# Fission Product Behaviour In The PHTS Following Fuel Failure Occurrence: New Developments

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## ABSTRACT

Recent studies using commercial reactor experience have shed more light on the behaviour of PHTS fission product evolution following failed fuel occurrence. Simulation capabilities of recently developed software tools such as STAR and VISUAL_DETECT permit characterization of failed fuel present in a Unit at any particular time and enable prediction of future evolution of the concentration of fission products in the PHTS. Also, empirical techniques have been developed for differentiating between iodine releases due to the initiation of a new fuel defect (or degradation of an existing one) from iodine spiking due to fuel-to-sheath gap washout.

## 1.0 INTRODUCTION AND BACKGROUND

In order to improve fuel performance with respect to fuel defects in its reactor Units, Bruce Power recently launched a Fuel Defect Investigation Program. In line with the objectives of the program, several integrated activities were initiated which are now in various stages of completion. One of the key elements of this program was to develop an improved understanding of radionuclide behaviour in the Primary Heat Transport System (PHTS) following fuel failure occurrence using the theory of fission products behaviour and available historical data on iodine increase events in Bruce B Units. Such improved understanding will aid the control and containment of radionuclide activity in PHTS within applicable Operating Policies and Principles by facilitating proactive

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management of in-core failed fuel elements.

The study, was carried out in collaboration with the Royal Military College Kingston Ontario, and included the following key elements:

- systematic assessment and analysis of the PHTS radionuclide activity increase events in Bruce Units (covering 28 years of commercial reactor operation)
- development of empirical technique to distinguish between iodine gap-washout events and iodine releases due to initiation of a new defect or degradation of an existing one, and
- development of software tools that can be used to interpret radionuclide evolution in the PHTS following fuel failure occurrence.

This paper is organized in four sections. Section 2.0 describes the collected historical fission products data and its assessment. Section 3.0 presents data analysis using the theory of fission products. Some conclusions are presented in Section 4.0.

## 2.0 HISTORICAL DATA COLLECTION AND ASSESSMENT

### 2.1 Data Collection

The historical data on radionuclide activity evolution in the PHTS constitutes a rich source of information for studying the behavior of fission products following fuel failure occurrence. Bruce Units use the Gaseous Fission Products (GFP) system to continuously monitor key radionuclides ($^{88}$Kr, $^{133}$Xe, $^{135}$Xe and $^{131}$I) in the PHTS by performing gamma spectrometric measurements on sample lines from the reactor header. Depressurized coolant samples are also taken regularly for 'snap-shot' measurement purpose. Details of these systems and fission products data collections are provided elsewhere [1].

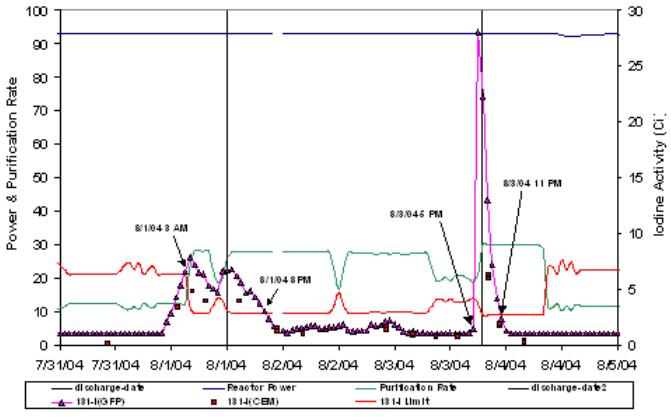
In all, about 28 years of Unit operation data was collected including historical concentrations of $^{131}$I and noble gases as well as purification rates, reactor power levels and other relevant information regarding confirmed failed fuel bundles. These data sets were examined for significant iodine release events, which were then analyzed. In examining the trends of noble gases and iodine in the collected data sets, the following key points were taken into consideration:

- Usually, the presence of a failed fuel in a Unit is indicated by steady increase in the level of noble gases (especially $^{133}$Xe) in the PHTS. The initiation of this steady increase is reasonably assumed to coincide with initiation of defects in the Unit. Since discharge dates of confirmed failed fuel bundles are available, particular fuel bundles can be linked to observations in a Unit PHTS.

