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Storage of defective fuel pins in SFR core

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Abstract

An open fuel pin failure is a breach in the fuel pin cladding that allows direct contact between the primary coolant and the nuclear fuel. In this paper we focus on the sodium-fuel interactions in a Sodium cooled Fast neutrons Reactor (SFR), reviewing the main aspects of the fuel pin failure evolution with an emphasis on the Reaction between the Oxide fuel and the Sodium (ROS). This reaction leads to the formation of an uranoplutonate phase with approximately half the density of the initial oxide. In turn this can cause significant fuel swelling and the potential further degradation of the fuel pin. The maximal fuel swelling due to the formation of the uranoplutonate can be estimated for non-irradiated fuel based on the physico-chemical properties of the pellets, as further described in this paper.

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Keywords: uranoplutonate, sodium fast reacors; fuel pin failure; sodium-fuel reaction, fuel swelling

1. Introduction

Of the several types of reactors considered for development as part of the generation IV, the Sodium Fast Reactors (SFR) have raised the most interest and over 400 reactor-years of experience was gained globally in operating them [1]. The renewed interest of the CEA in the fast-neutron reactor R&D have led to the present-day design studies for the construction of the sodium-cooled fast neutrons reactor ASTRID, with several improvements via-à-vis previous CEA-operated SFRs[2].

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Since the safe operation of the reactor is the highest priority concern for the CEA, this study is focused on the sodium-fuel interactions and the subsequent fuel pin failure evolution, in order to evaluate the available data relative to the requirements imposed by the ASTRID operational conditions.

A fuel pin failure represents a breach in the pin cladding which is the first confinement barrier of the nuclear fuel inside the reactor core. It can lead to the contact between the coolant and the compounds inside the fuel pin including the nuclear fuel, and to a chemical reaction that alters the fuel behavior and could potentially lead to fuel dissemination in the primary coolant. The phenomenology of the fuel pin failure evolution as a result of the Reaction between the Oxide fuel and Sodium (ROS), both under irradiation in reactor conditions as well as in internal storage will be addressed in the first part of this paper, followed by a brief discussion on some of the thermodynamic data available on the sodium-fuel systems. The kinetic aspects of the reaction are also addressed in this paper and since the data currently available on the sodium –fuel reaction rate is limited, the need for further research is underlined. In this regard a series of experiments could be performed in order to investigate the reaction rate variation with different parameters. Since the pellet swelling can give an indication of the advancement of the reaction, we describe a simplified method for estimating of the maximum radial fuel swelling due to the ROS for non-irradiated fuel, based on the initial fuel pellet characteristics such as its diameter, density, Pu/M and the reaction temperature. In kinetic studies this can provide an estimation of the progress of the reaction relative to the equilibrium but it could also be used to estimate the maximum fuel swelling and diametric deformation of the fuel pin in Beginning Of Life (BOL) fuel pin failures.

2. General phenomenology of a fuel pin failure

Once the sodium infiltrates the fissile column the Reaction between the Oxide fuel and Sodium (ROS) is initiated forming the Fuel-Sodium Reaction Product (FSRP) of which the main component is the trisodium uranoplutonate, more or less pure depending mainly on the fuel burnup. This compound has physical properties very different from the initial mixed-oxide. In particular the density and thermal conductivity are listed in Table 1.

Table 1. Main physical properties comparison of the $(U,Pu)O_2$ and Na_3MO_4

Compound	$(U,Pu)O_2$ [3]	*Na_3MO_4 [4]
Theoretical density (g/cm^3)	10.99 – 11.08 (10 - 30 at% Pu)	5.59 – 5.68 (depending on T)
Thermal conductivity ($W \times m^{-1} \times K^{-1}$)	1.7 – 5.4 depending on temperature, composition, burnup, O/M	0.9 – 1.0 (depending on T)
Morphology	Compact sintered grains	Laminar layers with a high porosity

