Journal of Nuclear Materials 204 (1993) 180–187
North-Holland

journal of nuclear materials

The development of the Enpanne code to model failed fast reactor fuel pins

I.J. Ford [a] and M.A. Mignanelli [b]

[a] *Theoretical Studies Department, AEA Industrial Technology, B424.4, Harwell Laboratory, Didcot, Oxon, OX11 0RA, UK*
[b] *Reactor Chemistry Department, AEA Reactor Services, B540.1, Harwell Laboratory, Didcot, Oxon, OX11 0RA, UK*

The Enpanne code is being developed in order to describe the behaviour of fuel pins after failure. This is important if non-essential reactor shutdowns to remove failed pins are to be avoided. The models cover both physical and chemical aspects of the problem: on the physical side the opening of a cladding crack and the ingress of sodium into the pin need to be described while on the chemical side it is necessary to model the reaction between sodium and fuel to form sodium uranoplutonates. Progress in the various areas is described. Some preliminary calculations of the behaviour of a failed pin are presented and compared with post-irradiation examination. This has helped to identify aspects of the modelling which are inadequate at present and which are currently under development.

1. Introduction

In a commercial fast reactor some failures of fuel pins will most probably occur, particularly as their dwell times in the core increase and high burn-up of the fuel is achieved. There is a strong economic incentive to continue the operation of the reactor with the failed fuel in-situ until the planned shutdown or until the consequences of the failure become unacceptable. An understanding of the changes of the chemical constitution of the fuel following sodium ingress is essential for the assessment of the behaviour of a reactor core with failed fuel pins. The main effects of these changes are that fission product species and fuel particles could be released into the sodium coolant, causing circuit contamination, and fuel swelling due to the formation of sodium uranoplutonate could cause the extension of the original defect. In addition the thermophysical behaviour of the fuel pin will be modified after sodium ingress and reaction. The development of a code to model the thermophysical and chemical behaviour of failed fast reactor fuel pins is important in order to be able to quantify these effects for a failure at any particular burn-up and during internal storage. In addition such a model could provide information for the interpretation of the signals monitored by the failed fuel detection system.

The code which is being assembled by AEA Technology at Harwell has been named Enpanne (Experimental Numerical Procedure for the Analysis of Non-intact Nuclear fuel Elements) and is based on the TRAFIC code and the Alchemist suite of subroutines [1]. This paper describes the development of the various modules of the Enpanne code which include the depressurization of the pin on failure, the extent of sodium logging with respect to reactor power fluctuations, the changes in chemical constitution in the presence of sodium and crack extension due to fuel swelling. In addition, results are presented of preliminary calculations carried out to validate the chemical models in the code against an artificially defected fuel pin irradiated in the Prototype Fast Reactor (PFR).

2. Development of the Enpanne code

In the following sections the various models of the Enpanne code are described. As already discussed the code is based on TRAFIC and the Alchemist suite of subroutines. The latter set of models determine the chemical constitution and radial distribution of the fuel and fission product species for the intact pin, which will strongly influence the extent of fuel swelling and the release of species after pin failure. At this stage of the development, the effect of the chemical interac-

tions following sodium logging, in particular the sodium-fuel interaction, on the thermophysical behaviour of the pin is not modelled. The coupling of these phenomena is a key objective of the overall failed fuel modelling strategy. The various stages required in the Enpanne code for the calculation of the chemical behaviour of failed fuel pins are shown in fig. 1.

