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Release of fission tritium through Zircaloy-4 fuel cladding tubes

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Abstract

In order to analyse a potential fission tritium migration from the fuel to the coolant trough the cladding, two experiments of effusion and permeation types, have been performed on ^3H release from Zry4 claddings. During the tests at 350 °C, the ^3H released activities were measured at regular intervals. In both cases, very fast release rates have been obtained in the first few days, followed by more steady release rates. A correlation has been obtained between the ^3H releases measured and the oxide formation kinetics after the initial burst. A mechanism of ^3H transport is proposed based on the behaviour of the precipitates during the oxidation of Zry4. Applied to the conditions of PWR fuels, the measurements performed and the mechanisms considered lead to an insignificant contribution of fission tritium permeation to the total inventory of the tritium in the primary coolant.
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1. Introduction

The operation of nuclear reactors is intrinsically linked to the creation of radioactive isotopes, each one having its specific health concern, depending on the mode of decay and the biological damage mechanisms. Among them tritium (^3H) is of high concern due to its β-decay and high diffusivity in any biological environment. Therefore, limitations exist concerning the release of ^3H during power reactor operation and outages (typically 50 TBq per year per reactor, depending on local conditions). In order to comply with these limits, the power plant operators have to maintain the ^3H concentration in the primary coolant at the lowest value achievable.

Among the principal sources of ^3H generation, ternary fissions contribute to the major part. For a typical 900 MWe PWR, the annual formation of tritium in the fuel is 600 TBq (i.e. 16 000 Ci). By contrast, the second source, which is due to boric acid (^{10}B $(n, 2\alpha)$ ^3H), creates only 17 TBq a year in the primary coolant. It is however assumed to be the predominant source of the ^3H, which is released during reactor operation and outages. Comparing these two quantities, it is clear that any significant release from the fuel to the coolant would significantly increase the amount of ^3H present in the primary coolant and therefore the release of ^3H from the plant.

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As hydrogen is known to ingress into the Zr alloy cladding during the oxidation process induced by the coolant water, some concern may arise with respect to any possibility of hydrogen isotopes migration during operation that would allow fission tritium to migrate from the fuel to the coolant [1,2].

Therefore the kinetics of ^{3}H permeation through the cladding is of major importance and has to be known with high accuracy. Indeed this quantity is required in order to forecast any possible additional contribution, due to fuel release, to the ^{3}H coolant inventory.

The aim of this paper is to present and discuss ^{3}H release, measured during an effusion and a permeation experiment, on Zry4 claddings under various conditions and types of ^{3}H sources. The experiments were performed at 350 °C using a set of shielded autoclaves in order to measure continuously the tritium concentration in the water.

2. Experimental procedures

2.1. Tritiated Zircaloy samples

Two sections of fuel cladding were used for the experiments. Both were made out of standard stress relieved Zry4 alloys.

The first one, used for the effusion experiment, has been obtained in the framework of a R&D programme related to tritium release of Li bearing ceramics currently underway for the development for blankets of fusion reactors [3]. The porous ceramic pellets were clad with two Zry4 tubes, the inner one was, thus, protected from the coolant by the outer one and remained unoxidized. During a 300 day irradiation the external tube was maintained at 320 °C and received a dose of 4×10^{20} n cm^{-2}. During this irradiation period, the ^{3}H was released by the Li bearing oxide and was trapped by the inner cladding. This ^{3}H doped Zry4 cladding was used as starting material for our ^{3}H release study.

The geometry of the section of the internal tube tested was OD = 8.1 mm, ID = 7 mm, height = 50 mm. The ^{3}H content of this tube after this irradiation has been measured on both ends by high temperature extraction (1700 °C) followed by gaseous activity (ionisation chamber) and/or liquid scintillation technique. Due to the high ^{3}H release fraction from the porous ceramic core, the internal cladding contained 350 GBq of ^{3}H after irradiation, i.e. about 240 ppm. Being well protected by a tight outer cladding, the tube tested was free of any external corrosion layer. However, due to its γ-radiation activity, it had to be tested in a hot cell equipped with a shielded autoclave allowing a remote control.

