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A Mechanistic Swelling Model of TRU Metal Dispersion Fuel for HYPER

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Abstract

An in-reactor performance computer code for the blanket fuel is being developed in the conceptual design stage of blanket fuel for HYPER. The present deveolpment status of the DIMAC code is in the second step which emphasizes on fuel swelling calculation particularly. In this paper, mechanistic swelling models were developed and the models were installed into the DIMAC program. The preliminary calculation results for (TRU-Zr)-Zr dispersion fuel predicted by DIMAC were compared with those of silicide dispersion fuel predicted by DIFAIR. It appeared that the swelling levels for (TRU-Zr)-Zr dispersion fuel were relatively higher than those of silicide fuel, and the dispersion fuel performance may be limited by swelling. Therefore, some experimental tests including inpile and outpile experiments are need for clarifing the integrity and the material properties of the inter-metallic fuel core.

1. Introduction

Either TRU-Zr metal alloy or (TRU-Zr)-Zr dispersion fuel is considered as a blanket fuel for HYPER (HYbrid Power Extraction Reactor). Recently, renewed interest in dispersion metallic fuel for ADS(Accelerator Driven System) has arisen[1] in the USA, France, Japan and Korea. In the case of dispersion fuel, the particles of TRU-Zr metal alloy are dispersed in a Zr matrix. A blanket rod is made of sealed tubing containing actinide fuel slug in columns. The blanket-fuel cladding material is ferritic-martensitic steel.

The rod deformation analysis in fuel rod design is essential to assure adequate fuel performance and integrity under irradiation conditions. This paper represents an

attempt to develop a swelling model to predict the deformation of the (TRU-Zr)-Zr dispersion fuel rod according to its power history. Therefore, modeling efforts for fuel swelling have been made and these models are installed in DIMAC (DIspersion Metallic fuel performance Analysis Code).

The deveolpment scheme of the DIMAC code consists of three steps : basic and main programming and temperature calculation scheme, rod deformation modelling, and thermo-mechanical modelling. The present status of the code deveolpment is in the second step which emphasizes fuel swelling calculation.

To develop a physical model, it is necessary to analyze the irradiation behavior of (TRU-Zr)-Zr dispersion and TRU-Zr alloy fuels. As described in section 2, one of the important irradiation performance characteristics of a TRU-Zr fuel rod in ADS/FBRs is its diameteral increase resulting from fuel core swelling. By analyzing the irradiation behavior in the literature on experimental data for TRU-Zr and U-Pu-Zr fuels, a analytical swelling model is derived. In Section 3, the key models that are significant in controlling fuel swelling are described. Section 4 describes some benchmark calculations to evaluate the validity of the DIMAC code. Conclusions and recommendations are given in the final section.

2. Physical Processes for In-Reactor Performance of a Dispersion Fuel Rod

In U-Pu-Zr, and U-Pu-Fs alloy fuels for FBRs, some contributing mechanisms for the swelling of the fuel during irradiation have already been identified. Also, in the silicide dispersion fuel of a research reactor, some contributing mechanisms have already been identified[2] for the swelling of the dispersion fuel during irradiation. However, there are so far no irradiation data for the dispersion fuel of (TRU-Zr)-Zr, it is expected that the contributing mechanisms similar to silicide dispersion fuel can be applied for (TRU-Zr)-Zr fuel. The main unknown is the reaction mechanism of the interfacial layer between (TRU-Zr) particles and the Zr matrix. Fortunately, the irradiation data of Pu-Zr is very important for TRU-Zr alloy fuel. One of the major component of TRU is Pu (> 90% Pu in TRU) which has already been well characterized[3,4]. Despite a lack of available (TRU-Zr)-Zr fuel characteristics, existing irradiation data and experience for silicide dispersion and metal alloy fuels have led to the following physical processes for (TRU-Zr)-Zr dispersion fuel.