*M=U+Pu ;physical properties of the $Na_3(U,Pu)O_4$ with less than 30 % molar Pu are considered similar to those of Na_3UO_4

Sodium uranoplutonate has roughly half the density of the original mixed-oxide leading to a lower thermal conductivity and considerable swelling of the fuel. This can favor the further degradation of the fissile column and the propagation of the fuel pin failure. While under irradiation in the reactor core the high temperature of the fuel surface and the high radial thermal gradient of the fuel that causes a radial oxygen migration, favor the ROS. Thus the defective assembly is placed in a low-power position referred to as Internal Storage where the reaction is significantly slowed by the low fuel temperature and by the absence of a thermal gradient that can supply the oxygen.

Although the ROS and its effects on the safety of the reactor operation have been the focus of multiple studies, many thermodynamic and kinetic aspects of the reaction are still unknown.

2.1. Fuel pin failure evolution under irradiation in core conditions

Stratin *et al*[5] observed the fuel pin failure evolution and propagation as a result of the ROS under irradiation in the reactor core in experimental “Run Beyond Cladding Breach” irradiations. They have shown the deleterious effects of the ROS coupled with significant fluctuations in the reactor power. However the ROS alone is unlikely to

lead to fissile matter dissemination since the FSRP layer forms around the mixed-oxide, coating the fuel with a cohesive structure. As long as the operational conditions (power level and temperature) are constant, the fuel pin failure will eventually reach an equilibrium state imposed by the oxygen potential. This is also consistent with observations made on fuel pin failures in the PHENIX reactor (Figure 1).

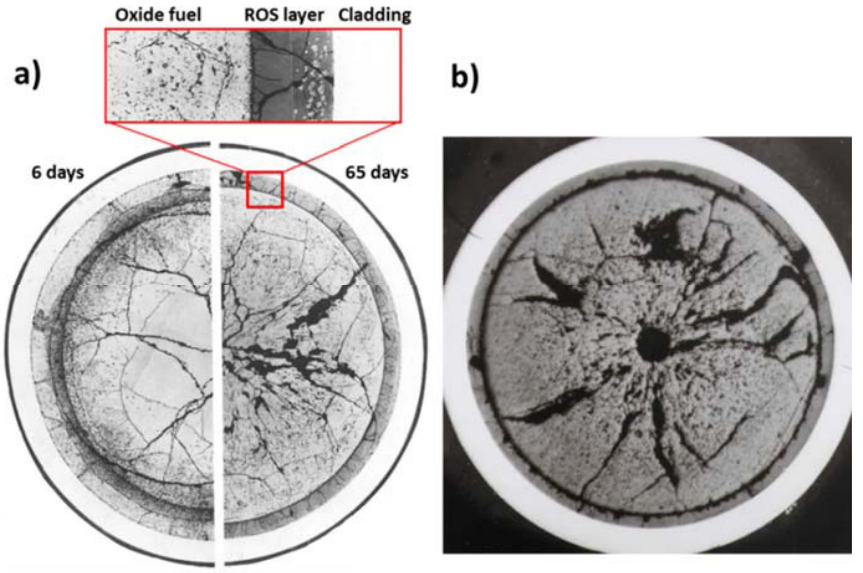

Figure 1. Comparison of ROS aspect in RBCB tests [5] (a) and a fuel pin failure in the PHENIX reactor (b)

In nominal conditions there is an important radial temperature gradient in the fuel. In these conditions the ROS is limited to the outer region of the fuel by oxygen redistribution and the operating temperature of the fuel. The reaction occurs initially somewhat inward from the fuel surface, near the dissociation isotherm of the sodium uranoplutonate (≈1400°C) where the temperature allows the fastest kinetics. The ROS layer increases progressively outwards and forms a monolithic layer between the fuel and the cladding [5].