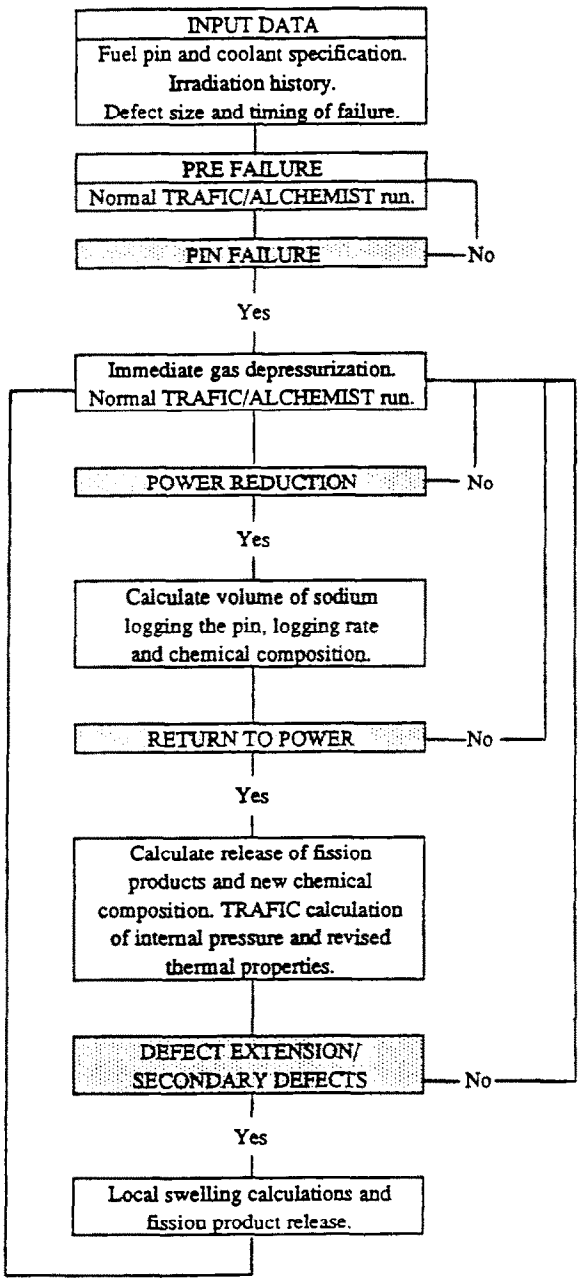

Fig. 1. Calculation of the chemical behaviour of failed fuel pins.

2.1. Pin depressurisation and sodium ingress models

The description of the entry of sodium into a failed fuel pin can be greatly simplified if it is assumed that sodium exchange through the cladding breach occurs rapidly, so that significant pressure differences between the inside and outside of the pin are not maintained for very long. Scoping calculations have indeed shown that the transfer is fast. Therefore, instead of calculating the sodium velocity through the breach driven by a particular pressure difference, an equilibrium pin pressure condition is assumed, and the corresponding level of sodium in the pin determined. Transients in the ebb and flow of sodium through the cladding rupture are ignored.

The consequences of this assumption as regards the pin mechanics are simple and can be handled easily. The inner stress boundary condition on the (assumed annular) pellet is altered at the instant of failure and the stresses in the fuel and clad subsequently allowed to adjust.

When the fuel pin fails, there will usually be an initial mismatch in pressure between the gas inside the pin and the sodium in the coolant channel. Depending on which pressure is larger, and assuming that the crack in the cladding is not plugged by a tightly fitting fuel pellet, there will either be an expulsion of gas or an ingress of sodium to equalise the pressures, as illustrated in fig. 2. Subsequent behaviour depends on the size of the crack. If the hole in the cladding is large, then the expulsion of gas can proceed after the pressures have nominally equilibrated: there would be a sufficient variation in hydraulic pressure across the cladding breach for sodium to leak into the pin. The level of sodium in the pin would consequently rise to the position of the failure. At the opposite extreme, a small hole would remain leak-proof since surface tension effects would oppose sodium entry.

Simple considerations, involving Laplace's equation for stability of the meniscus, can help to decide what size of hole will remain leak-proof after the initial pressure equilibration. Estimates of crack sizes, in section 2.4, suggest that initial breaches are small enough to resist ingress until changes occur in the pin operating conditions.