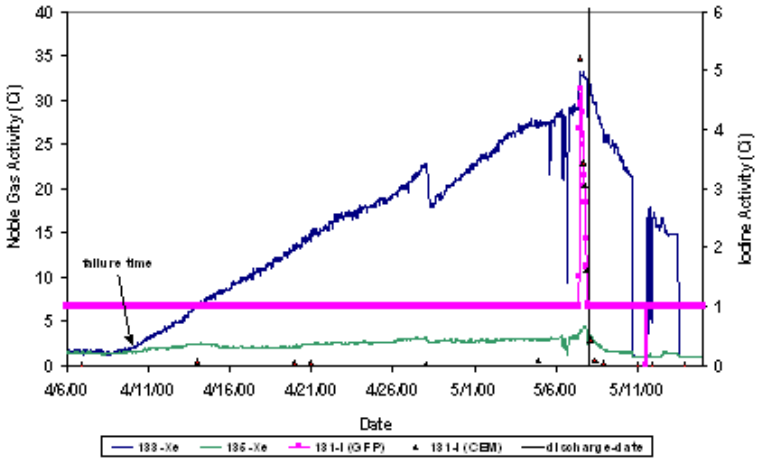
- For $^{135}$Xe atoms, a steady increase in concentration following a fuel failure can not be sustained due to absorption from neutrons.
- The onset of noble gas release into the PHTS following fuel failure occurrence may or may not be accompanied by release of measurable concentrations of $^{131}$I. Hence for identifying initiation of defect in a Unit, changes in the concentrations of noble gases are more reliable.

- Available information on discharged failed fuel bundles provides additional guidance for interpreting radionuclide data since times of channel refuelling can usually be correlated with changes in release rates from failed fuel bundles. However, when multiple failed fuel elements/bundles are involved (or the defect deteriorates), it may be difficult to separate the contributions from individual fuel elements/bundles.
- Information on defect size can typically be obtained from inspection data (including pictures of defects).
- All retrieved data was examined to identify periods with significant $^{131}$I concentration increase in the Units. These periods were then grouped into cases for further analysis. Some of the identified cases cover large periods involving multiple discharge of confirmed failed fuel bundles from a Unit, and this facilitates the assessment of the effects of both simultaneous and single failures.

Figure 1 shows an example plot containing time evolution of $^{133}$Xe and $^{135}$Xe as well as $^{131}$I from GFP and grab samples[2]. The plot also indicates the data and time that the fuel bundle containing the failed element (which caused the concentrations of the fission products to increase) was discharged. As shown in the figure, an iodine excursion (referred to as gap-washout or iodine spiking) occurred when the failed fuel was discharged from the Unit.




[2] Measurements from grab samples are labelled CEM
name of the internal Bruce Power database on which they are electronically stored.




**(a)** **(b)**

**Figure 1: Example of the time-evolution of fission products following fuel failure occurrence**

## 2.2 Data assessment

A systematic assessment of the identified iodine increase events was carried out. The selected cases represent fission products release under different scenarios: e.g. steady state reactor power versus transient (startup/shutdown), single versus multiple failed fuel elements, small versus large defect sizes etc. Given that the cases represent different scenarios, a detailed assessment will help to identify the effect of these key parameters on release behaviour.

Prior to failure, the available inventory of fission products in the fuel to sheath gap of a fuel element is a direct function of the power of the fuel and the time it has spent under irradiation (until equilibrium is attained). When the fuel element fails, $^{131}$I and other fission products are released into the PHTS coolant from the gap. The amount of fission products available for release after failure occurrence is enhanced (compared to pre-failure gap inventory) by oxidation of the failed fuel element. Depending on the fuel power at the occurrence of failure, the gap is either filled with steam or with water (i.e. due to steam condensation). It is known that mobility of $^{131}$I is much higher in water than in steam unlike noble gases [2]. As a result, a much higher rate of release of iodine into the PHTS is expected when the power of a previously failed fuel is reduced such that the temperature in the gap drops below the saturation temperature. This iodine gap-washout has traditionally been referred to as iodine spiking.

The rate of release of iodine into the PHTS is also known to depend on the size of the

defect on the fuel sheath. However, for noble gases this dependency is not a function of the defect size but the path of diffusion to the defect site. At any time, the concentration of fission products in the PHTS coolant is expected to be a balance of what is being released from the failed fuel elements and losses by decay (which depends on the decay constant) and losses through ion-exchange purification (in the case of iodine) and degassing operations (in the case of noble gases).