(i) In reactor irradiation experiments with inter-metallic uranium compounds, Hofman(1) showed that an enormous increase in gas bubble growth occurs when a compound becomes amorphous during irradiation. This accelerated swelling

phenomenon occurs only in compounds that undergo a crystalline-to-amorphous transformation, and that it is a manifestation of radiation enhancement of diffusion and plastic flow in amorphous solids[3].

(ii) (TRU-Zr)-Zr dispersion fuel is not thermodynamically *stable*. The type and amount of an interfacial layer formed obviously varies with concentration and temperature, and can be predicted from diffusion behavior. Constituent migration[5] occurs around the interfacial layer between TRU-Zr fuel particles and the Zirconium matrix, the Zirconium having reacted with particle surfaces and also penetrated along the networks of grain boundaries to attack sub-grains throughout the TRU-Zr.

(iii) Total swelling is comprised of three major components, a volume change due to transformation to a higher Zirconium phase as a result of TRU burnup, a volume increase due to the accumulation of non-gaseous fission products, and a volume increase due to fission gas accumulation. The volume change due to TRU burnup will be calculated using the most current TRU- Zirconium equilibrium phase diagram and measured densities of the phases involved. The amount of non-gaseous fission products was determined with the aid of published fission yield data. These contributions to volume change were estimated by evaluating their solubility in fuel and their tendency to form compounds with each other and with TRU and Zirconium. These two contributions to swelling, i.e., phase transformation and non-gaseous fission products were combined and yielded as a linear function of fission density. The largest component of fuel swelling is due to the formation of fission gas bubbles.

(iv) The component of fuel swelling due to the formation of fission gas bubbles is not linear with fission density at lower swelling values but tends to a linear behavior when swelling reaches higher levels. The non-linear behavior is caused by the capability of smaller bubbles to contain more gas atoms according to the equilibrium between the gas pressure in the bubble and the surface tension of the fuel. The apparent fission rate dependency is due to a delay in development of larger bubbles when the fission rate increases[6].

(v) In a dispersion fuel, the most important driving force for deformation is believed to be fuel core swelling. When compared to commercial UO_2 fuels, the discharge burnup of blanket fuel for ADS is very high (up to about 20at% BU), therefore, fuel swelling can also be high enough to cause large permanent plastic deformation.

From the foregoing description, the volume changes in irradiated (TRU-Zr)-Zr were shown to be dependent on temperature and fission rate. The quantitative amount of swelling for (TRU-Zr)-Zr fuel can be estimated as follows by considering temperature, fission rate, solid fission product build-up and gas bubble behavior.

$$\left\{\frac{\Delta V}{V}\right\}_{ts} = \left\{\frac{\Delta V}{V}\right\}_{gb} + \left\{\frac{\Delta V}{V}\right\}_{sp} + \left\{\frac{\Delta V}{V}\right\}_{il}$$

where,

$\left\{\frac{\Delta V}{V}\right\}_{ts}$: the total fractional volume change due to the swelling for (TRU-Zr)-Zr dispersion fuel

$\left\{\frac{\Delta V}{V}\right\}_{gb}$: the fractional volume change due to the accumulation of gas bubble nucleation, growth and coalescence

$\left\{\frac{\Delta V}{V}\right\}_{sp}$: the fractional volume changes due to transformation to a higher zirconium phase as a result of TRU depletion as burnup increases and the accumulation of solid fission product swelling

$\left\{\frac{\Delta V}{V}\right\}_{il}$: the fractional volume change due to a thermal-chemical reaction between TRU-Zr fuel particles and the Zr matrix .

3. Swelling Model Description

3.1 Modelling of the Volume Change due to the Accumulation of Solid Fission Products

The solid fission products introduced into the lattice of fuel particles lead to volumetric swelling. The swelling of the fuel core due to the formation of solid fission products is caused by incorporation of the products in the solid lattice and is, therefore, proportional to the number of fissions which have taken place in the fuel core. Also, the volume change due to transformation to a higher zirconium phase as a result of TRU bumup is proportional to the fission rate. Therefore, these two contributions to swelling are combined and yield a linear function of fission density.

