

SOURCE IST 2.0 PHENOMENA MODELLING

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ABSTRACT

This paper traces the phenomena modelled by SOURCE IST 2.0 from the Technical Basis Document for Validation of CANDU Computer Programs to the final coding. For each primary phenomenon for fission product release identified in the Technical Basis Document (and the Fission Product Release and Transport Validation Matrix), the model selected is described.

The SOURCE IST 2.0 code employs a subdivision of the fuel into basis units. Since the fuel volume changes with temperature and porosity, the mass of uranium initially present in the basis unit serves as the constant parameter for describing the basis unit. SOURCE IST 2.0 simulates the transfer of fission products between the following inventory partitions: the fuel grain matrix, the fuel grain boundary, the fuel surfaces, the fuel-clad gap, and that released to the coolant. SOURCE IST 2.0 models all of the primary phenomena affecting fission-product release from CANDU fuel under accident conditions, namely

- Fission yield, decay and transmutation
- Fission-product diffusion
- Grain growth
- Grain boundary sweeping
- Fission-product redistribution due to matrix stripping (fuel volatilization), UO₂/Zircaloy interaction, UO₂ dissolution by molten Zircaloy, fuel melting, grain boundary separation, and grain boundary coalescence / tunnel interlinkage
- Fission-product vaporization
- Gap transport
- Releases due to rewet
- Leaching releases.

SOURCE IST 2.0 includes models for the effect of fuel stoichiometric deviation (x in UO_{2+x}) on the fission-product diffusion coefficient. The models reflect the current state of knowledge. In the few cases in which consistent conceptual models can not be constructed, the respective phenomena are implemented with limit-consequence approaches. The corresponding numerical solutions include solution of coupled partial differential equations (PDE), differential-algebraic equation (DAE) systems, ordinary differential equation (ODE) systems, and algebraic equations. Each implementation of each phenomenon has its own internal time step control to maintain stability of each model within the calculational stream. The model of fission product diffusion within the grains uses a dynamic grid that grows with increases in grain size.

INTRODUCTION

SOURCE IST 2.0 models fission-product releases due to the primary phenomena listed in Table 1. The phenomena identification codes used in Table 1 were taken from the validation matrix. SOURCE IST 2.0 also simulates the production of fission products and actinides in the fuel during normal operation and their release under accident conditions. The effects of fission, radioactive decay and neutron transmutation processes on the fission-product and actinide inventories are modelled.

In SOURCE IST 2.0 terminology, a basis unit is the geometric unit for which release calculations are performed. This may be a fuel element, an axial segment, or an azimuthal ring within a segment, or some other geometry. SOURCE IST 2.0 uses the following inventory partitions within a basis unit to track releases from the fuel: the fuel grain, the fuel grain boundary, the fuel surface, the fuel-sheath gap, and the released inventory. The fractional release of each radionuclide is defined as the total of all releases of that nuclide (not accounting for decay, during both normal operating conditions, any decay steps, and during the transient) divided by the total inventory at the beginning of the transient.

FPR-01 ATHERMAL RELEASE

This phenomenon compiles all fission-product release mechanisms that are not temperature activated. These mechanisms are recoil, knockout and thermal spike. The first is the direct recoil of fission fragments from within a layer equal to the range of the fission fragments in UO_2 . The second describes the releases produced by the elastic collisions (and the associated collision cascade) that a fission fragment experiences in passing through the fuel. The last mechanism addresses the release of the fission products inside the volume of a fission spike that intersects a fuel free surface. Athermal release is not a primary phenomenon in any CANDU reactor accident scenarios. This phenomenon is treated as an irradiation-enhanced athermal diffusion term in the diffusion model in SOURCE IST 2.0. (See FPR-02 below).

FPR-02 DIFFUSION

The dominant mechanism for the release of fission products from a single fuel grain, at temperatures higher than 1000 K is the escape of fission-generated atoms by lattice diffusion to the grain surface. This phenomenon describes the releases of fission products from the grain to the grain boundaries.

