SAND2010-0800C

MELCOR Fission Product Release Model for HTGRs

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Abstract - *A fission product release and transport model for High Temperature Gas cooled Reactors (HTGRs) is being developed for the MELCOR code. HTGRs use fuel in the form of TRISO coated fuel particles embedded in a graphitized matrix. The HTGR fission product model for MELCOR is being developed to calculate the released amounts and distribution of fission products during normal operation and during accidents. The fission product release and transport model considers the important phenomena for fission product behavior in HTGRs, including the recoil and release of fission products from the fuel kernel, transport through the coating layers, transport through the surrounding fuel matrix, release into circulating helium coolant, settling and plate-out on structural surfaces, adsorption by graphite dust in the primary system, and resuspension. The fraction of failed particles versus time is input by a particle failure fraction response surface of particle failure fraction as a function of fuel temperature, and potentially, fuel burn-up. Fission product release from the fuel kernel and transport through the particle coating layers is calculated using diffusion-based release models. The models account for fission product release from uranium contamination in the graphitized matrix, and adsorption of fission products in the reactor system. The dust and its distribution can be determined from either MELCOR calculations of the reactor system during normal operation, or provided by other sources as input. The distribution of fission products is then normalized using the ORIGEN inventory to provide initial conditions for accident calculations. For the initial releases during an accident, the existing MELCOR aerosol transport models, with appropriate modifications, is being explored for calculating dust and fission product transport in the reactor system and in the confinement. For the delayed releases during the accident, which occur over many hours, and even days fission product release is calculated by combining the diffusion-based release rate with the failure fraction response surface input via a convolution integral. The decay of fission products is also included in the modeling.*

I. INTRODUCTION

MELCOR is a fully integrated, engineering-level computer code that was originally developed to model the progression of light water reactor severe accidents. The code is being modified to include models for HTGR confirmatory safety analysis. This paper describes a fission product release model for HTGRs under development for MELCOR. This model is to be used both for release under accident conditions and during normal operation. HTGRs use fuel in the form of TRISO particles embedded in a graphite matrix. The graphite matrix is either in the form of pebbles, for the Pebble Bed Reactor (PBR) or in a cylindrical fuel compact, for the Prismatic Modular Reactor (PMR).

The PBR design is among the designs currently being considered by the US DOE for the Next Generation Nuclear Plant (NGNP) Very High Temperature Reactor (VHTR). The pebble bed reactor design has fuel consisting of TRISO coated fuel particles about 0.5 mm in diameter, embedded in a graphitized spherical matrix (fuel pebbles) 6 cm in diameter. There are about 15,000 TRISO coated fuel particles randomly embedded in each fuel pebble in the central 5 cm diameter fueled region of the pebble, surrounded by a 0.5 cm thick unfueled region. The fueled region of the active core may be cylindrical or annular, with outer, top, and bottom reflectors. The central reflector region in the annular PBMR268 reference design was dynamic, consisting of unfueled graphitized pebbles that circulated downward through the reactor core with the fueled pebbles.[1] This design was later changed to incorporate a solid fixed central graphite reflector.[2] The most recent PBMR design is to be cylindrical dynamic core with fueled pebbles across the active core.

The PMR design is also currently being considered by the US DOE for the NGNP VHTR.[2] One of the PMR reactor designs is to be based on the General Atomics Modular High-Temperature Gas-Cooled Reactor (MHTGR) design, which consists of an annular fueled region with fixed inner, outer, top and bottom graphite reflectors. The active core is comprised of hexagonal graphite fuel blocks with cylindrical channels drilled for the fuel and coolant. The fuel in the fuel channels consists of 5.0 cm long by 1.25 cm diameter cylindrical graphitized fuel compacts containing TRISO coated fuel particles.

TRISO fuel particle for both the PBR and PMR designs consists of a fuel kernel surrounded by four coating layers: a buffer layer of porous pyrolytic carbon, a dense inner pyrolytic carbon layer, a dense silicon carbide layer, and a dense outer pyrolytic carbon layer, see Figure 1.