Given the dependency of PHTS radionuclide evolution on these various parameters, a useful assessment of these iodine increase events should at least consider the following:

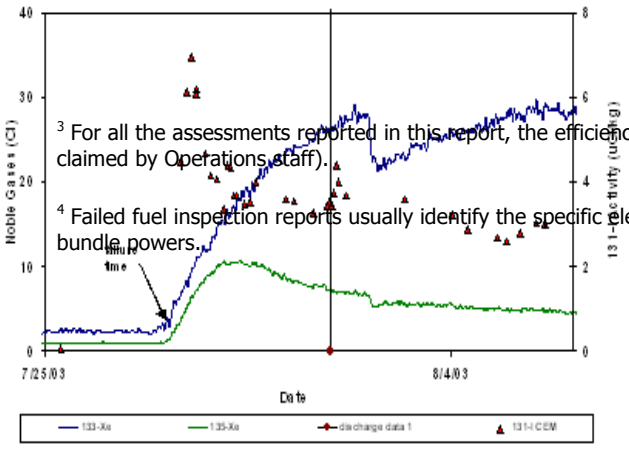
- Changes in power history of the failed fuel bundles that are associated with the periods of interest, including times of bundle shifts,
- Size of defect and presence or absence of any secondary defects,
- Changes in purification rate[3] and degassing operations,
- Time of discharge of the fuel bundles from the Unit,
- Changes in rates of release of fission products,
- Number of failed fuel elements/bundles present in the Unit during the period of interest.

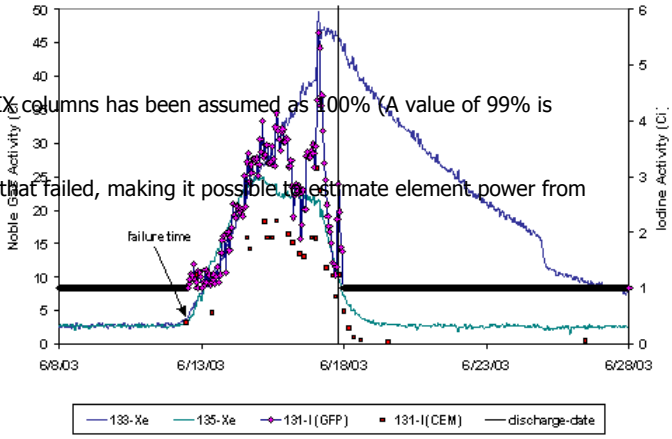
Considering the foregoing, plots of $^{131}I$, $^{133}Xe$ and $^{135}Xe$ along with purification rates and reactor power were produced for each case. The specific times of discharge of identified failed fuel bundles for each case were indicated on the plots. Power histories for the failed elements were calculated from bundle power histories[4], and times of power changes due to bundle shifts were noted. Release rates of $^{131}I$, $^{133}Xe$ and $^{135}Xe$ into the coolant were also determined. The trends of observed iodine and noble gases release into the PHTS were then examined along with these pieces of information.

## 2.3 Observations from Assessment

The following general observations and deductions were made from the exercise as described in the previous section.

- At the initiation of defect (for a gap filled with steam), there are two release rates of interest that directly determines the evolution of fission products in the PHTS: $RR_{p->g}$, the rate of release from fuel pellet to fuel-to-sheath gap and $RR_{g->c}$, the




[3] For all the assessments reported in this report, the efficiency of the IX columns has been assumed as 100% (A value of 99% is claimed by Operations staff).

[4] Failed fuel inspection reports usually identify the specific element(s) that failed, making it possible to estimate element power from bundle powers.

rate of release from fuel-to-sheath gap to the coolant. $RR_{p\text{-}>g}$ depends on fuel element power while $RR_{g\text{-}>c}$ depends on the size of defect for iodine. This means that for relatively small defect sizes (small stress corrosion cracking failure), the release rate of iodine into the coolant will be very small. If a high rate of iodine release into the coolant is observed at the initiation of defect, then the defect size must be relatively large (compare Figures 2a and 2b).