The Booth diffusion model is used in SOURCE IST 2.0 to describe the releases of fission products from the fuel grains to the grain boundaries [1]. The Booth model represents the average behaviour of the fuel grains as a set of "equivalent" spheres whose total area is comparable to the total grain boundary surface. The original model has been generalized to describe the cumulative diffusional release of various members of a radioactive decay chain during several cycles of reactor operation followed by a high-temperature transient [2,3]. The implementation in SOURCE IST 2.0 simulates diffusional releases from equiaxed (spherical) and columnar (cylindrical) fuel grains using the Booth equivalent sphere, and takes into account intrinsic (temperature-activated) diffusion, vacancy-assisted diffusion due to irradiation damage and fuel oxidation, and irradiation-enhanced athermal diffusion [4]. Actinides are not released from the fuel grains by the diffusion mechanism.

In the time between diffusion steps, the grains may grow. This growth is accounted for by adding a shell of uranium oxide to the grain that is free of fission products. The old concentration profile is fitted to the new grain by cubic spline interpolation preserving the zero concentration gradient at the grain centre and maintaining the same gradient at the location of the old surface.

FPR-03 GRAIN BOUNDARY SWEEPING/GRAIN GROWTH

Under certain conditions, some of the grains that form a sintered pellet (polycrystalline material) may grow at the expense of the other grains. The effect of this growth is the increase in size of the average grain in the fuel as a function of time. During this process, the displacement of the grain boundaries 'sweeps' the insoluble fission products existing in the shrinking grains. These fission products accumulate on the grain boundaries [5]. The SOURCE IST 2.0 grain boundary sweeping model transfers 100% of all fission products initially present in the swept portion of the fuel grains from the fuel grain inventory partition to the grain boundary inventory. The actinides are not released from the fuel grains by the grain-boundary sweeping module.

Equiaxed grain growth is modelled in SOURCE SIT 2.0 using the model of Hastings *et al.* [6]. In the event that the equiaxed grain growth results in a change in grain length that is smaller than the change calculated for columnar grain growth (FPR-05) the grain length increases by value calculated for columnar grain growth, and the grain diameter increases by the amount calculated for equiaxed grain growth.

FPR-04 GRAIN BOUNDARY COALESCENCE/TUNNEL INTERCONNECTION

The UO_2 used in nuclear power reactors is in the form of a sintered pellet. The boundaries between neighbouring grains are sites for the accumulation of fission products (FPR-02 and FPR-03). These fission products form grain boundary bubbles in equilibrium with the surroundings. As more fission products arrive at the grain boundaries, the diameters of the bubbles increase. When the bubbles coalesce, the excess fission products are transported to the grain corners where tunnels are formed. When enough fission products are accumulated in the tunnels, the tunnels will interlink and the excess fission products will be released to the fuel free surfaces.

The pressure of noble-gas fission products in the bubbles on the fuel grain boundaries, the surface tension of the grain boundary bubbles and the local hydrostatic stress on the fuel are used to calculate bubble growth. The grain boundary bubbles are assumed to interlink at a radius of about $0.7 \mu\text{m}$ [7]. When the bubbles interlink, the grain boundary coalescence / tunnel interlinkage model in SOURCE IST 2.0 transfers the excess fission products from the grain boundary inventory partition to the fuel surface and fuel-sheath gap inventories. The excess amount of the noble-gas fission products is transferred directly to the fuel-sheath gap inventory. The same fraction of the other fission products is transferred to the fuel surface inventory.

FPR-05 VAPOUR TRANSPORT/COLUMNAR GRAINS

At high temperatures and in the presence of a steep temperature gradient, columnar grains can grow towards the centre of the fuel. The mechanism for columnar grain formation is

the movement of large pores up the temperature gradient by the molecular diffusion of molecules of the fuel matrix from the hot side of the pore to the cold side through the contained gas. The migrating pore collects the fission products dissolved in the evaporating region of the fuel. Columnar grain growth is simulated up to a maximum grain length of 200 μm . Once the grain length reaches 200 μm , the grain boundary bubbles continue to sweep a 200 μm length. SOURCE IST 2.0 simulates releases from the fuel grains to the grain boundaries due to columnar grain growth using the grain-boundary sweeping model (FPR-03).