The fractional volume change for TRU-Zr particles, f_p due to the formation of solid fission products is given by :

$$f_{tp} = C \cdot 10^{-3} \cdot B_a \qquad (1)$$

where, C is a coefficient ranging from 1.26 to 1.86 which is derived from the experimental data[7,8], and B_a is the atomic % burnup. In the DIMAC code, C =1.26 is used. Not only is the overall density for solid fission products under fast fission condition slightly higher than that under thermal fission condition, but also the densitiy of soild fission products due to the fission of Pu is slightly higher than those due to the fission of U.

In order to model the swelling behavior of (TRU-Zr)-Zr fuel, it is assumed that the initial pores only existed at the boundary area between (TRU-Zr) particles and the Zr matrix. For the unit volume of the fuel core, the fractional volume change due to the formation of solid fission products is given by

$$\left(\frac{\Delta V_{uc}}{V_{uc}} \right)_{tp} = \left\{ \left(\frac{WT_{zr}}{\partial_{zr}} \right) + V_{ip} + (V_{tp} N_{tp})(1 + f_{tp}) \right\} - 1 \qquad (2)$$

where,

$V_{uc} = V_{tp} \cdot N_{tp} + \dfrac{WT_{zr}}{d_{zr}} + V_{ip} = 1$, the unit volume of (TRU-Zr)-Zr fuel

$WT_{zr} = d_{fc} \cdot F_{zr}$, the Zr-matrix weight per unit volume of (TRU-Zr)-Zr fuel

d_{fc} = the density of (TRU-Zr)-Zr fuel

d_{fp} = the density of (TRU-Zr)

d_{zr} = the density of Zr

F_{zr} = the weight fraction of the Zr matrix in the (TRU-Zr)-Zr fuel core

V_{tp} = the volume of a (TRU-Zr) particle

$V_{ip} = 1 - (V_{tp} \cdot N_{tp} + VT_{zr})$, the initial pore volume per unit volume of the fuel

$N_{tp} = d_{fc} \cdot F_{tp} / M_{tp}$, the number of TRU-Zr particles per unit volume of fuel

VT_{zr} = the volume of Zr-matrix per unit fuel core volume, WT_{zr} / d_{zr}

F_{tp} = the weight fraction of (TRU-Zr) in (TRU-Zr)-Zr fuel

M_{tp} = the weight of a (TRU-Zr) particle.

3.2 Diffusion Behavior of the (TRU-Zr)-Zr Dispersion Fuel

One of the swelling mechanisms for dispersion fuel is associated with a reaction zone between TRU-Zr fuel particles and the Zirconium matrix. Pu-20wt%Zr dispersion fuel as annealed at 873K for 1hr, and the reaction zone between the Pu matrix and Zr particles was observed by a Scanning Electron Microscope. The phases[9] of some regions near the reaction zone between the Pu matrix and Zr particles were identified using an energy dispersive X-ray(EDX) detector installed in the SEM. Fig. 1 shows the reaction zone between the Pu matrix and Zr particles in