Following the initial equilibration, further exchanges of sodium and gas are driven by changes in pressure either inside or outside the pin. A decrease in reactor power results in a cooling and reduction in

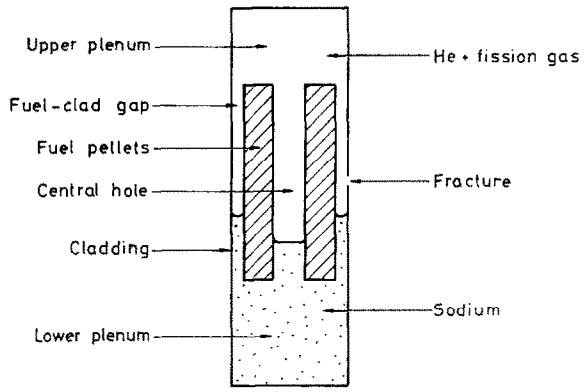

Fig. 2. A failed fuel pin (not to scale) into which sodium has been drawn.

pressure of the gas in the pin, and therefore leads to the drawing in of more sodium. A reduction in coolant flow rate both raises the pin temperatures and reduces the coolant pressure, leading to an expulsion of gas from the pin. The basic equilibrium condition observed at all times is

$$N = \int \frac{p \, dV}{kT}, \tag{1}$$

where N is the total number of gas atoms in the pin, p is the external pressure, kT is Boltzmann's constant multiplied by absolute temperature, and the integral is taken over all connected gas spaces in the pin, including the upper and lower plena, the fuel pellet central hole and the fuel-clad gap. When a change occurs in p or T, either the level of sodium in the pin or the number of gas atoms N changes. The one constraint is due to geometry. Sodium in the pin up to the level of the fracture position cannot be expelled: ingress up to this point is therefore irreversible.

The evolution of the level of sodium inside the pin is followed by calculating the volume occupied by the remaining gas within the pin, under the given temperature conditions. Gas may be expelled, if necessary, while the sodium level lies below the fracture. Example calculations demonstrate the operation of this model. Fig. 3 shows the evolution of the in-pin sodium level for a case where a pin is irradiated to a burn-up of 16% at a linear power of about 430 W cm^{-1}. The fissile column is of length 1 m and the upper and lower plena have lengths of 15 cm and 1 m, respectively. The inlet and outlet temperatures at full power are 668 K and approximately 900 K, respectively. Following an initial irradiation period of length 4×10^7 s, shutdowns occur at intervals of 5×10^6 s. A failure of the cladding at the mid-point of the fuel column is supposed to occur after 3×10^7 s. The initial gas pressure inside the pin is 0.1 MPa while the coolant pressure at failure is about 0.6 MPa, but the initial period of intact operation generates sufficient fission gas within the pin to exceed the external pressure, so that at failure, gas is expelled from the pin. A failure before the pin pressure had exceeded the coolant pressure would have resulted in immediate sodium ingress.

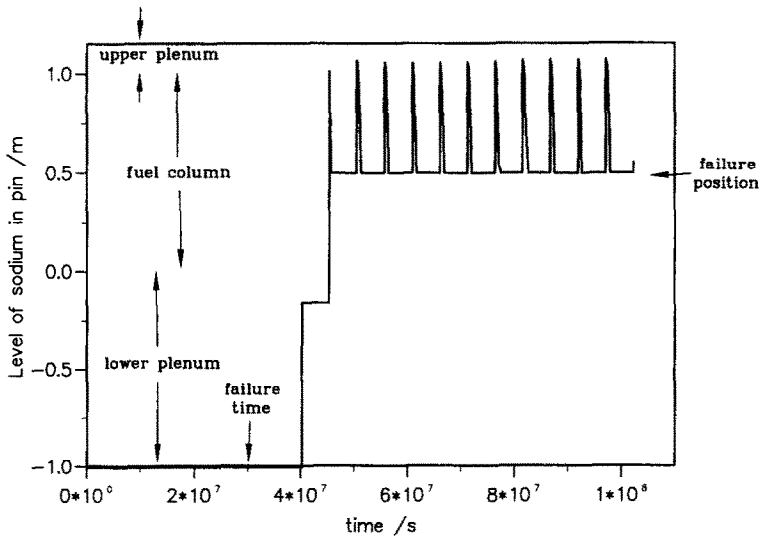

Fig. 3. Rise of sodium inside a failed fuel pin due to power cycling.