FPR-06 FUEL CRACKING (THERMAL)

When the fuel is subjected to temperature changes it may crack to relieve thermal stresses. The cracks will most likely be produced in the pellet periphery during heating and in the pellet centre during cooling. If the cracks intersect intra-granular bubbles, grain boundary bubbles or tunnels, the fission products contained in these structures will be released to the fuel free surfaces. If the fuel is subjected to rapid cooling from high temperature during the rewet accident phase (water quench), the cracks are normally large in number and very small (microcracking). The fission-product releases due to fuel cracking during temperature changes are addressed in SOURCE IST 2.0 by providing the surface area of each axial fuel segment to the code as an input parameter.

FPR-07 GAP TRANSPORT (FAILED ELEMENTS)

The fission products stored in the gap may be released to the coolant if the cladding fails. In this case, the stored fission products will migrate from their location to the defect site and release to the coolant. The presence of water in the gap accelerates the releases of soluble fission products (FPR-19). It is generally accepted that this migration can be described by a desorption/ adsorption reaction and atomic diffusion in the gap environment.

SOURCE IST 2.0 uses a simple bookkeeping model for fission-product releases from the gap. After the fuel element failure time (provided as input to the code), all noble gases in the fuel-sheath gap inventory partition are transferred to the released inventory. Fission products that are subsequently transferred into the fuel-sheath gap are immediately transferred to the released inventory partition. Fission products other than noble gases are released from the fuel surface by fission-product vaporization (FPR-15).

FPR-08 GAP RETENTION

Some fission products (e.g., Te) that are released from the fuel matrix can chemically react with the cladding under certain conditions. The fission products react with metallic components of the cladding. Once the cladding is fully oxidized these fission products will be released according to their volatility (FPR-15). Gap retention is not simulated in SOURCE IST 2.0. Releases of Te and other fission-products that undergo chemical interactions with the Zircaloy fuel sheathing will be overestimated by the code.

FPR-09 UO_{2+x} FORMATION

When UO_2 is exposed to an oxidizing environment, it oxidizes to an equilibrium stoichiometry, which depends on the fuel temperature and the oxygen potential of the steam. For a certain range of conditions, the fuel matrix accommodates the excess oxygen without a phase transformation, forming UO_{2+x} . The equilibrium composition can be calculated by equating the oxygen potential in the steam to the oxygen potential in the uranium oxide. Stoichiometry changes in the uranium oxide have a significant effect on the material properties of the fuel and on the release of fission products. Small changes in fuel stoichiometry may produce very large changes in the release kinetics of some fission products.

The stoichiometric deviation in the UO_2 fuel is an input to SOURCE IST 2.0. The diffusion coefficient used by the code to simulate releases from the fuel grains to the grain boundaries depends on the stoichiometry of the fuel (FPR-02). The Killeen and Turnbull model for diffusional releases from hyperstoichiometric fuel is used in SOURCE IST 2.0 [4].

FPR-10 U_4O_9 - U_3O_8 FORMATION

At certain temperatures and oxygen partial pressures, UO_2 oxidation will involve a phase change. The formation of these other phases (e.g., U_4O_9 or U_3O_8) is more likely in highly oxidizing environments (e.g., air). When phase changes are involved, the fuel oxidation process is very complex and may involve the sequential formation of several uranium oxides. Fission-product releases can be affected by cracking of the matrix due to volume changes associated with the phase transformation, as well as by the changes in material properties (e.g., thermal conductivity, diffusion coefficients).