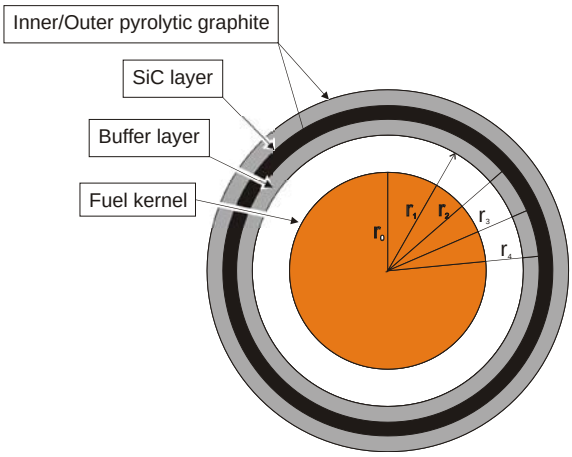

Figure 1. TRISO particle

The HTGR fission product model for MELCOR is to calculate the released amounts and distribution of fission products in the modeled reactor system. To do this, it is necessary to calculate the amount and species of fission products generated and released during normal power operation. This includes the amount (and species) of fission products generated in the fuel kernels, the amount recoiled and diffused to the buffer layer, the amount diffused through the dense coating layers, the amount released from the kernels of particles with failed coating layers , the amount which diffuses through the graphitized matrix (and for PMRs the graphite fuel block) and the distribution of fission products released and distributed to the reactor system and adsorbed on dust and other surfaces in the reactor system.

For HTGR accident scenarios, literature has primarily focused on a few fission products: Kr, Xe, I, Ag, Sr and Cs. This is partly because of the important contribution of these fission products to public and worker doses. Moreover, these fission products are the only ones whose release has been extensively measured.

II. TECHNICAL APPROACH

The approach for the MELCOR fission product release model has been developed based on considerations of the important phenomena for fission product behavior in HTGRs.[3] There are two general populations of TRISO particles: those with intact coating layers and those with failed coating layers. A small fraction of fresh fuel particles are also modeled as failed from the start owing to manufacturing defects, consistent with the design specification, and generally about 10^{-5}. The fraction of failed particles during operation will be provided by an empirical fuel particle failure model. Development of a particle failure model or implementation of suitable existing models is separate from the effort described in this paper.

For intact particles, the gaseous fission products released are assumed to accumulate in the buffer; for failed particles, fission products are assumed to go directly to the graphitized matrix. The condensable (metallic) and non-condensable (gaseous) fission product release will be calculated using diffusion-based release models. This calculation is performed during normal operation, to determine the partition of the fission product inventory between the kernel and the buffer layer, and the amount released from failed particles.

There are also contributions to fission products from uranium contamination in the graphite matrix. In HTGRs, fission products are not just present in the fuel; they may also accumulate, via adsorption, in graphite dust that is generated and subsequently distributed in the reactor system, for a PBR, graphite dust is generated primarily due to abrasion during the circulation of the pebbles through the core and transport within the fuel pebble recirculation system. Dust is also present, but to a lesser degree in the PMR reactor system. The quantity and distribution of dust during normal operation could be calculated by a stand-alone model or determined from MELCOR calculations of the operating reactor system provided appropriate models are incorporated into MELCOR. Once again, development of a dust generation model or implementation of suitable existing models is separate from the effort described in this paper. A liftoff model for the dust is necessary for calculation of the accident consequences. Existing MELCOR aerosol transport models can provide the framework for calculation of dust transport in the reactor system.

II.A Main Features of Model

MELCOR nodalizes the active core into axial levels and radial rings; the two indices, axial and radial, then define a cell location. Within a core cell, there are separate temperatures for each component, such as fuel particle, graphite matrix, surrounding graphite structures, etc. There would be a separate calculation of, for instance, fission product transport, for each core cell using the cell component temperatures.