The second sample, used in the permeation experiment, was basically a capsule of tritiated water. It was made of a section of a fresh standard PWR fuel cladding rod (OD = 9.5 mm, thickness = 0.57 mm, height = 100 mm), on which two plugs were welded at the ends after insertion of 2 cm^3 of highly tritiated water (total activity of the water introduced = 117 GBq).

2.2. Autoclave conditions and ^{3}H activity measurements

The two samples were tested in the shielded autoclaves under similar conditions. In both cases, the temperature was 350 ± 3 °C, the pressure 16.3 ± 0.2 MPa but the water chemistry differed slightly: pure water was used for the effusion experiment, while typical PWR water conditions were used for the permeation test (1000 ppm boric acid + 2 ppm Li). The first experiment was carried out for almost two years (580 days), while the second one was discontinued after about eight months (252 days).

The ^{3}H released activities were measured at regular intervals, with high sampling rates at the beginning of the experiments (every day or every few days) and the rates were reduced after obtaining a steady release rate (weekly or monthly sampling). For each measurement, two samples of a fraction of a cm^3 have been analyzed. At each measurement operation, the autoclaves were emptied and small samples of water were used for liquid scintillation. The tritium measurement techniques were rather standard. The accuracy of the activity measurements by scintillography is considered to be better than 0.1% for counting, but reduced to 5% due to sampling procedure accuracy, while the ones corresponding to ionisation chamber and chromatography are in the range of 12–15%. Due to the long duration of the experiments, corrections were done for the natural decay of ^{3}H, considering a half life of 12.3 years, and all the activities reported in this paper have been normalized to the initial conditions.

3. Results

Results of the first experiment, the effusion of ^{3}H during the oxidation of a portion of a tritiated cladding tube, are given in Fig. 1. A very fast release rate has been obtained in the first few days, corresponding to a fractional release in the water of the autoclave of 2.8×10^{-3} of the initial inventory, followed by a steady state release with an average release rate of 1.4×10^{-6} d^{-1}. Thus almost all the release occurred at the very beginning of the experiment. A detailed analysis (Fig. 2) of the steady state region shows that an initial parabolic behaviour is followed by a more linear region after about 200 days.

In the second experiment, the permeation through the fresh cladding filled with tritiated water inside the Zry4 capsule, similar results were obtained: As seen in
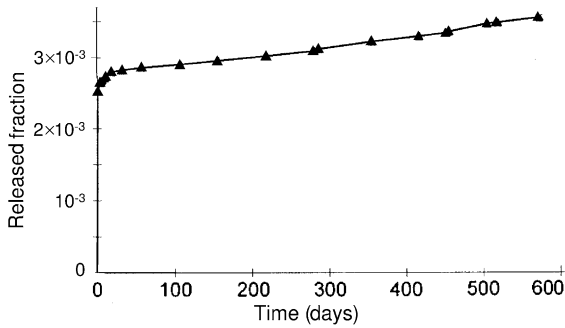
Fig. 1. Kinetics of ^{3}H activity release from the tritiated Zry4 cladding at 350 °C (Effusion experiment).
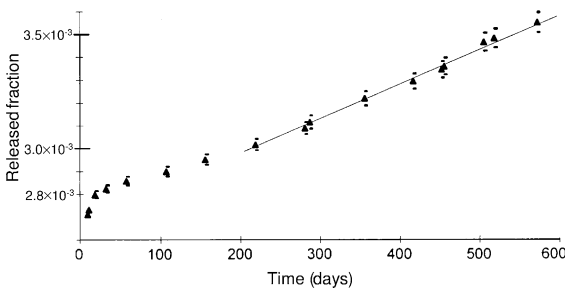

Fig. 2. Fractional release after the initial burst (detail of Fig. 1).

Fig. 3, most of the release occurred during the first week, followed by a steady release of limited amount. The amount of the release is however much lower than the release in the first experiment. The initial burst and the steady release rate are roughly three to four orders of magnitude lower than in the first experiment: the initial burst amounts to 1.7×10^{-7} of the initial ^{3}H inventory present in the internal water, and the average steady release is only 1.7×10^{-10} d^{-1}.