This mechanism is predominant in Beginning Of Life fuel pin failures, in which the fuel-cladding gap is open and the sodium can easily infiltrate the fissile column. In the case of End Of Life fuel pin failures the ROS morphology can vary significantly due to the presence of fission products that can alter the reaction kinetics[6]. Also the closed fuel-cladding gap may cause a non-uniform sodium infiltration.

2.2. Fuel pin failure evolution in internal storage conditions

In internal storage or during reactor shutdown, the temperature of the fuel is much lower and there is no significant radial temperature gradient to cause the oxygen migration to the reaction site. The low temperature also allows the uranoplutonate formation in the whole volume of the pellet, but the reaction is much slower and limited to the grain boundary[7]. In these conditions the main oxygen source is present as an impurity in the primary sodium (less than 2 ppm) especially if the available oxygen in the fuel has already been consumed by the ROS in core conditions. Although this oxygen concentration is extremely low, it can still be considered as a significant supply due to the vast total amount of primary sodium. However the ROS kinetics is slowed by the lower temperature[6] and limited by the sodium flow rate in the fuel and the Na-fuel oxygen exchange.

Observations from the PHENIX reactor fuel pin failures have shown little evolution of the ROS in internal storage over an experimental 300 Effective Full Power Days (EFPD) storage time Figure 2.

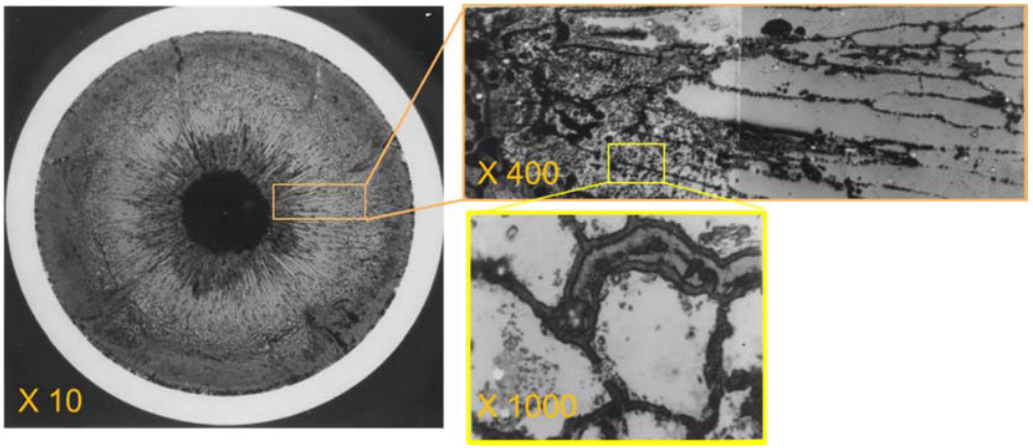

Figure 2.PIE image of the sodium-oxide reaction after prolonged internal storage in the PHENIX reactor of a defective fuel pin

Although the ROS can be observed at the center of the pellet, which is possible only in internal storage conditions, the progress of the ROS over the 300 EFPD relative to the 90 EFPD storage time (standard storage time for the PHENIX) reactor could not be quantified and no significant overall evolution of the fuel pin failure was observed.

3. Chemistry of the sodium fuel interactions

The main parameters controlling the sodium-fuel reaction are the oxygen potential and the fuel burnup. In non-irradiated fuel the oxygen potential is dependent on the Pu loading which controls the O/M limit to which the fuel can be reduced and thus controls the amount of oxygen available for the ROS[6,8].

3.1. Thermodynamic limitation of the ROS

The thermodynamic limitations of the sodium-oxide fuel reaction have been studied in several out-of-pile tests investigating the influence of the Pu/M, $\Delta\bar{G}(O_2)$ and temperature on the uranoplutonate formation (see Table 2).