An initial fission product inventory for an accident scenario is provided by a calculation using a burnup code such as ORIGEN. Fission products generally of interest are Kr, Xe, I, Cs, Ag, and Sr.[4] Note that although a burnup code like ORIGEN provides the fission product inventory, it does not provide data on the fission product distribution, either for release from the kernel into the buffer or for release to the matrix from initially failed particles and contamination. The fission product distribution in the TRISO particle and in the reactor system must either be calculated by MELCOR from a normal reactor power operation run or provided by another code. The fuel failure fraction is provided by a user-input particle failure

fraction versus temperature curve or particle failure fraction versus temperature and burnup.

Neutronic input parameters are also needed for the reactor power shape, point kinetics model, and reactivity feedback coefficients if it is desired to run accidents without scram.

Other input would include initial failure fraction of TRISO particles, distribution and amount of graphite dust in the primary system, and distribution and amount of fission products plated out during operation. The distribution information would either be provided by the user from experimental data or be calculated.

II.B Solution Strategy with MELCOR

MELCOR uses a steady-state calculation for normal operation, followed by the accident simulation. The normal operation calculation is used to generate initial conditions for the accident simulation and would be done in three stages: (1) establishment of thermal steady-state using an "accelerated steady-state" option; (2) calculation of fission product distribution and release to the coolant using a diffusion model; (3) and distribution of the released fission products in the reactor system, using a "accelerated" run to establish deposition rates and locations. To do the reactor normal power operation calculation, MELCOR requires the rate of fission product generation, rate of release of fission products from failed TRISO particles and fuel matrix contamination, and graphite dust generation rate and size distribution. During normal operation, there would be releases of metallic fission products, notably Ag^{110m} and Cs, from both intact TRISO coated fuel particles and the TRISO coated particles that fail. The radionuclide transport models in MELCOR would distribute the dust in the primary system, and released fission products would accumulate on the dust and surfaces of the reactor system. For typical particle burnup of over three years and the reactor operating lifetime of 40 years, it is necessary to run an "accelerated" steady state calculation, as mentioned above, to reduce computation time. Once the trends in the distribution of dust and rate of deposition are established, the results would be scaled up to the full operating time. The final step before the transient run is to scale the fission product distributions to the reactor inventory provided by ORIGEN.

After the reactor operation run, the initial conditions for the transient problem are established. During the transient calculation, the release from intact particles and transport in the fuel matrix are again treated by the finite-difference diffusion model. To account for particle failure over time in the transient using a convolution integral (described later), the additional release from failed particles is done via an analytic model.

III. DIFFUSION RELEASE MODEL

III. A Normal Operation

Release to the graphitized matrix is modeled differently for failed and intact particles as documented in the review report.[3] For intact particles, the model includes transport through the intact coating layers and by the kernel diffusive resistance. For failed particles, the model considers only the effects of kernel fissive resistance and fission priduct recoil. For failed particles no credit is taken for the diffusive fission product transport through the particle coating layers. If any, or all, of the dense coating layers are intact, it is assumed that the gaseous fission products, such as Kr, are effectively retained in the fuel particle. However, the modeling of intact coated particles provides for the diffusive transport and release of metallic fission products, such as Cs. Fuel particle failure is assumed to involve the simultaneous failure of all of the high density coating layers.

The diffusion rate through the graphitized fuel matrix can be important for Kr and Cs. For release from the dust in the system, the surface to volume ratio is so large that immediate release is considered a reasonable approximation.

Another factor in PMRs is diffusive transport of fission products through the web of graphite between the fuel channel and the coolant channel in the graphite fuel blocks. A porous flow model is currently being considered for this diffusion effect, accounting for transport through the connected porosity of the graphite blocks.