At each shutdown, the power is reduced to zero and the sodium inlet pressure also falls to about 0.16 MPa, reflecting a reduction in coolant flow. Initially, the introduced sodium collects in the lower plenum: two shutdowns are required for the level to reach the fissile column. Once the level reaches the lowest fuel pellet, the rate of rise is accelerated since the cross-sectional area of available space across the pin is much lower than within the plena, as shown in fig. 2. The sodium will rise up the central hole, and penetrate into the fuel-clad gap if it is open. Once the level passes the failure position, the subsequent behaviour is reversible, rising and falling at each shutdown and return to power.

Concerning the fuel-pin chemistry, the level of sodium within the pin provides a boundary between a region where the chemical equilibria should be determined with the inclusion of sodium, and a region where sodium is excluded from the reactions. The interface moves, changing the local chemical environment as it does. However, this simple picture ignores the effects of capillary action, whereby the level of sodium in the narrow fuel-clad gap could differ from that in the relatively wide central hole, as indicated in fig. 2. Some initial calculations suggest that capillary action is strong and that sodium rises rapidly up a narrow fuel-clad gap as soon as the level in the pin reaches the bottom of the fuel column.

2.2. Sodium-fuel reaction model

Following sodium logging of the pin during a period of power reduction, the reaction of the sodium with the fuel to form sodium uranoplutonate can occur in those regions of the fuel column determined by the threshold equilibrium thermodynamic conditions [2]. This reaction along with the reactions between the sodium and the phases formed in the fuel-clad gap prior to failure are modelled in the code using the equilibrium chemistry routines. The kinetics of the sodium-fuel reaction will be determined using algorithms derived from the experimental programme carried out at Harwell [3]. The rate of reaction at a particular temperature, T, can be expressed by the equation,

$$\mathrm{Vol_s} = A(1 - \exp(-kt)), \tag{2}$$

where $\mathrm{Vol_s}$ is the percent volume increase, k the rate constant, t the time in hours and A a constant equal to the maximum volume increase. The latter value can be derived from the threshold plutonium valency, $\mathrm{Pu_{val}}$; the theoretical densities of the fuel oxides and the sodium uranoplutonate [4]; and the reaction equation,

$$3(y-z)\mathrm{Na(l)} + (2+y)\mathrm{U_{1-x}Pu_xO_{2-z}(s)} \rightarrow (y-z)\mathrm{Na_3U_{1-x}Pu_xO_4(s)} + (2+z)\mathrm{U_{1-x}Pu_xO_{2-y}(s)},$$

where (s) and (l) refer to the solid and liquid phase respectively.

The value of y is set by the equation,

$$y = 2 - (4(1-x) + \mathrm{Pu_{val}}x)/2. \tag{3}$$

Although the influence of burn-up on the kinetics of the reaction also needs to be included, there are only limited data available. A simple linear dependency will be used as an initial approximation.

Following failure, further fission gas release from the pin will still occur and the generation and radial migration of the other fission products and oxygen will continue.

2.3. Release of fission products and fuel particles

On cycling between low and high power, the sodium content of the fuel pin will be continually changing. Fission product species calculated to be present initially in the fuel-clad gap that are soluble or miscible in sodium, typically phases of the volatile fission products Cs, Te and I, will be expelled into the main coolant channel during this process. The rate of release of these highly soluble species will depend, therefore, on the logging behaviour rather than chemical effects. Subsequent to the release, the chemical composition of the fuel pin will be calculated using the revised inventory of the fission products.