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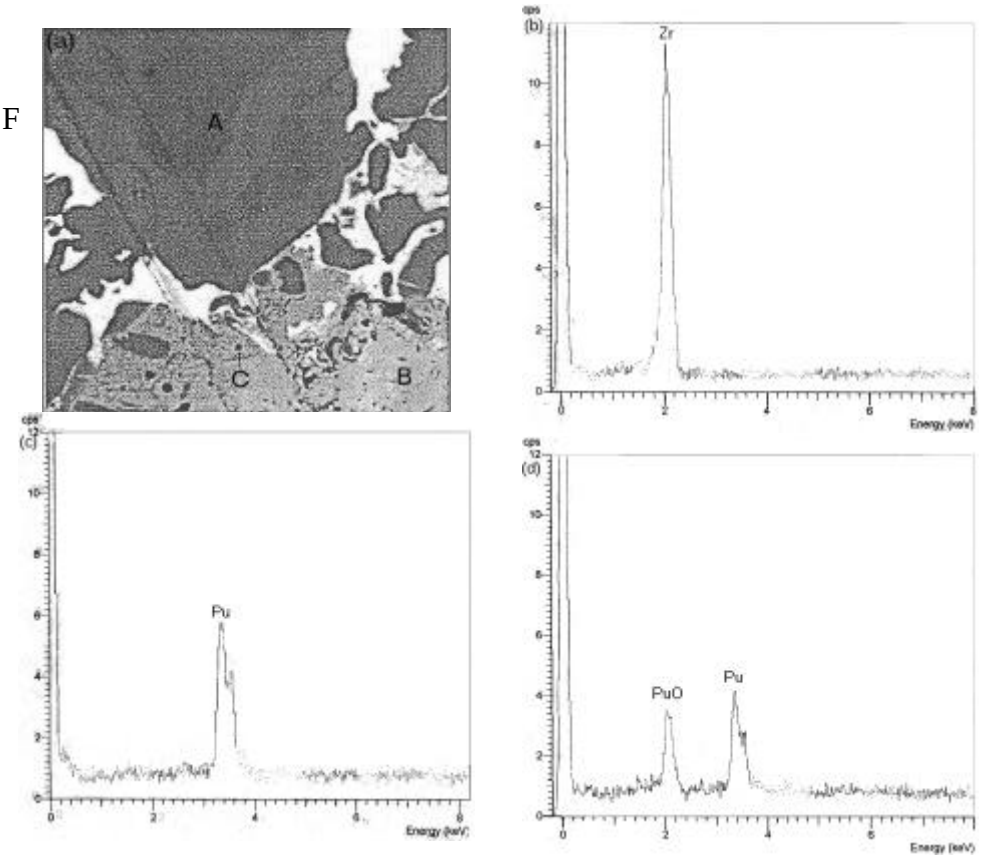

Fig. 1. SEM micrograph(a) showing the reaction zone between the Pu matrix and Zr particles, and EDS spectrums(b,c,d) at each spots in the alloy annealed at 873 for 1hr.

Pu-20Zr alloy annealed at 873 K for 1hr. There was no interfacial layer within the reaction zone between the Pu matrix and Zr particles. The dark phase, A, was identified as pure zirconium, and the grey phase, B, was pure plutonium. The tiny dark phase, C, was identified as impurities such as plutonium oxide, which seem to be contaminated during polishing. Comparing the Pu-20Zr sample with the (TRU-Zr)-Zr sample, it is inferred that both samples have similar results in the interfacial layer. Therefore, there is assumed to be no interfacial layer in (TRU-Zr)-Zr dispersion fuel, and it is postulated that there is no density variation near the reaction zone between (TRU-Zr) particles and the Zr matrix. Therefore, the density change of the interfacial layer in (TRU-Zr)-Zr dispersion fuel is neglected in present swelling mechanisms.

3.3 The Modelling of Volume Change due to the Accumulation of Gas Bubble Nucleation

Gaseous fission products, primarily Xe and Kr, are an even more important source of swelling. These atoms agglomerate into gas bubbles which have volumes many times greater than the fuel atoms from which they originated. In order to estimate the volume change due to the accumulation of gas bubble nucleation, it is necessary to calculate the bubble size distribution formed by the gas atoms produced within the fuel particles. The procedure for calculating the bubble size distribution on grain boundaries consists of dividing the bubbles into equal size ranges on a logarithmic scale and averaging their properties over the ranges as demonstrated by Li et al. [10].

The bubble sizes are defined according to the number of gas atoms. The gas is assumed to obey a Van der Waals equation, where the Van der Waals constant is a function of temperature and pressure. n_i is the number of gas atoms in the bubble with radius r_i, which is given by the following expression [11].

$$r_i = \left\{ \left(\frac{3kT}{8\pi g} \right) n_i \right\}^{1/2} . \tag{3}$$

Hence, the mean radius, $\overline{r_i}$, at a region which ranges from r_i to r_{i+1} is given by

$$\overline{r_i} = \left\{ \left(\frac{3kT}{8\pi g} \right) \left(\frac{n_i + n_{i+1}}{2} \right) \right\}^{1/2} , \tag{4}$$

where, g is the surface tension of TRU-Zr alloy, k is the Boltzmann constant, and T is the Kelvin temperature.