Release from an intact particle is described by the diffusion equation:[5]

$$\frac{\partial C}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 D \frac{\partial C}{\partial r} \right) - \lambda C + \beta \qquad (1)$$

where

C	=	Concentration of nuclide (kmol/m^3)
D	=	Diffusion coefficient (m^2/s)
r	=	radial dimension (m)
λ	=	Decay constant (1/s)
β	=	Generation rate - in kernel only (kmol/m^3-s)

For diffusion during the accident phase, with reactor shutdown, the generation rate in the kernel, β, *is set to* 0. A symmetry boundary condition is used at the centerline with zero concentration at the outer surface. The diffusion coefficient D is given by the Arhennius expression

$$D(T) = D_o e^{-Q_i / RT} \qquad (2)$$

where D_o is the precoefficient, Q is the activation energy, R is the gas constant, and T is temperature. The values for D_o and Q are user specified and is different for the different fission products (see Reference 6).

Figure 2 shows the evolution of concentration profiles for Cs as calculated by a finite difference code and for steady state conditions at 1200 K. As can be seen, the SiC provides most of the diffusion resistance. This finite difference solution approach is used in a number of other fission product release codes. [7,8]

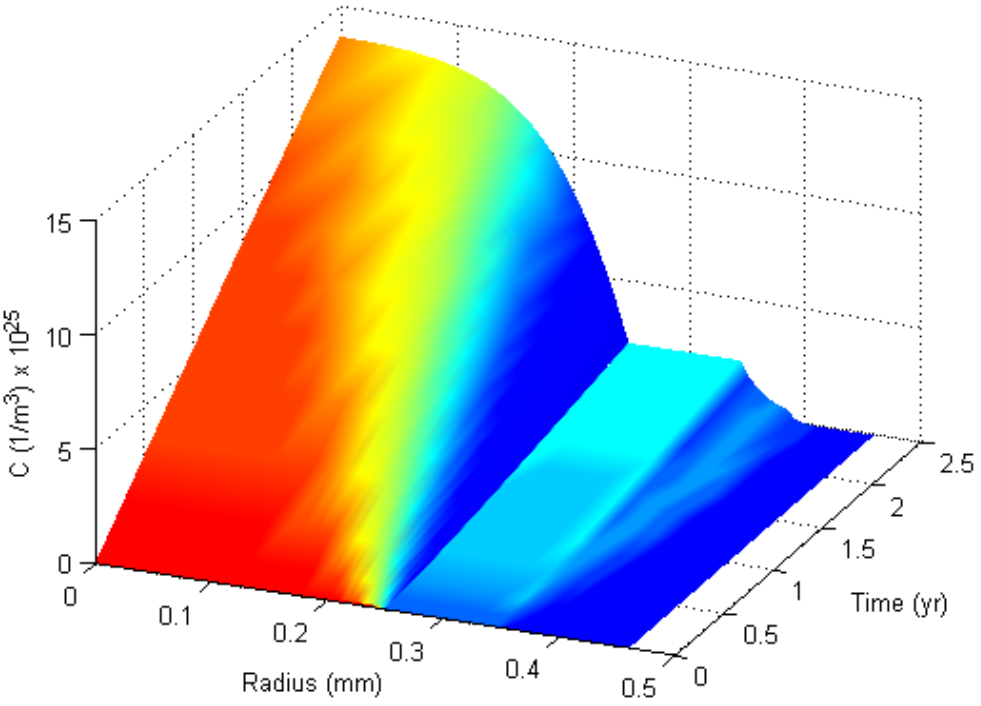

Figure 2. Cs concentration profile in TRISO fuel particle

Approximately 1.5% of the fission product inventory is transported to the buffer from the kernel via fission product recoil for the reference particle design shown in Figure 1. For some fission products, such as strontium, this exceeds the calculated release from the kernel via diffusional transport at fuel particle maximum operating temperatures. During an accident core heatup, when the diffusive transport increases with increasing fuel temperature, the recoil release still dominates the fission product transport for some fission products.

The recoil release from the kernel can be calculated by the following equation:[9]

$$f_r = \frac{3}{4}\frac{R}{a} - \frac{1}{16}\left(\frac{R}{a}\right)^3 \qquad (3)$$

where

R = Recoil length (m)

a = Kernel diameter (m)

The recoil distance for a heavy fission product is given as 7.7 μm by Nabielek,[10] 10 μm in Olander.[11] The recoil release to the buffer means that, on failure, the amount of fission products in the buffer layer available for release from the particle through the failed outer layers is greater than that calculated by diffusive release alone.