In a similar way, a detailed examination of the steady release, ignoring the first burst, allows us to analyse it as an initial parabolic step, with a continuously decreasing rate, followed by a faster linear regime after about 170–200 days (Fig. 4).
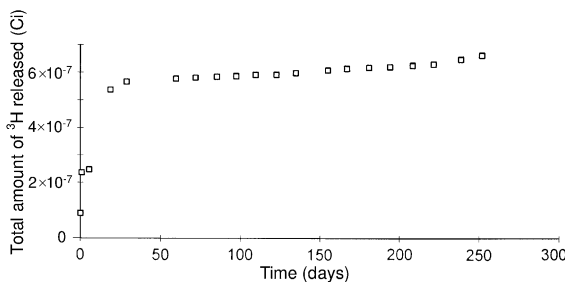

Fig. 3. Kinetics of ^{3}H activity release from tritiated water inside a Zry4 cladding capsule at 350 °C (permeation experiment).
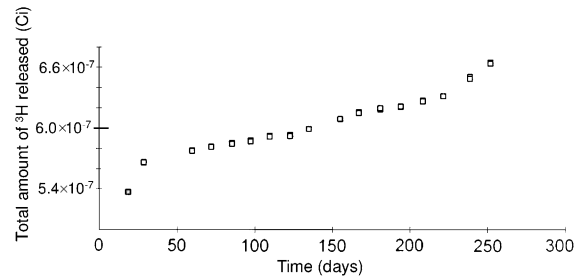

Fig. 4. ^{3}H release after the initial burst during capsule release (detail of Fig. 3).

4. Discussion

4.1. Hydrogen behaviour and Zr alloy corrosion kinetics

During the operation of nuclear reactors a fraction of the hydrogen released by the water reduction via the oxidation of the Zr alloys is picked up by the alloy. The hydrogen goes into solution or precipitates as hydrides, depending on temperature and H concentration of the alloy. Due to the brittle behaviour at low temperature of these hydrides, the kinetics of H pick-up, as well as the physical mechanisms responsible for the H ingress, has been widely studied [4].

A general observation is that a constant fraction of the H released by oxidation is picked up by the alloys. The pick-up fraction is usually dependant on the composition of the alloys, the microstructure, the corrosive environment and the temperature. Typical values of the H pick-up fractions are in the range of 10–15% for Zry4 in PWR, 60–80% for Zry2 in BWR and 5% for Zr–2.5Nb in CANDU pressure tubes. Several studies give a very sharp correlation between oxide thickness and total amount of H that was found present in the alloy [5,6]. This behaviour can be observed for very different conditions of corrosion, e.g. from long durations of a few years in operation or short oxidations performed in a few days during experimental testing. The only changing parameter is the pick-up ratio.

Since the behaviour of hydrogen in Zr alloys is involved during tritium release, a link between the behaviour of ^{3}H release measured in our experiment and the oxide formation has been searched. For that purpose, the kinetics of ZrO_2 formation has been computed using a program developed for the corrosion studies of fuel cladding tubes [7].

Due to the presence of the initial burst release of very high intensity compared to the total amount released, a direct correlation between ^{3}H release and oxidation cannot be obtained. However, assuming the initial burst to be due to a specific mechanism that will be proposed below and therefore ignoring the initial burst release,
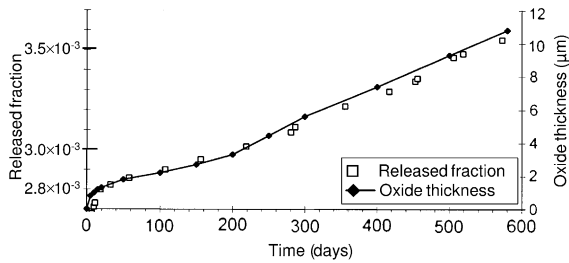
Fig. 5. Correlation between the ^{3}H release after initial burst and the oxidation kinetics (effusion experiment).

such a correlation between corrosion kinetics and ^{3}H release has been found. As shown in Fig. 5, in which both the ^{3}H release and the ZrO_2 oxide thickness of the first experiment are plotted, a matching of the two curves can be obtained using adequate scaling of the vertical axes. It corresponds to a fractional release in the linear regime of 7×10^{-5} per each μm of ZrO_2. Similar results can also be obtained for the second experiment, but then the correlation leads to a fractional release of 10^{-8} per each μm of ZrO_2, a lower value that is consistent with the lower total release in this case.