Table 2. Equilibrium oxygen potential variation with the temperature, for the uranoplutonate formation

Equilibrium	$\Delta\bar{G}_{O_2}^{eq}(J\cdot mol^{-1})$	Reference
$3Na_{(l)}+ UO_{2(s)} + O_2 \rightleftarrows Na_3UO_{4(s)}$	$-945000 + 261\cdot T$ ($\pm$ 4.2)	Adamson [8]
$3Na_{(l)}+ UO_{2(s)} + O_2 \rightleftarrows Na_3UO_{4(s)}$	$-949789 + 253\cdot T$ ($\pm$ 2.7)	Mgnanelli [7]
$3Na_{(l)}+ UO_{2(s)} + O_2 \rightleftarrows Na_3UO_{4(s)}$	$-949072 + 264.6\cdot T$	Smith [10]
$3.014Na_{(l)}+0.986\ UO_{2(cr)} + 1.014O_2 \rightleftarrows Na_{3.014}U_{0.986}O_{4(cr)}$	$-944773 + 255.6\cdot T$	Smith [10]
$3Na_{(l)}+ U_{1-x}Pu_xO_{2-y(s)} + (1+y/2)O_2 \rightleftarrows Na_3U_{1-x}Pu_xO_{4(s)}$	$-907394 + 244.9\cdot T$	Lorenzelli [11]

The equations reported in the literature for the equilibrium oxygen potential of the uranoplutonate formation are in very good agreement (Figure 3). However the errors associated with these equations are not always available, thus given the uncertainties [7,8] they could be considered valid to within experimental error for fuels containing up to 30% Pu.

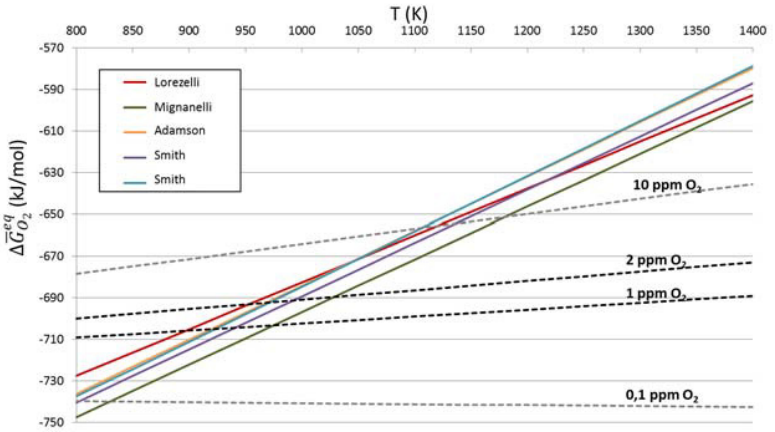

Figure 3. $\Delta\bar{G}_{O_2}^{eq}$ for the formation of the uranoplutonate relative to the oxygen potential of the sodium containing 0.1 to 10 wppm O_2

The oxygen potential threshold of the uranoplutonate formation was estimated at 1.5 wppm of O_2 in sodium at 900 K and 5 wppm of O_2 in sodium at 1000 K[9]. This is however valid only for non-irradiated fuel, and further studies are required for a complete thermodynamic description of the systems involved in the ROS.

In order to accurately predict the fuel pin failure evolution, especially over long internal storage periods the reaction kinetics also needs to be considered. There is little data available so far in this regard[6, 7] and new studies could bring important insights into the fuel-sodium reaction rate and its variation with various parameters.

3.2. Estimating the maximum fuel swelling due to the ROS

The maximum fuel swelling could be used as criteria for the in-pile evolution of fuel pin failures as well as for the determination of the progress of the reaction relative to the equilibrium in out-of-pile studies of the ROS kinetics.