In a failed particle, fission products previoulsy released from the kernel are assumed to be instantly released through the failed coating layers. Further release from the kernel is then governed by diffusive transport in the kernel to the buffer layer and, during operation, by the recoil effect.

Of the gaseous fission products considered, only Xe and I have decay constants that are short enough to model fission product decay during the accident phase, and are in equilibrium during normal power operations.

III. B Accident conditions

For fission gas release from the kernels of failed particles, the general solution to the diffusion equation in an equivalent sphere after irradiation (no power, no further generation of nuclides) with time-dependent diffusion coefficient can be expressed as [5]

$$F(t) = 6\sum_{n=1}^{\infty}\int_0^t \exp\left[-\left(n^2\pi^2\int_0^{\mu} D'(t')dt' + \lambda\mu\right)\right] D'(\mu)d\mu \qquad (4)$$

where

$F(t)$ = Fractional release of fission product up to time t

$D'(t)$ = Reduced diffusion coefficient = $\frac{D(t)}{a^2}$

a = Radius of equivalent sphere (m)

The radius a was taken as the radius of a fuel grain in the original equivalent sphere model of fission product release. It can be taken as the kernel radius if an effective diffusion coefficient is used.

The short-time approximation ($\pi^2 D't \le 1$) to the general solution, eqn(4), for the case of a constant reduced diffusion coefficient D' can be given as

$$F(t) = \frac{3D'}{\lambda}\left(e^{-\lambda t} - 1 + \sqrt{\frac{\lambda}{D'}}erf\sqrt{\lambda t}\right) \qquad (5)$$

If it is further assumed that radioactive decay can be ignored over the time of interest, we get the short-time approximation of the Booth solution:

$$F(t) = 6\sqrt{\frac{D't}{\pi}} - 3D't \qquad (6)$$

For the present case the diffusion coefficient changes with time due to the fuel kernel temperature change with time. The product D't can in this case be taken as

$$\tau_D = \int_0^t \frac{D(t')}{a^2} dt' \qquad (7)$$

and τ_D substituted for *D't* in the above equations to get equations for the case of non-constant *D'*.

As mentioned previously, in a transient calculation it is desirable to use an analytical solution for the failed particles. Figures 3 and 4 show the comparison of the finite difference solutions (Eq. 1) with the approximate solutions (Eq. 6) for typical fission product releases at a constant temperature. It is evident that the analytical solutions provide reasonable agreement with the finite difference solution during the transient time of interest. From a computational point of view, the analytic solution is much easier to implement in combination with the particle failure rate.