**(a)** **(b)**

**Figure 2: PHTS Radionuclide time history following a fuel failure occurrence**

- With a given amount of iodine inventory in the fuel-to-sheath gap available for release, an iodine peak can be observed in the PHTS at the initiation of failure depending on the following relationship:
  - for $RR_{p\text{-}>g} \geq RR_{g\text{-}>c}$, no activity peak is observed
  - for $RR_{p\text{-}>g} < RR_{g\text{-}>c}$, activity peak is observed

- For noble gases, the release rate to the coolant does not depend on size of the defect (it only depends on the length of the path from the release point to the defect site).

- Since the mobility of iodine is much higher in water than in steam, when steam condensation takes place in the fuel-to-sheath gap (due to bundle power shift or Unit shutdown), iodine gets washed out of the gap into the coolant. This process due to ionic diffusion and is also convection assisted; hence the dependence of release rate on defect size is different from when the gap is filled with steam. For small defect sizes, it is therefore common not to observe any significant increase in PHTS iodine level for long period while the concentrations of noble gases are steadily increasing (indicating the presence of a failed fuel) until the channel containing the failed fuel bundle is refueled or the Unit is shut down.

- In some cases, spikes in the activity of noble gases were also observed (especially $^{133}Xe$ and $^{135}Xe$) as shown in Figure 1a.

It is possible to distinguish iodine release due to gap-washout from the release that is primarily controlled by diffusion (i.e. release in steam due to new fuel failure initiation or degradation of pre-existing defects).

In general, release rate of a fission product can be approximated by the following equation:

$$R = \frac{\Delta C}{\Delta t} + (\lambda + \beta)C \qquad \text{(E-1)}$$

where

$R$ is the release rate in Ci/hr,
$\frac{\Delta C}{\Delta t}$ is the rate of change of concentration *C* (Ci) with time (at time t)
$\lambda$ is the isotope decay constant ($hr^{-1}$)
$\beta$ ($hr^{-1}$) is the ratio of purification flow rate to the mass of coolant in the PHTS multiplied by the purification efficiency of the Ion Exchange column (assumed in this assessment to be equal to 100%). For noble gases, $\beta = 0$ (i.e. no degassing).

In order to avoid the effect of purification rate changes on the calculation where necessary, concentrations of iodine were corrected for purification rate before release rates were determined.

For each case, the release events were classified as either *iodine gap-washout* or *release in steam*. The classification was done by examining the timing of release along with available data on fuel bundle shift, bundle discharge, reactor power and bundle power history data.

By comparing the release rates (RR) calculated for release events due to iodine-gap washout with the events without gap steam condensation, the following rules can be used to classify iodine release events:
Releases due to gap-washout are characterised by

- Iodine release rates, RR ($^{131}I$) > 1 Ci/hr and $\frac{RR(^{133}Xe)}{RR(^{135}Xe)} > 1$

Releases in steam (due to new fuel failure or degradation of a previously failed fuel) are characterised by:

RR ($^{131}I$) ≤ 1 Ci/hr and $\frac{RR(^{133}Xe)}{RR(^{135}Xe)} \leq 1$

## 3.0 FISSION PRODUCTS MODELLING AND ANALYSIS

The concentration of a fission product in the PHTS can be determined as a function of time by performing a mass balance across the fuel pellet, the fuel-to-sheath gap and the PHTS coolant. The following sections present a development of the relevant mass balance equations in each of these three systems as coupled partial differential equations.

## 3.1 Governing equations

### 3.1.1 Mass conservation in the fuel pellet

Under irradiation conditions, fission products are released into the free void space (fuel-to-sheath gap) within the fuel element via solid-state lattice diffusion. The mass balance equations for describing radionuclide concentration distribution in a fuel grain of radius *a*, which accounts for generation of fission products (by fission) and losses by diffusion and radioactive decay is as follows (see Reference 3 for derivation):

$$\frac{\partial u(\eta,t)}{\partial t} = \underbrace{\frac{D'(t)}{\eta^2}\frac{\partial}{\partial \eta}\left(\eta^2 \frac{\partial u(\eta,t)}{\partial \eta}\right)}_{DIFFUSION\ IN\ \ GRAIN} - \underbrace{\lambda u(\eta,t)}_{LOSS\ BY\ DECAY} + \underbrace{F(t)y}_{\substack{PRODUCTION \\ BY\ FISSION}} \qquad \text{(E1)}$$