4.2. Proposed mechanisms for ^{3}H transport in Zry4

4.2.1. The role of the intermetallic precipitates for the ^{3}H release

The diffusion of hydrogen through the zirconia layer is known to be very limited. Due to the low values of the diffusion coefficients measured a rather large scatter exists among the reported values [8–10]. However, the orders of magnitude remain the same and the different experiments agree with a typical value of $D_{T/ZrO_2} \approx 10^{-19\ to\ -20}$ m^2 s^{-1} at 350 °C. With such a low diffusion coefficient, significant transport of hydrogen through the zirconia layer cannot be considered unless the oxide thickness is very low, i.e. at the very early stage of oxidation, for a thickness typically below 50 nm. This process is sketched in Fig. 6(a). Since in the two cases the cladding that was used was free of oxide at the beginning of the experiment, a mechanism based on ^{3}H diffusion through the early zirconia layers could be considered and will be analysed quantitatively below.

The two other mechanisms to be considered are related to the specific behaviour of the intermetallics precipitates during oxidation of Zry4. In this alloy, almost all the alloying metallic elements (Fe and Cr) are precipitated as $Zr(Fe,Cr)_2$ phases. For the standard heat treatments, the size of theses precipitates is typically 150–200 nm [11,12]. As the oxidation front progresses in the alloy, these precipitates are embedded in the first oxide layer where they remain metallic at first in the oxide layer. As the oxidation proceeds, new layers of oxide form, while the initial dense oxide layer trans-

forms in a more porous and less protective oxide. The precipitates can then be oxidized by direct contact with the water and/or local increase of the oxygen potential [13,14].

In addition to this delayed oxidation behaviour, these types of precipitates are known to have strong interactions with hydrogen. These intermetallic phases dissolve hydrogen to a very large amount, and allow fast diffusion of hydrogen even at low temperature. With such properties they are indeed considered for reversible H_2 storage materials. Up to one atom of hydrogen for each metallic atom can be stored in such phases [15,16]. With that respect, the role played by the unoxidized precipitates acting as diffusion short circuits through the zirconia layer has already been proven by various techniques [11,17]. For the early stages of oxidation, when the oxide thickness remains lower than the average precipitate size, such a mechanism could be active for release of ^{3}H during the initial burst (Fig. 6(b)).

The fast reduction of release rate observed would then correspond to the time at which the dense oxide thickness is larger than the precipitate size. Being then surrounded by dense zirconia, the precipitates cannot act as diffusion paths for ^{3}H from the bulk of the Zr matrix to the water anymore.

At that stage, the precipitates could however still contribute to the release of ^{3}H to the water, but only by playing the role of ^{3}H carriers. When the precipitates are in contact with the metallic alloy, they dissolve ^{3}H, to a concentration corresponding to the chemical equilibrium between the H in the Zr solid solution and the H in solution within the precipitates. Then, due to the progression of the oxidation front, they are isolated by a surrounding dense zirconia and they remain unchanged. Finally, the degradation of the oxide microstructure to a porous zirconia allows the release of ^{3}H to the water (Fig. 6(c)). The release rate will then correspond to the corrosion rate multiplied by the fraction of ^{3}H in the precipitates.

4.2.2. Behaviour during the effusion and permeation experiments

In the effusion experiment, the tritium is already present in the alloy at the beginning of the experiment, and the release occurs with constant boundary conditions on the metallic side. Indeed the total amount of ^{3}H released is only a marginal part of the initial quantity of tritium, and the total release does not affect significantly the concentration of ^{3}H in the cladding tube. We assume a homogeneous distribution of ^{3}H in the sample at the beginning of the effusion experiment, due to the long time in the testing reactor, during which long range diffusion should have occurred.

