersed in aluminum (Durazzo and Ramanathan, 1987);
- after these 24 h, the activity of fission products in the water of each recipient is measured by means of gamma-ray spectroscopy;
- each miniplate is withdrawn from the recipient and immersed immediately inside a new recipient containing a known volume of deionized water, there remaining in a steady vertical position in order to simulate the storage condition;
- water of each new recipient is very gently stirred in order to permanently homogenize the solution, whereas its temperature and pH are continuously monitored;
- small water samples are collected from each new recipient in regular time intervals and submitted to gamma-ray spectroscopy measurements;
- graphs of the specific activity of gamma-emitting fission products are plotted as a function of immersion time for each miniplate.

If the parameters f and D_i are well known, the main advantage in using the proposed model is to avoid the employ of destructive methods (dismantling of irradiated, breached fuel

elements inside pool water or in hot cells) or sophisticated techniques and equipments (non-destructive inspections using either eddy current or radiation-resistant industrial endoscopes) to evaluate the size of breach in the cladding of irradiated MTR fuel elements.

7. Conclusion

This work proposes a model in order to describe the release of radioactive fission products from a breached fuel plate into the storage pool water, considering diffusion through a postulated small cylindrical breach or directly out of a large hole.

The application of the model to results of sipping tests performed at the IEA-R1 research reactor on leaking MTR fuel elements correlates successfully the specific activity of ^{137}Cs , measured as a function of time, with the evaluated size of the cladding breach.

Experiments must be carried out under controlled conditions to measure more accurately the fraction of the average concentration in the irradiated nuclear fuel that is released into the water as well as the diffusion coefficient of the fission product inside irradiated nuclear fuels, both at usual storage temperatures.

Once these parameters are known, the proposed model may be used to evaluate the breach size in the cladding of irradiated MTR fuel elements and to estimate the activity released from them into the storage pool water, based only on the results of sipping tests and without the employ of destructive methods or sophisticated techniques and equipments.

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