As the initial defect is enlarged and the surface area of exposed fuel is increased, or if a secondary defect is formed at a different axial position of the pin, sodium entry may not require power cycling. The sodium-fuel reaction, either at the enlarged defect site or along the flow path within the pin, will occur during normal power operation, which will also involve the excess oxygen available in the flowing sodium. The growth of the reaction product layers will then be determined by the main coolant flow rate, the fuel surface temperature and the oxygen content of the sodium. The modelling of the extent of the reaction in the code will be based on the results from small-scale loop experiments carried out at Harwell [5]. Similarly, the modelling of the erosion of fuel particles from the fuel surface, which will also be incorporated in the code, will be estimated from data derived from in-pile tests [6]. The release of fission products under these conditions will

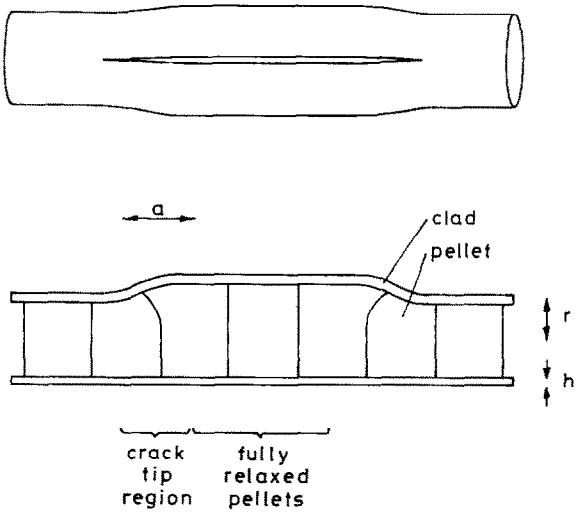

Fig. 4. Geometry of axially cracked fuel pin driven by PCMI.

be determined by the radial transport processes in the fuel and fuel-clad gap. The inventory in contact with flowing sodium will be assumed to be removed from the pin.

2.4. Crack formation and extension

The initial formation of an axial crack in the clad driven by stresses in the fuel pellets can be modelled using fracture mechanics. The main points of interest are (a) the size of crack formed initially, and (b) the likelihood of crack extension driven by swelling caused by chemical reactions following sodium ingress. A simple model of crack geometry and pellet stress relaxation has been formulated. The situation is shown in fig. 4. Clad and fuel deformation are taken to be elastic. The length of each crack tip is a and the radial displacement of the clad wall is taken to be

$$w(z,\theta)=\begin{cases}0 & \text{if } z<0,\\ \delta_{\max}\sin^2\left(\dfrac{\pi z}{2a}\right)(\theta/\pi)^2 & \text{if } 0\le z\le a\\ \delta_{\max} & \text{if } z>a,\end{cases} \quad (4)$$

where $\delta_{\max}$ is the maximum displacement and z and θ are cylindrical polar coordinates with the crack lying along $\theta=\pm\pi$. The loading pressure on the clad is taken to be

$$p(z,\theta)=\begin{cases}0 & \text{if } z<0,\\ p_0-B\Delta V(z)/V & \text{if } 0\le z\le a,\\ 0 & \text{if } z>a,\end{cases} \quad (5)$$

where p_0 is the pin pressure at failure, B is the bulk modulus and $\Delta V(z)/V$ is the fractional volume expansion of the fuel pellet made possible by clad deformation at position z. Using the loading and deformation patterns, it can be shown that $\delta_{\max}=3rp_0/(2B)$ with r the pin radius. The maximum width of the crack is taken to be approximately $u=4\bar{p}ahM/(rE)$ with $\bar{p}$ a representative constant pressure acting on the crack, h is the clad wall thickness and E is Young's modulus. $M=(1+1.61a^2/(rh))^{1/2}$ is an enhancement factor due to the clyindrical geometry. Using $\bar{p}\approx p_0/2$ and $u=\pi\delta_{\max}$ we have

$$Ma=3\pi hE/4B, \quad (6)$$

which is a quadratic equation for a. For $r=4$ mm, $h=0.5$ mm, $E\approx B$ we have $a\approx 1$ mm and the crack width is about 2 μm. A crack with these approximate dimensions just relaxes the pellet stresses.