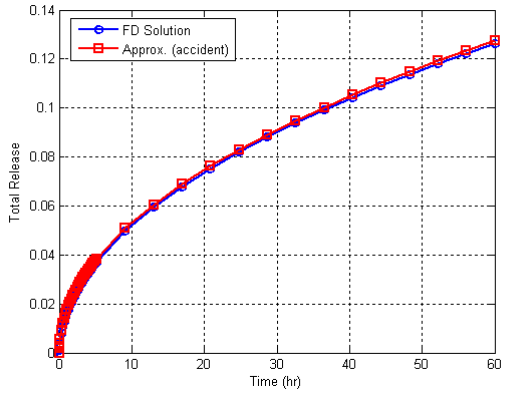

Figure 3. Ag release during accident

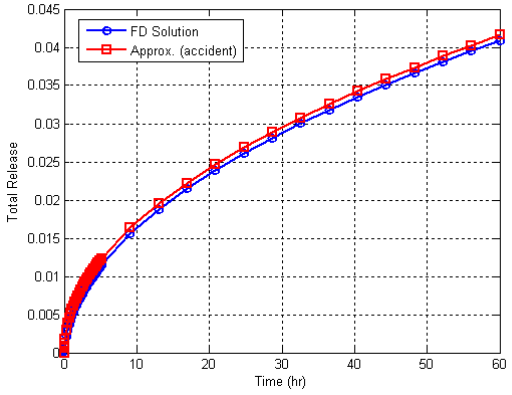

Figure 4. Cs release during accident

III.C Release from buffer and from buffer porosity

The release from the buffer to the matrix is assumed immediate, although some approximation could be made to simulate a delay. This is essentially release from the buffer graphite to the porosity, so an effective sphere model using a graphite grain size could be used. The buffer porosity is the main source of release on failure. It is usually considered as immediate, although it is probably actually vapor diffusion through the porosity. Further additional release is governed by the kernel release rates, which can be obtained via the approximate solutions to the equivalent sphere model given above.

III. D Matrix Contamination

The graphite matrix is contaminated by U metal during manufacture. Release from this contamination is not the same as from a bare kernel because of the high surface-to-volume ratio of the U contamination. The contamination release can be treated as immediate release from the U to the matrix, followed by diffusion through the matrix. Diffusion through the matrix provides a delay in the transport of fission products released by failed particles to the coolant. A release coefficient model is probably not adequate here, because the assumption of steady state in the matrix does not capture the delay effect. Probably a short-time approximation to the Booth solution in the matrix would be adequate to capture the delay.

III.E Dust

The graphite dust in a PBR results, in large part, from the pebbles abrading against each other and the graphite walls. During normal operation, this dust may be distributed throughout the primary system, and may contain fission products from both matrix contamination and that adsorbed during operation. This dust is also present, although to a lesser degree, in a PMR. The amount of fission products in the dust, the amount of dust generated, and the dust distribution must be provided as initial conditions for an accident calculation, either through input or via the steady-state MELCOR calculation that includes a dust generation model.

During an accident involving depressurization, dust deposited in the primary system may be resuspended by the increased helium flow. The liftoff depends on the flow velocities and size distribution of dust deposited. When resuspended, the dust will carry with it any fission products adsorbed during operation. A liftoff model is necessary for MELCOR to treat this phenomenon. Also, some treatment to distinguish the dust actually available for resuspension, versus dust that has been essentially plated onto the metal surfaces in areas of high temperature, will be needed. We suggest a parametric model based on helium mass flow and dust particle size as an approach for a MELCOR dust liftoff model. The fraction plated onto surfaces in the primary system and unavailable for liftoff will have to be a user input parameter, owing to lack of a model or data. [12]

A dust generation model is also needed. There does not seem at present to be a verified analytic model for dust generation. Thus, the information available is empirical evidence from existing reactor experience.[12] The present state of knowledge suggests that a parametric model, probably a constant generation rate during operation, would be best for MELCOR.

III. F Integration of releases from failures

During an accident scenario, the core temperature rises, resulting in increased particle failures from the increased temperatures and increased diffusional release from intact and failed particles due to increased diffusion rates. Particles fail at various times during the accident, so the total fission product release must account for these different failure times.

The releases from failed particles versus time are the integrated result of particle failures over the course of the accident. This can be expressed as a convolution integral. In addition, since the temperature is changing with time, the diffusion coefficients also change, so the release rate will also change due to changes in the diffusion rate versus time.

If we take the failure fraction at time *t* as $F_W(t)$, the rate of failure is $\frac{dF_W}{dt}$. The fission product release fraction from a particle at time t due to failure at time τ is $F_R(t-\tau)$. The total release fraction of a population at time *t* due to failures at time τ is then

$$\frac{dF_W(\tau)}{d\tau} F_R(t-\tau) d\tau$$

To get the total release fraction at time *t* due to all failures between a starting time 0 and time *t*, we need to integrate over all failure times τ: as in Nabielek and Verfondern:[13]

$$F_{tot}(t) = \int_0^t \frac{dF_W(\tau)}{d\tau} F_R(t-\tau) d\tau$$

It would be desirable from a computational standpoint to not have to perform this integral at each system timestep, since we do not usually keep the time histories of the failure fraction or release fraction during the computation. Although the integral can be divided into old and current timestep parts, the dependence of F_R on $(t-\tau)$ causes computational problems, in that some information from previous times must be kept. We would like to limit the amount of time history information that is kept in order to limit computational storage requirements. It is possible that no extraordinary methods will be necessary to limit the amount of time history data, since the release timesteps can be much longer than MELCOR system timesteps; typical accident timescales are 50-100 hrs.