where

$u = CV$ i.e. fission product concentration, $C$ (atom $m^{-3}$) $\times$ fuel volume $V$ ($m^3$) (E2)

$$\eta = r/a,\ D' = D/a^2 \qquad \text{(E3)}$$

*a* equivalent fuel grain sphere radius (m)
*r* radial coordinate
*D* fission product diffusion coefficient in fuel matrix ($m^2\ s^{-1}$)
*D′* empirical fission product diffusivity in the $UO_2$ fuel matrix (= $D/a^2$) ($s^{-1}$)
$\eta$ radial coordinate (m)
*t* time (s)
$\lambda$ radioactive decay constant ($s^{-1}$);
*F* average fission rate per defective rod (fission $s^{-1}$); $F_f(t)$ (fission/s) = $1.489 \times 10^{13}$ $P(t)$ (kW/m) [2]
y fission product yield (atom fission$^{-1}$)

The initial and boundary conditions are defined as follows:

Initial condition: $u(\eta,0)=0$ for $0 < \eta < 1$ (E5)

Boundary conditions: $u(1,t) = 0$ $\eta=1$ (E6)

$$\frac{\partial u}{\partial \eta} = 0 \qquad \eta=0 \qquad \text{(E7)}$$

### 3.1.2 Mass conservation in the pellet-sheath gap

The fission products released into the fuel-to-sheath gap will migrate towards the fuel defect site where they may be eventually released into the PHTS coolant. The mass balance equation for the fission product inventory in the gap ($N_g$) is a balance of the diffusional release from the fuel matrix ($R_{dif,}$ atoms $s^-$), losses due to radioactive decay and release from the failed element.

$$\frac{dN_g}{dt} = \underbrace{R_{dif}(t)}_{RELEASE\ \text{FROM FUEL}} - \underbrace{\lambda N_g}_{\text{LOSS BY DECAY}} - \underbrace{\nu(t)N_g}_{RELEASE\ \text{TO PHTS COOLANT}} \quad \text{(E8)}$$

where the diffusional release rate from the fuel pellet is given from Fick's law as:

$$R_{dif} = -3D' \left.\frac{\partial u}{\partial \eta}\right|_{\eta=1} \quad \text{(E9)}$$

The loss to the PHTS coolant is a function of the escape rate coefficient $\nu$ ($s^{-1}$). The diffusion coefficient $D'$ can be empirically related to linear power rating (kW/m) for a single defect element as follows [2]:

for $^{131}I$ $$D' = 10^{9.857\log P - 25.1314} \quad \text{(E10)}$$

and for noble gases $$D' = 10^{8.632\log P - 23.4091} \quad \text{(E11)}$$

for linear element power ratings $P$ in the range 25 kW/m to 60 kW/m.

It should be noted that these effective diffusion coefficients were derived from experimental data for oxidized fuel pellets. This means that the gap inventory calculated by this model already accounts for enhanced diffusion due to oxidation. In this case, under pre-failure condition, $N_g$ refers to the inventory of fission product that will be available for release once the fuel sheath fails, assuming instantaneous oxidation. A more advanced model can incorporate a fuel oxidation model at the initiation of fuel failure to account for the variation of diffusion coefficient with oxidation. Also, a diffusion coefficient relationship corresponding to normal fuel stochiometry can be incorporated for accurate prediction of $N_g$ prior to failure.

### 3.1.3 Mass Conservation in the PHTS

The fission product inventory in the PHTS coolant ($N_c$) can be determined by a mass balance of the source release from defective fuel rod(s) and losses due to radioactive decay and coolant purification as given in equation E12:

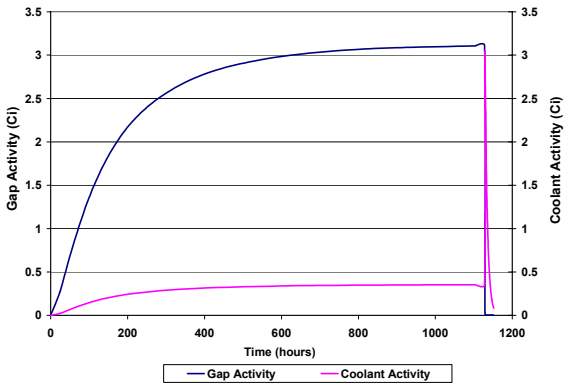
$$\frac{dN_c}{dt} = \underbrace{\nu(t)N_g}_{RELEASE\ FROM\ ELEMENT} - \underbrace{\lambda N_c}_{LOSS\ BY\ DECAY} - \underbrace{\beta(t)N_g}_{LOSS\ BY\ PURIFICATION} \quad \text{(E12)}$$