UO_2 oxidation to U_4O_9 or U_3O_8 involves a phase change, but in SOURCE IST 2.0 both of these phases are considered to be hyperstoichiometric UO_2 . Diffusion in U_4O_9 is simulated using the UO_{2+x} diffusion coefficient for $x = 0.25$. The Lindner and Matzke diffusion coefficient is used for U_3O_8 [8]. If the stoichiometric deviation is between 0.25 and 0.67, the uranium oxide is partitioned into two phases, $UO_{2.25}$ and U_3O_8 , for the purposes of the diffusion calculation using the lever rule. (This approach inherently assumes that U_4O_9 does not exist as a separate phase but is merely the upper limit of UO_{2+x} .) The concentration distributions of the fission products in the fuel grains in a basis unit arising from diffusion in $UO_{2.25}$ and U_3O_8 are calculated separately within a time step and merged at the end of the time step. If the stoichiometric deviation of the fuel in a basis unit exceeds 0.458 (half way between U_4O_9 and U_3O_8), the fuel is deemed to have undergone grain boundary separation (see FPR-18).

FPR-11 UO_{2-x} FORMATION

In highly reducing environments (e.g., hydrogen-rich) or if the UO_2 is in contact with Zircaloy or other materials, it is possible to reduce UO_2 to UO_{2-x} . Stoichiometry changes in the UO_2 have a significant effect on the release of fission products. The reduction and associated release kinetics are sensitive to the reducing agent as well as temperature and the contact characteristics.

No model, comparable to that of Killeen and Turnbull [4], exists for the diffusion coefficient of fission products in hypostoichiometric uranium oxide (UO_{2-x}). The results of Miekeley and Felix, however, suggest that the rare gas diffusivity will decrease in hypostoichiometric fuel [9]. In the absence of a detailed model, the diffusion coefficient

expression for stoichiometric uranium dioxide fuel is also used for hypostoichiometric urania in the SOURCE IST 2.0 code.

FPR-12 UO_2 - ZIRCALOY INTERACTION

Uranium and zirconium can form an alloy. The melting point of the alloy varies with its composition, and can be as low as $\sim 1150^\circ C$. This alloy can be formed through reduction of the UO_2 (to $UO_{2-x} + U$) by Zircaloy, which is still in the solid state. With contact between Zircaloy and UO_2 , a series of metallographic structures will develop, i.e., outward from the fuel pellet surface one observes: (i) a mixture of α -Zr(O) and (U,Zr); (ii) a layer of (U,Zr); (iii) a layer of α -Zr(O); and (iv) the remainder of the Zircaloy sheath. In this process, some fuel "liquefaction" may result in the fuel element, well below the melting point of UO_2 . The occurrence of this liquefied fuel can contribute to an enhanced fission-product release as a consequence of the fuel reduction.

Uranium and zirconium can form an alloy through interactions at temperatures below the melting point of Zircaloy. The fraction of the original uranium in a basis unit that has liquefied due to UO_2 - Zircaloy interaction is provided as an input to SOURCE IST 2.0. Simple rules are used to transfer the nuclide inventories from this liquefied fraction to the appropriate inventory partition. The fuel grain and grain boundary inventories of noble-gas fission products in the liquefied fraction are transferred to the fuel-sheath gap. The fuel grain and grain boundary inventories of non-noble-gas fission products in the liquefied fraction are transferred to the fuel surface inventory. The fuel grain inventories of actinides in the liquefied fraction are transferred to the fuel surface inventory.

FPR-13 UO_2 DISSOLUTION BY MOLTEN ZIRCALOY

If fuel heatup in an accident is sufficient to produce molten Zircaloy, the Zircaloy can act to reduce the UO_2 , producing a liquid (U,Zr)O + (U,Zr) O_{2-x} mixture. The (U,Zr)O is a liquid at temperatures ($\sim 1150^\circ C$) which are significantly below the melting point of UO_2 . This dissolution and liquefaction of the UO_2 matrix may result in significant fission-product release of all isotopes which either have a large partial pressure or which can form volatile compounds in the prevailing environment. The fraction of the original uranium in a basis unit that has liquefied due to UO_2 dissolution by molten Zircaloy is provided as an input to SOURCE IST 2.0. The same simple rules used for releases due to UO_2 - Zircaloy interaction are also used for UO_2 dissolution by molten Zircaloy.