The main source of oxygen for the ROS under irradiation in core conditions is the oxygen in the fuel: either available at the sodium–fuel interface or supplied to the reaction site mostly by radial migration from the fuel in the center of the pellet to the periphery. In non-irradiated fuel the oxygen potential is dependent on the Pu loading which controls the O/M limit to which the fuel can be reduced and thus controls the amount of oxygen available for the ROS. The overall sodium-fuel reaction in core conditions can be written as:

$$\mathbf{(z\text{-}y)Na_{(l)} + (2+z)U_{1-x}Pu_xO_{2-y(s)} \rightleftarrows (z\text{-}y)Na_3U_{1-x}Pu_xO_{4(s)} + (2+y)U_{1-x}Pu_xO_{2-z(s)}} \text{ with } z > y[9] \quad (1)$$

Considering a non-irradiated fuel pellet, the ROS can be represented according to Figure 4 where:

ϕ_p is the initial diameter of the pellet

ϕ_f is the final diameter of the pellet at equilibrium

τ is the thickness of the uranoplutonate layer

φ is the thickness of the fuel layer in the initial pellet that reacted to form the uranoplutonate

$\mathbf{r_1}$ is the radius of the reduced fuel in the center of the pellet

$\mathbf{r_2}$ is the final radius of the pellet at equilibrium

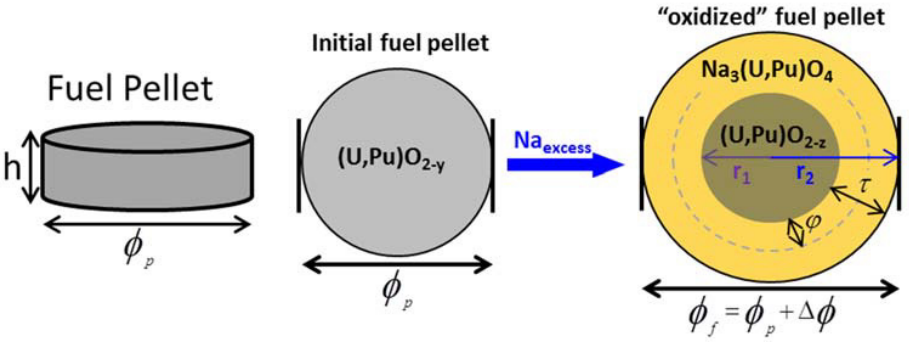

Figure 4. Schematic representation of the fuel pellet swelling due to the ROS

The total oxygen mass per unit of height in the pellet can be given by:

$$m_{O_{2,MOX}} = \frac{\pi \emptyset_p^2}{4} \cdot \rho_p \cdot x_{o_{2,MOX}} \quad (2)$$

where:

$m_{O_{2,MOX}}$ is the total oxygen mass in the initial pellet.
ρ_p is the initial density of the pellet.
$x_{o_{2,MOX}}$ is the mass fraction of oxygen in the initial fuel.

Part of this oxygen will be incorporated into the uranoplutonate phase directly by the ROS. Since the uranoplutonate is formed as a layer surrounding the reduced fuel at the center of the pellet, the total oxygen mass per unit of height in the uranoplutonate layer can be given by:

$$m_{O_{2,Na_3MO_4}} = \pi \cdot (r_2^2 - r_1^2) \cdot \rho_{Na_3MO_4} \cdot x_{o_{2,Na_3MO_4}} \quad (3)$$

Considering: $(r_2 - r_1) = \tau$ and $(r_2 + r_1) \approx \emptyset_p$

$$m_{O_{2,Na_3MO_4}} = \pi \cdot \emptyset_p \cdot \tau \cdot \rho_{Na_3MO_4} \cdot x_{o_{2,Na_3MO_4}} \quad (4)$$

where:

$m_{O_{2,Na_3MO_4}}$ is the total oxygen mass in the uranoplutonate layer (M = U, Pu).
$\rho_{Na_3MO_4}$ is the density of the uranoplutonate.
$x_{o_{2,Na_3MO_4}}$ is the mass fraction of oxygen in the uranoplutonate.