Whether the crack continues to run is determined by energy release and dissipation. The elastic energy released by the pellet is approximately $U_p=\pi ra\delta_{\max}p_0/12=\pi r^2ap_0^2/8B$ and the energy release by the clad is at most $U_c=2\pi rp_0^2r^2a/(2hE)$. The crack driving force is approximately

$$F_d\approx\frac{U_p+U_c}{a}=\pi r^2p_0^2\left(\frac{1}{8B}+\frac{r}{hE}\right). \quad (7)$$

The crack resistive force is not less than $F_r=K^2h/E$ where K is the clad fracture toughness so that the ratio of driving to resistive forces on the crack is given by

$$F_d/F_c<\pi r^2p_0^2\left(\frac{E}{b}+\frac{r}{h}\right)/K^2h, \quad (8)$$

which is about 0.03 for typical parameters $p_0=20$ MPa and $K=100$ MPa m$^{1/2}$. This means that the clad must be severely embrittled (low K) for the crack to continue to run. It is more likely that the crack will arrest when it has propagated far enough to relax the stresses at the failure point and has met clad which is relatively undamaged. An approximate resulting crack size is 2 mm according to the preceding argument. Once the fuel reacts with the sodium and begins to swell, it is possible that the crack may be extended further but this aspect of the model is still to be developed.

3. Comparison of the chemical model with PIE results

The purpose of these calculations was to test the thermodynamic models for the sodium- fuel reaction in the Enpanne code against the defect pin test HSH in

the PFR. As the coupling between the chemistry calculations and the thermophysical behaviour of the pin has not been developed yet, the code cannot be used to model the changes in the fuel temperatures due to the formation of a peripheral band of sodium uranoplutonate.

For the irradiation conditions experienced by cluster HSH, which involved considerable power cycling, complete logging by sodium of the fuel-clad gap of the fuel and upper breeder columns would be expected.

3.1. Details of the defect pin

Cluster HSH contained one artificially defected pin and 18 standard reference fuel pins. The pins contained standard length fuel columns of annular pellets of composition $U_{0.69}Pu_{0.31}O_{1.98}$ and were clad in 20% cold worked M316. The external and internal diameters of the pellets and clad were 4.978/1.829 mm and 5.842/5.080 mm, respectively. The defect in the pin was a hole 0.25 mm in diameter located at the top of the lower plenum. The cluster was irradiated in PFR to an equivalent of 201.3 effective full power days reaching a maximum burn-up of 5.8% heavy atoms. After removal from the reactor, the cluster was stored for 5.5 years in the primary sodium prior to destructive examination. An intact pin and the defect pin were then examined by non-destructive and destructive techniques.

3.2. Results from the PIE

The examination of the intact pin indicated that the fuel had behaved as expected for the irradiation conditions experienced [7]. No evidence of columnar grain formation was found indicating that the fuel temperatures were low. The examination of the defected pin showed that sodium had logged the entire length of the fuel column and entered into the upper axial breeder section during the period in the reactor. Extensive reaction had occurred between the peripheral regions of the fuel and the liquid sodium coolant resulting in a band of reaction product, sodium uranoplutonate, up to 450 μm thick along the length of the fuel column. This differed from the morphology of the interaction of the sodium with the upper axial breeder which was intergranular with the reaction front penetrating the pellets to a depth of up to 250 μm . The low thermal conductivity of the reaction product had resulted in high fuel temperatures and extensive columnar grain growth in the region of the core centre line. The fuel was rendered sufficiently plastic that it was capable of accommodating the volume increase due to the sodium-fuel reaction by the contraction or closure of