On the other hand, in the permeation experiment, the initial cladding tube is free of tritium at the beginning of the experiment. Therefore, some tritium has to be
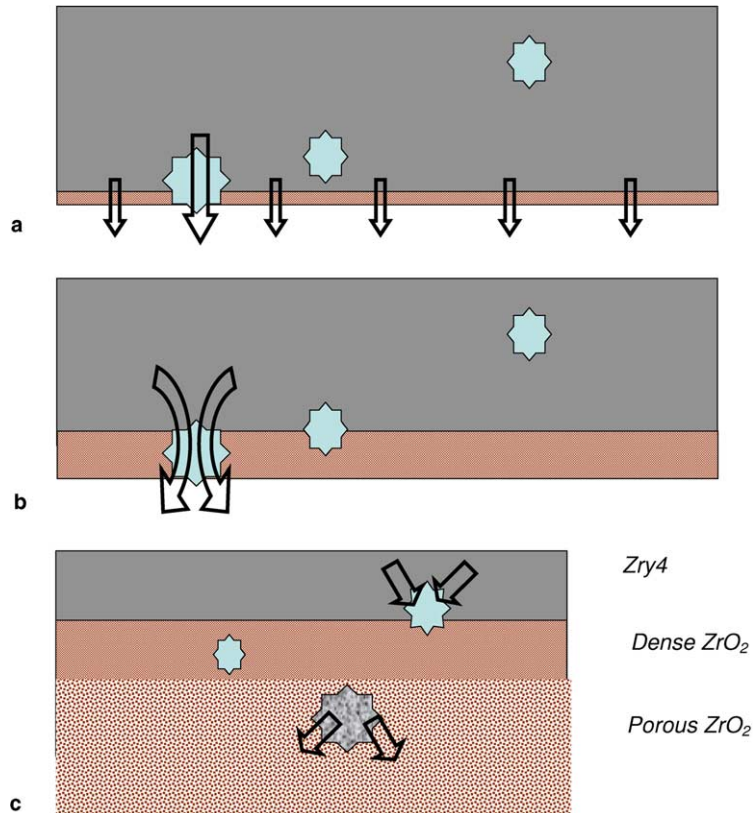
Fig. 6. Mechanisms of ^{3}H release (the arrows indicate the flow of ^{3}H from Zry4 to the coolant): (a) Diffusion through the thin initial ZrO$_2$ layer (initial burst). (b) Diffusion through the unoxidized precipitates, acting as short circuits in the dense ZrO$_2$ layer (before the transition). (c) Transport by the precipitates and release in the porous ZrO$_2$ layer (long term mechanism).

transferred from the internal tritiated water to the Zry4 cladding, before leaving the alloy and escaping to the external water. In some ways this configuration is a closer representation of an actual fuel rod, as the ^{3}H is released by the fuel, where the ternary fission takes place, before any release from the cladding to the coolant could occur. However, a difference has to be noticed, regarding the internal oxide layer. In the permeation experiment, the internal source of ^{3}H was tritiated water. Zry4 in contact with ^{3}H doped water at the temperature of the experiment will develop a layer of oxide at the typical rate presented in Fig. 5. This layer, as described above, acts as a barrier for the transfer of ^{3}H from the internal water to the alloy. Based on the mechanism of hydrogen pick-up, it is expected that the quantity of tritium picked up by the alloy during the oxidation corresponds to the pick-up fraction, assumed to be H isotope independent. After the experiment, the measurement of the total amount of ^{3}H present in the cladding by high temperature extraction gave a concentration of 9.2 MBq of ^{3}H per gram of alloy. Assuming that the H pick-up mechanisms are H isotope independent, this mount of ^{3}H in the cladding corresponds, for an oxide

thickness of 3.8 μm, to a total pick-up fraction of 8%, a value close to typical ones for such thin oxide layers [18].

In a nuclear fuel rod, the very low oxygen potential inside the fuel rod inhibits the formation of the internal oxide layer, at least up to the solid contact between pellet and cladding that is obtained only after 18 months in PWR's [19]. Thus, the tritium has no barrier to pass across for ingress into the alloy. It is, however, necessary to consider the release of ^{3}H from the fuel itself. The corresponding mechanisms will be discussed below.

4.3. Modelling

The various steps corresponding to the physical mechanisms described above have been modelled for an accurate computation of the total amount of ^{3}H released in these two experiments, aiming at extending these results to typical power reactor conditions. In this modelling, based on various diffusion and transport mechanisms, the H atoms are considered alike whatever the isotope and the ^{3}H transport is considered as the H flux multiplied by its fractional content.