It can be shown that, if the fission product release on failure was temporally expressed as a delta function, then the integral would be simply the failure fraction curve times the release amount. The major effect of slower releases is to shift (i.e., delay) the release fraction curve in time from the failure fraction curve.

The release rate for failures in one timestep is envisioned as being an initial burst release on failure, followed by a further diffusion-controlled release from the exposed kernels. The temperature dependence of the diffusive release rate is accounted for by using diffusion coefficients evaluated at the current temperature to calculate the incremental release for each timestep.

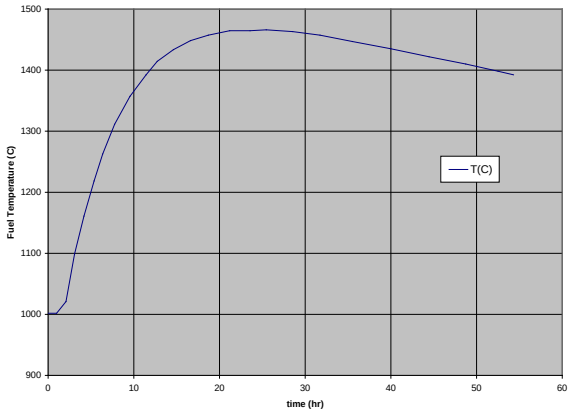

Figure 5. DLOFC temperature history

As mentioned previously, if the particle release rate were a delta function, the release curve would follow the failure fraction curve and the convolution integral collapses to a simple integral. Another approximation is to consider the kernel release rate only; the fission products are then considered to be either held up in the buffer, for intact particles, or to be released immediately to the coolant, for failed particles. The release fraction from the kernel in both cases is the same. For example, Figure 6 shows the DLOFC release fraction from a kernel failing at a reference time of 0 hr, and again for kernels at times 15 hr and 20 hr. The release fraction curves overlap after failure and merely start at different times.

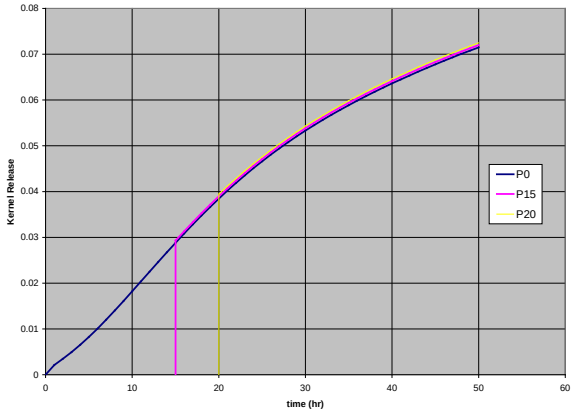

Figure 6. DLOFC release fraction for Kr from kernel for two example particles

Since the release fraction curve is the same for all kernels, the total release is then the release fraction times

the failure fraction. Again, the convolution integral is collapsed.

If the particle release to the coolant is assumed to be held up by the matrix, then the main effect is that the release curve is delayed by a constant time amount from the release curve determined assuming no matrix holdup. Using an effective time delay, rather than calculating the diffusional transport through the matrix, may be an adequate approximation for the matrix transport.

IV. SUMMARY

A fission product transport model is being developed for MELCOR which handles fission product distribution and release in HTGR cores. Both reactor normal power operation and transients can be calculated. The results of the normal power operation phase can be used to provide the initial conditions for the transient phase. The operation phase is calculated using a three-stage solution procedure to give the distribution of fission products in the fuel particles, matrix, and in the primary system. A finite difference diffusion model is used to calculate fission product distributions for the intact and failed particles, and the graphite matrix. A failure fraction curve is combined with the fission product release model via a convolution integral to account for particle failures over time during a transient.

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