The three-coupled conservation equations can be solved to obtain the concentration of a specific fission product in the PHTS coolant and the fuel-to-sheath gap as a function of time. The only parameters that are unknown are the escape rate coefficient, $\nu$ and the element power P.

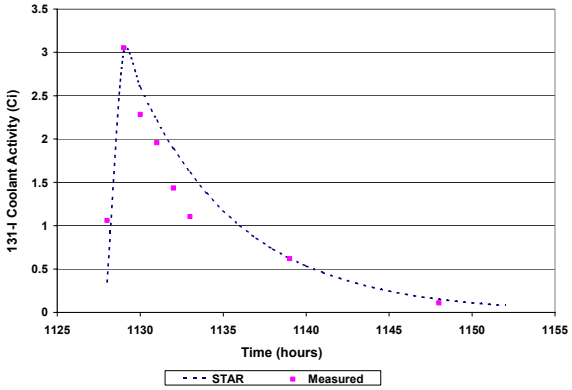
The newly developed STAR (Steady-state and Transient Activity Release) numerically implements these three coupled conservation equations, thereby permitting further detailed analysis of the collected historical data. Figures 3a[5] show example simulations of $^{131}$I evolution following a single failed fuel occurrence using data from the case shown in Figure 3c. Being able to simulate the fission product concentration in the fuel-to-sheath gap enables proper accounting of releases into the PHTS. STAR simulation works by estimating the defect size (through the gap escape rate coefficient) and power rating of the failed element using fission products measurements obtained by the GFP. Once the escape rate coefficient and the power of a suspected failed element have been estimated, further releases from the fuel into the coolant can be estimated as a function of time.

STAR can be used as a predictive tool whereby coolant activity levels that will result from a steady-state situation or during iodine transient (at a given reactor power and purification flow) following a fuel failure occurrence, can be estimated. Such estimation can be done by tuning gap escape rate coefficient and the failed element power to fit measured coolant activity data from GFP or grab samples data. Further details on how this type of analysis can be used for proactive failed fuel management in a reactor Unit are provided elsewhere [1].

[5] Figure 3a shows STAR's prediction of the evolution of iodine in the fuel-to-sheath gap of a failed fuel (data shown in Figure 3c) as well as iodine evolution in the coolant. The iodine spike that was observed on discharging the fuel is compared with STAR's prediction in Figure 3b.

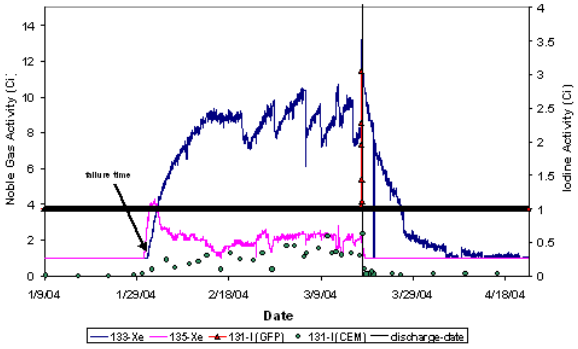



(a)




(b)

**Figures 3: STAR simulated $^{131}$I evolution in the fuel-to-sheath gap and the coolant (5,6) using data from the case presented in Figure 7.**




(c)

## 4.0 CONCLUSIONS

Using data from 28 years of commercial reactor experience, improved understanding of fission products behavior in the PHTS following fuel failure occurrence has been developed. Iodine releases that occur as a result of gap-washout (i.e. due to condensation in the fuel-to-sheath gap of a failed fuel) can be distinguished from iodine increases due to failed fuel degradation or the due to the initiation of new defects, by calculating the source release rate. Newly developed software tool such as STAR provides capability to analyze fission products data making it possible to predict future evolution of PHTS radionuclide concentration.

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