The first step is to determine the kinetics of ^{3}H ingress and diffusion in the cladding in the permeation experiment. As described above this was obtained by scaling the oxide thickness with the pick-up fraction. In this experiment, it will lead to a continuously increasing concentration of ^{3}H in the alloy.

The diffusion coefficient of H in Zr alloys has been measured by various authors and is known to be high, typically $D_{H/Zr\,350\,°C} \approx 10^{-10}$ m^2 s^{-1} [20–22]. An isotope effect could be expected, reducing it by a factor of $\sqrt{3}$ for ^{3}H, but this has not been observed experimentally, due to the scatters in the measurements. The diffusion through the cladding thickness of 0.57 mm requires therefore only a few minutes, a time span negligible at the scale of the experiment, and this step will not be considered further as a rate controlling step for kinetics computation.

The initial burst release is a combination of the direct diffusion through the zirconia layer, when it is very fine, and diffusion through the unoxidized precipitates when they are in contact with both the water and the alloy.

For the diffusion through the zirconia layer, its thickness has been considered to be controlled by the oxidation kinetics law, i.e. a cubic root of time dependency, with an activation energy of 130 kJ mol^{-1} [23]. In addition to the low diffusion coefficient, the solubility of H in the zirconia is also known to be very low. For instance, maximal concentrations below 0.15 wt% have been reported in the porous layer of CANDU pressure tubes [24], but were measured to be much less in the dense part of the oxide layer which is the region of interest. There the concentrations of different H isotopes were found to be in the range of 0.01 wt% [25]. The computed value of the ^{3}H release by this mechanism in the effusion experiment, give a release fraction of 6×10^{-7}, i.e. a contribution corresponding to only 2×10^{-4} of the initial release. Thus, while this mechanism has to be considered theoretically, its contribution is also negligible, compared to the next one.

The diffusion through the precipitates, when in contact with both the metallic alloy and the water, is a problem of flow by diffusion of a given species through a conductive phase acting as a short circuit across an isolating layer. The diffusion of H in the Zr (Fe, Cr)$_2$ intermetallic phase has been found to be larger than the diffusion of H in Zr alloys [26]. Then, the problem is a standard diffusion problem, and has been solved analytically in the case of an isolated particle bridging the diffusing phase and the sink [27]. The flux of species is then given by

$$F = 4D_H . R_{eff} . \Delta c, \tag{1}$$

where D_H is the diffusion coefficient of H in the metallic phases, R_{eff} the effective radius of the precipitates, Δc the H concentration difference between the bulk of the Zry4

sample and its equilibrium concentration in contact with water.

The effective mean diameter of the precipitates in contact simultaneously with the metallic Zry and the water, R_{eff}, depends on the precipitate size $2R_P$, and on the dense oxide layer thickness t_{do}. Its value can be obtained from standard stereology computation [28] and was found to be given by the relation

$$R_{eff} = \frac{1}{2(2R_p - t_{do})} \left[\frac{\pi R_p}{2} - (t_{do} - R_p)\sqrt{t_{do}(2R_p - t_{do})} \right.$$
$$\left. - R_p^2 \sin^{-1}\left(\frac{t_{do} - R_p}{R_p}\right) \right]. \tag{2}$$

At the microstructural scale, the precipitates can be considered as remote from each other and therefore their diffusion fluxes are not interacting. The dense oxide thickness is indeed of the order of the precipitate size [14,29].

Combining all the quantities leads to the fractional release flux via the precipitates.

For the mechanisms controlled by the transport by the precipitates (Fig. 6(c)), the flux of ^{3}H is simply the volume fraction of precipitates multiplied by the partition factor for hydrogen between matrix and precipitates and by the corrosion rate. Thermodynamic computations were used to assess the partition of hydrogen between the matrix and the precipitates. It has been done using the Thermocalc® software, using the thermodynamic database developed for Zr alloys [30].