FPR-14 FUEL MELTING

At sufficiently high temperatures, UO_2 melts. Stoichiometric UO_2 melts congruently at a unique temperature, but hypo- or hyper-stoichiometric UO_2 does not and has separate solidus and liquidus temperatures. Non-stoichiometric UO_2 has solidus and liquidus lines that are lower in temperature than the congruent melting point of $UO_{2.00}$. This liquefaction of the UO_2 matrix may result in significant fission-product release of all isotopes which either have a large partial pressure or which can form volatile compounds in the prevailing environment. The fraction of the original uranium in a basis unit that has liquefied due to UO_2 melting is provided as an input

to SOURCE IST 2.0. The same simple rules used for releases due to UO_2 - Zircaloy interaction are also used for UO_2 melting.

FPR-15 Fission Product Vaporization/Volatilization

The rate-limiting process for release of the volatile or relatively volatile fission products (Xe, Kr, Cs, I, Te, Sb) is solid-state diffusion through the UO_2 fuel matrix. However, for non-volatile fission products (e.g., Sr, Y, Zr, Nb, Mo, Tc, Ru, Rh, Pd, Ba, La, Ce, Pr, Nd), their escape from free surfaces into the gas phase may be so difficult that the surface or gas-phase transport processes can control the overall kinetics. This rate-limiting step is related to the chemical state of the fission products (i.e., vapour pressure of the thermodynamically-favoured compounds) that exist at the fuel surface, and to the mass transfer from the free surface of the fuel to the bulk, hydrogen-steam gas flow.

The vaporization of non-noble-gas fission products from the fuel surface inventory partition to the released inventory in SOURCE IST 2.0 is based on the equilibrium partial pressures of the gaseous fission products and boundary layer mass transfer considerations [10,11]. The equilibrium partial pressures were determined by Gibbs-energy minimization utilizing the F*A*C*T thermodynamic program [12] for a system containing specific proportions of fuel and fission products, and a gas phase atmosphere containing steam and hydrogen. The extensive F*A*C*T results were incorporated into a database for integration into SOURCE IST 2.0. The complete partial pressure results are reconstructed from the fitted element partial pressure representations and the F*A*C*T Gibbs energy functions utilizing the method of chemical potentials. The boundary layer mass transfer coefficient is calculated using a heat/mass transfer analogy [13,14].

FPR-16 MATRIX STRIPPING

If the Zircaloy sheath is severely oxidized, the oxidizing environment can react with UO_2 , and, under these conditions, the UO_2 will incorporate excess oxygen to form hyperstoichiometric UO_{2+x} or be converted to a higher oxide (see FPR-09 and 10). At temperatures above 1800 K, UO_{2+x} is the stable condensed phase in both air and steam. A significant vapour pressure of uranium-bearing species is predicted at equilibrium. Since the gases in contact with the fuel do not constitute a closed system, the condensed phase is vaporized at a relatively rapid rate due to incongruent vaporization of the UO_{2+x} matrix. As the matrix is volatilized, the remaining noble gases (Xe, Kr) and volatile fission products (Cs, I, Br, Rb) will be released from the volatilized fuel volume. The low volatility fission products that were previously inside the fuel matrix become concentrated at the fuel surface, where they can accumulate or else be vaporized (FPR-15) or entrained in the flowing gases as particulates. This phenomenon has been described as "matrix stripping" or fuel volatilization. The fraction of the original uranium in a basis unit that has vaporized due to UO_3 formation is provided as an input to SOURCE IST 2.0. Simple rules are used to transfer the nuclide inventories from this vaporized fraction to the appropriate inventory partition. The fuel grain and grain boundary inventories of noble-gas fission products in the vaporized fraction are transferred to the fuel-sheath gap. The fuel grain and grain boundary inventories of non-noble-gas fission products in the vaporized fraction are transferred to the fuel surface inventory. The fuel grain and fuel surface inventories of actinides in the vaporized fraction are transferred to the fuel-sheath gap inventory.