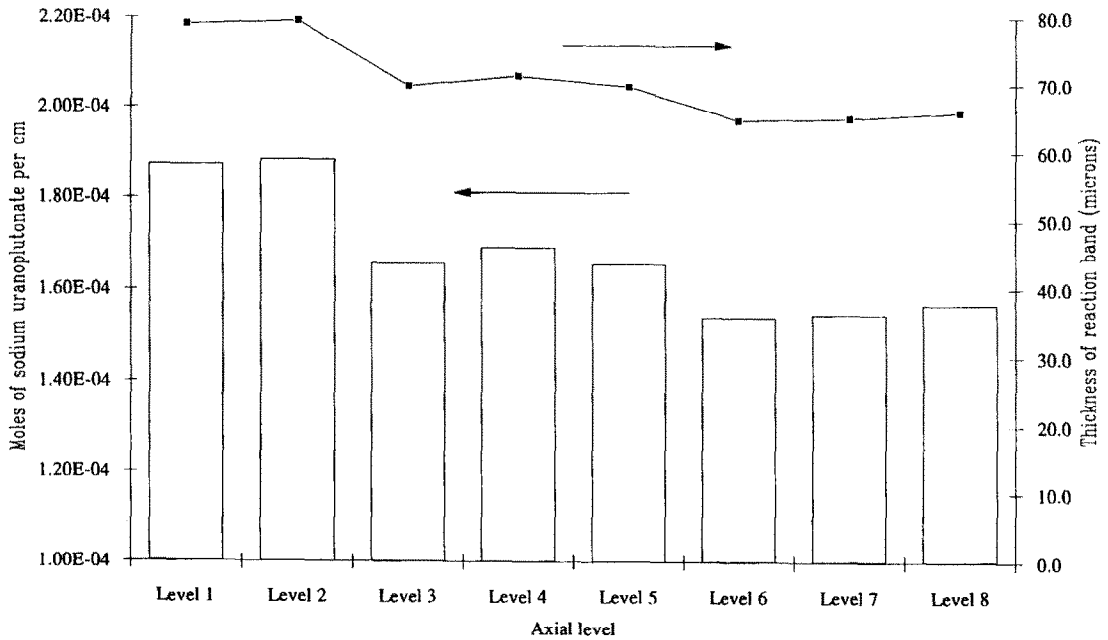

Fig. 5. Variation of the amount of sodium uranoplutonate formed with axial position.

the central hole. No clad strains were detected and propagation of the defect had not occurred. An analysis of the gamma-scan profiles for ^{137}Cs indicated a 47% loss from the fuel column.

3.3. Comparison with calculated results

Enpanne calculations were performed assuming free access of the sodium within the fuel- clad gap. This was achieved by adding as an input quantity an excess of sodium to the fuel prior to the determination of the equilibrium composition. The results from the calculation of the sodium-fuel reaction expressed as the maximum number of moles of sodium uranoplutonate formed along the fuel column are shown in fig. 5. The results show that the amounts formed are fairly uniform along the pin, with slightly increased amounts in the lower axial region where the centre fuel temperatures are lower. As expected, sodium uranoplutonate was not formed in the inner fuel rings of the axial levels centred about the core mid-plane.

The only comparison that can be made between the calculations and the PIE results is to compare the observed band width of the reaction product with the calculated band width derived from the amount of product formed, assuming that the internal diameter of the clad has not changed. The formation of peripheral bands of reaction product is made possible either by the radial migration of oxygen in the fuel during power operation, or by the oxygen dissolved in the sodium reacting with the fuel surface. The calculated values are also shown in fig. 5; the reaction product band width is a maximum of 80 μm. The results from the PIE indicate that the reaction product band width in the upper and lower regions of the fuel column is a maximum of 450 μm with typical values of 350 μm. In the case of samples taken from the central regions of the fuel pin, the band width is approximately 225 μm. Radial sections of the pin taken from these regions clearly show that sodium has completely penetrated the grain boundaries of the fuel, including the zone of columnar grains. This indicates that some sodium-fuel reaction occurred either during periods of low power operation or during in-vessel storage.