A code has been developed, named DIMITRI, (acronym of DIffusion MIgration TRItium). It is based on the mechanisms described above, but neglects the initial burst, as this initial contribution is not relevant for the behaviour of power reactor fuels. Indeed at the beginning of the irradiation of a fresh rod, obviously not ^{3}H is present in the fuel. As shown in the Fig. 7, the release kinetics obtained using this modelling follows quite well the experimental results. The agreement, better than 30%, is a noticeable achievement taking into account the absence of any tuneable parameters in the model.
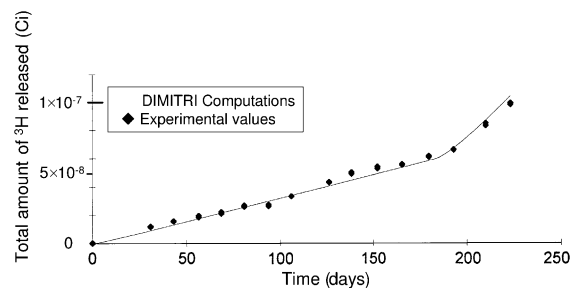

Fig. 7. Comparison between the computed and the measured ^{3}H release, in the permeation experiment, after the initial burst.

4.4. Power reactor conditions

Extending these results to the case of power reactors requires an analysis of the kinetics of release of ^{3}H out of the fuel, where it is created. Two mechanisms can be proposed: direct recoil and diffusion.

The tritium ions formed by ternary fission have a Gaussian distribution of energy, with a mean value of 8.4 MeV for the averaged fission mechanisms [31]. TRIM computations give a corresponding recoil distance of 130 µm in UO_2 or Zr alloys. This recoil distance is high enough that the ^{3}H release by direct recoil has to be considered. For the typical geometries of the PWR or BWR fuel rods, these quantities lead to a fractional release by recoil of 1.5%. The corresponding ^{3}H is delivered within the cladding directly by recoil.

For the ^{3}H escaping from the fuel by diffusion, detailed analysis has to be performed. The diffusion of tritium out of the UO_2 has been measured directly. Its coefficient was found to be several orders of magnitude larger that the diffusion coefficient of fission gases (Kr, Xe). For the centreline of the fuel (1250 K), it is in the range of 10^{-13} m^2 s^{-1} with an activation energy of 200 kJ mol^{-1} [32]. The total release by this mechanism has been computed, using standard fission product release codes based on the solution of the Booth equation. For the case of a mean fuel rod of a PWR running on a standard 4 cycles fuel management to a burn-up of 43 GW d t^{-1}, a maximum release of 40 GBq was obtained, corresponding to a total fractional release of 60%. This computation is consistent with the partition of tritium between the Zry4 cladding and the fuel measured in hot cells after irradiation [33,34].

Combining this tritium pick-up of the cladding during the irradiation, with the release to the coolant described in Section 4.2, gives a typical total release of 0.65 MBq per fuel rod during four years of irradiation, i.e. a total release for a full core of 29 GBq per year for a 900 MWe plant. In PWR, this quantity is orders of magnitude below the direct creation by the boric acid and therefore cannot be considered as a matter of concern, even for very high burn-up fuel operations.

5. Conclusions

In order to analyse the contribution of fission tritium to the total inventory of ^{3}H in the primary coolant of the PWR's, two experiments have been performed aiming at measuring the tritium release and at providing a mechanistic understanding of the mechanisms involved in the ^{3}H permeation from the fuel to the coolant.

The release kinetics have been obtained in two different experiments of effusion and permeation types. During the tests at 350 °C, the ^{3}H released activities were measured at regular intervals. In both cases, very fast release rates have been obtained in the first few days, followed by more steady release rates. The total fractional release was about 3.5×10^{-3} for the effusion test, and below 7×10^{-7} in the permeation test.

With the help of the knowledge of the behaviour of the precipitates with respect to the H pick-up mechanisms in Zr alloys, physical processes for the ^{3}H transport have been proposed. The main mechanisms induced by the precipitates (short-path diffusion through the zirconia at the beginning and ^{3}H carriers for large zirconia thicknesses) are in good agreement with the release kinetics measured.

Transposed to the actual fuel rod behaviour, the modelling proposed allows us to forecast a maximum contribution of the fission tritium to the coolant inventory of a few thousandths of the other contributions. Therefore this contribution cannot be considered as a matter of concern in pressurized water reactors.

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