FPR-17 TEMPERATURE TRANSIENTS

In-pile power-ramp tests have produced greater gas (Xe and Kr) release than expected from a pure diffusion mechanism. The gas release rates are also higher than those measured during steady state irradiations. Experimental evidence showed that for heating rates higher than 50 K/s the most probable mechanism of release was grain boundary separation (see FPR-18). Enhanced releases from the grains were required to supply the grain boundaries with the required fission product inventories to match the observed releases. The most probable cause of this higher release was the production and movement of over-pressurized intra-granular or grain boundary bubbles.

The work of Une and Kashibe [15] and more recent studies at CRL suggest that the fission-product releases observed during temperature ramps are predominantly from the fuel grain boundaries. No effect of temperature ramp rate on the fission-product diffusion coefficient in the fuel grains is modelled in the SOURCE IST 2.0 code. The grain boundary coalescence / tunnel interlinkage model (FPR-04) releases fission products from the grain boundaries to the fuel-sheath gap during temperature ramps.

FPR-18 GRAIN BOUNDARY SEPARATION

If rapid releases from the grains to the grain boundaries occur (see FPR-17), the grain boundary bubbles will be over-pressurized until vacancy migration re-establishes equilibrium. If the forces produced by the bubble over-pressure are higher than the cohesive forces between grains, the grains will separate. The fission products accumulated on the grain boundaries may be released to the fuel free surfaces.

In SOURCE IST 2.0, grain boundary separation occurs either when the fuel temperature reaches 2400 K or when more than half of the fuel in a basis unit is converted to U_3O_8 (see also FPR-10). Once a basis unit undergoes grain boundary separation, it remains in that state. The grain boundary separation model transfers the fission-product grain boundary inventory to the fuel surface inventory (or directly to the fuel-sheath gap inventory for the noble gases).

FPR-19 FISSION PRODUCT LEACHING

After rewet, the fuel will be submerged in water. The UO_2 in all elements with cladding failures will be in contact with water and soluble fission products stored in the gap and the grain boundary will leach out from the fuel elements.

SOURCE IST 2.0 includes models for two aspects of fission-product release due to water ingress into a fuel element, namely rewet and leaching. The rewet model transfers (1) the soluble fraction of the fission-product inventory from the fuel surface, and (2) the entire fission-product inventory from the fuel-sheath gap to the released inventory. The rewet model is called only once during a SOURCE IST 2.0 calculation, at the time of water ingress into the fuel element provided as input to the code. Longer-term releases are simulated using the leaching model. The fraction of the original uranium in a basis unit that has dissolved or mechanically eroded due to the action of water on the fuel is provided as an input to SOURCE IST 2.0. The fuel grain, grain boundary and fuel surface inventories of fission products and actinides in the dissolved fraction are transferred to the released inventory partition.

CONCLUSIONS

This paper has described the models and the theoretical basis for the phenomena simulated by SOURCE IST 2.0 which is undergoing qualification by the Canadian Nuclear industry as the Industry Standard Tool for calculating fission-product release from fuel.

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Table 1: PRIMARY FISSION-PRODUCT RELEASE PHENOMENA
SIMULATED BY SOURCE IST 2.0

ID	Title
FPR-02	Diffusion
FPR-03	Grain Boundary Sweeping/Grain Growth
FPR-04	Grain Boundary Coalescence/Tunnel Interlinkage
FPR-05	Vapour Transport/Columnar Grains
FPR-06	Fuel Cracking (Thermal)
FPR-07	Gap Transport (Failed Elements)
FPR-09	UO_{2+x} Formation
FPR-10	U_4O_9 - U_3O_8 Formation
FPR-11	UO_{2-x} Formation
FPR-12	UO_2 Zircaloy Interaction
FPR-13	UO_2 Dissolution by Molten Zircaloy
FPR-14	Fuel Melting
FPR-15	Fission Product Vaporization/Volatilization
FPR-16	Matrix Stripping
FPR-17	Temperature Transients
FPR-18	Grain Boundary Separation
FPR-19	Fission Product Leaching