It would appear that the agreement between the Enpanne calculations and the PIE results for the defect pin in HSH is poor. However, the contribution of the oxygen dissolved in the sodium, in excess of the threshold concentration, to the formation of sodium uranoplutonate is not yet considered in the code. The theoretical maximum band width that could form based on the oxygen available in the fuel alone, using a threshold plutonium valency of 3.48, is 80 μm. Taking into account the effect of burn-up, this value could rise to a maximum value of about 100 μm. Therefore, the formation of sodium uranoplutonate in the defect pin is due mainly to the reaction with the oxygen dissolved in the sodium. These data are in agreement with results from small-scale sodium loop experiments carried out at Harwell [5], involving a study of the sodium-fuel reaction in flowing sodium of controlled oxygen content. The experiments confirmed that the oxygen dissolved in the sodium, at concentration levels as low as 2 wt ppm, can react with fuel pellets and in HSH this apparently has resulted in up to twice the theoretical swelling expected if only the fuel is considered.

4. Further development of the Enpanne code

The influence of these chemical changes on the thermophysical behaviour of the fuel pin needs to be established. The effect of the presence of a peripheral band of sodium uranoplutonate, which has a lower thermal conductivity than the fuel [4], on the heat transfer properties of the fuel could be determined within the TRAFIC part of the code. In addition, the increase in volume on forming the reaction product will cause an additional internal stress on the clad and, in particular, at the site of the initial defect. The TRAFIC code is ideally suited to handle the effect of an increase in volume of an outer band of an unspecified phase. The thermal and mechanical properties of sodium uranate, that are required for these calculations, have recently been assessed [4] and could be applied to the analogous uranoplutonate phase. The feedback of these data to TRAFIC and the processing of the data will require significant code development. In addition, further development of the chemical models to account for the contribution of the oxygen dissolved in the sodium to the formation of sodium uranoplutonate is required, particularly for internal storage situations.

On the mechanical side, the extension of an existing defect by the chemically-induced swelling of the fuel is an area which needs to be addressed. It is expected that a simplified approach within fracture mechanics will be adequate. The effect of additional failures on the logging behaviour also needs to be addressed. Slight extension of the existing models describing the ingress of sodium into the pin will be necessary to take this into account.

5. Conclusions

The Enpanne code is being developed to provide models to describe the many thermophysical and chemical effects that would occur during the failure of a fast reactor fuel pin. The code is based on the TRAFIC code and the Alchemist suite of subroutines. The phenomena that need to be modelled include the rate and extent of sodium logging, the sodium-fuel reaction, the release of fission products and fuel particles and the further deterioration of the pin. An important consideration in the development of the model is the treatment of periods of power reduction or shutdown after pin failure followed by the return to full power. During this phase, sodium ingress and hence sodium-fuel reaction will occur and this will influence the subsequent thermophysical behaviour of the pin. The effect of power cycling is cumulative in the sense that not all the sodium taken in by the pin on a reduction in power is expelled on returning to power, until sodium has filled the pin up to the failure site.

Although some of the stages will require new routines to be written, several of the consequences arising from the failure could be handled by existing routines within the TRAFIC code or the Alchemist routines. Preliminary calculations have been performed to model the chemical behaviour of the defect pin in the cluster HSH using the Enpanne code. The agreement between the calculations and the PIE results is poor. We believe that this is due to a lack of a model in the code to represent the chemical interaction between the excess oxygen dissolved in the sodium coolant and the fuel; the process that is responsible for the formation of most of the observed band of the sodium uranoplutonate phase in the defect pin.

Acknowledgement

This work was funded by the U.K. Department of Trade and Industry.

References

[1] M.A. Mignanelli and R. Thetford, in these Proceedings (Int. Symp. on Fuels for Liquid Metal Reactors, Chicago, IL, 1992), J. Nucl. Mater. 204 (1993) 173.
[2] M.A. Mignanelli and P.E. Potter, Proc. Int. Conf. on Science and Technology of Fast Reactor Safety, Guernsey, 12–16 May 1986, vol. 1, p. 53.
[3] M.A. Mignanelli and P.E. Potter, Harwell report, AERE-R 10991 (1983).
[4] H. Kleykamp, KfK Report no. 4701 (1990).
[5] D.J. Brown and M.A. Mignanelli, AEA Technology, unpublished work.
[6] M. Tetenbaum, EPRI report NP-1609.
[7] S. Tod, I.G. Clark and E.J. Fulton, AEA Technology, unpublished work.