This oxygen comes in part from the fuel that reacted with sodium according to :

$$m_{O_{2,ROS}} = \frac{M_{O_{2,MOX}}}{M_{M_{MOX}}} \cdot x_{M\ Na_3MO_4} \cdot \pi \cdot \emptyset_p \cdot \tau \cdot \rho_{Na_3MO_4} \quad (5)$$

where:

$m_{O_{2,ROS}}$ is the mass of oxygen in the uranoplutonate coming from the ROS.

$M_{O_{2,MOX}}$ is the molar mass of oxygen in the fuel.

$M_{M_{MOX}}$ is the molar mass of metal (U, Pu) in the fuel.

$x_{M\,Na_3MO_4}$ is the mass fraction of metal (U, Pu) in the uranoplutonate.

The rest of the oxygen is supplied by diffusion from the fuel at the center of the pellet that is reduced to the O/M limit according to:

$$m_{O_{2,mig}} = \frac{z}{2} \cdot \left(\frac{\pi \times \emptyset_p^2}{4} \cdot \rho_p \cdot x_{O_{2,MOX}} - \frac{M_{O_{2,MOX}}}{M_{M_{MOX}}} \cdot x_{M\,Na_3MO_4} \cdot \pi \cdot \emptyset_p \cdot \tau \cdot \rho_{Na_3MO_4} \right) \quad (6)$$

Where:
$m_{O_{2,mig}}$ is the oxygen mass in the uranoplutonate coming from the radial oxygen migration.
z is the final oxygen stoichiometry in the reduced fuel $(U,Pu)O_{2-z}$ [6,8].
$x_{M\,Na_3MO_4}$ is the mass fraction of metal (U, Pu) in uranoplutonate.

Thus, according to (5) and (6), the thickness of the uranoplutonate layer can be estimated according to :

$$\tau = \frac{x_{O_{2,MOX}} \emptyset_p \cdot \rho_p \cdot z \cdot M_{M_{MOX}}}{4 \cdot M_{O_{2,MOX}} \cdot x_{M\,Na_3MO_4} \cdot (2+z) \cdot \rho_{Na_3MO_4}} \quad (7)$$

Knowing the value for the thickness of the uranoplutonate layer τ, allows for the estimation of the maximal diametral swelling of the pellet. Assuming no axial mass transfer, the thickness of initial fuel layer that reacted with the sodium can be expressed as:

$$\varphi = \frac{x_{M\,Na_3MO_4} \cdot \rho_{Na_3MO_4}}{x_{M\,MOX} \cdot \rho_p} \cdot \tau \quad (8)$$

Thus the maximal diametric expansion of the pellet due to the ROS can be estimated to:

$$\Delta_{\max} \phi = 2 \cdot (\tau - \varphi) \quad (9)$$

assuming no axial expansion, or

$$\Delta_{\max} \phi = \frac{2 \cdot (\tau - \varphi)}{3} \quad (10)$$

assuming an isotropic 3 dimensional expansion.

4. Conclusions and perspectives

The sodium-fuel reaction is the driving force behind the fuel pin failure evolution and the main parameters controlling this reaction are the temperature, fuel burnup and the oxygen potential. The phases in the Na-U-O system have been identified and their structural and thermodynamic properties have been assessed[9]. Applying the same approach to phases in the Na-Pu-O and the phases containing the main soluble fission products such as Zr, and lanthanides (in particular Ce)[6,8] could allow a complete thermodynamic description of the fuel-oxide interactions

under irradiation in core conditions as well as in internal storage for all burnup values. However in order to accurately model the fuel pin failure evolution in internal storage, especially for long-term periods of over 1000 days, a kinetic description of the ROS is also required and in this regard the available data is limited. New insights into the kinetics of the ROS could however be made by studies of the reaction rate and its variation with several parameters. The progress of the reaction relative to the equilibrium could be experimentally determined using an estimation of the maximal fuel swelling at equilibrium based on the initial fuel pellet properties: diameter, density, Pu/M and the reaction temperature.

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