An equivalent volume diffusion coefficient for surface diffusion migration of a bubble on grain boundary is given[12] by

$$D_b = 0.301\, D_s a_o^4 \left(r_i^{-4} + r_j^{-4} \right), \tag{5}$$

where, r_i and r_j are the radii of bubbles i and j, and a_o is the lattice constant. It is assumed that TRU-Zr has a primitive tetragonal structure with :

i) T $\leq$1038 K, A=6.02x10^{-8} cm,
a_o = 8.69x10^{-8} cm,

ii) 1038 K $\leq$T $\leq$1198 K ; cubic
a_o = 4.34x10^{-8} cm.

D_s is the surface diffusion coefficient[12] of the bubble on a grain boundary [cm^2/sec, 5.4x10^{-5} exp(-108000/RT)].

Since one fission yields 0.310 atoms of Xe and Kr, and 3.1209415x10^{10} fissions per second yield 1 W/sec, the number of gas atoms produced within a fuel particle is given by

$$m = 0.310 \cdot C_F \cdot q''' \cdot t\ , \tag{6}$$

where, C_F = 3.1209415x10^{10}, t is the irradiation time (sec),
q''' = heat generation rate per fuel particle.

Based on the multiple bubble size distribution[13] on a grain boundary according to the random migration of bubbles, the average number of bubbles per fuel particle in a given bubble size range, $\overline{f_i}$ is given by

$$\overline{f}_i(n_{i+1} - n_i) = 0.23 m' \cdot t^{-4/5} \int_{n_i}^{n_{i+1}} \{\exp(-A(nt^{-2/5} - 0.5)\} \times \{\sinh[B(nt^{-2/5} - 0.5)]^{1/2}\} dn\,. \tag{7}$$

Hence, using $\overline{f_i}$ from Eq. (7), a balance equation can be written as follows.

$$m = C_s \sum_{sizerange} \overline{f_i} \left(\frac{n_i + n_{i+1}}{2} \right), \tag{8}$$

where, C_s is a correction coefficient for the concentration per fuel particle. Therefore, the equivalent number of bubbles for the median atom size at a given bubble size range *i*, is given by

$$f_i = \overline{f_i} \cdot C_s\,. \tag{9}$$

Eq. (9) can be solved for the bubble size distributions incrementally by using the Gaussian quadrature formula.

Finally, swelling due to the accumulation of gas bubbles on the grain boundary is calculated. The volume increase per fuel particle due to the accumulation of gas bubbles is calculated by considering the bubble size distribution, VT_{zr} using

$$V_b = \frac{4p}{3} \sum_{sizerange} \left(\overline{r_i}\right)^3 \cdot f_i , \tag{10}$$

where, V_b is the amount of volume increase due to the formation of fission gas bubbles per a fuel particle. Therefore, fractional swelling due to the formation of fission gas bubbles, per unit fuel core volume, is given by

$$\left(\frac{\Delta V_{uc}}{V_{uc}}\right)_{gb} = \frac{\left(\left(V_{tp} + V_b\right) \cdot N_{tp} + VT_{zr} + V_{ip}\right) - V_{uc}}{V_{uc}} = \left(\left(V_{sp} + V_b\right) \cdot N_{tp} + VT_{zr} + V_{ip}\right) - 1 \tag{11}$$

where, VT_{zr} is the volume of the Zr-matrix per unit fuel core volume, WT_{zr} / d_{zr},

V_{uc} is the unit volume of the fuel core,

$\left\{\frac{\Delta V}{V}\right\}_{sp}$ is the volume of a fuel particle which is treated as an equivalent sphere of uniform size characterized by a single equivalent radius, and

$\left\{\frac{\Delta V}{V}\right\}_{sp}$ is the volume of the initial pore per unit volume of the fuel meat, $\left[1 - \left(V_{tp} \cdot N_{tp} + VT_{zr}\right)\right]$.

4. Benchmark Calculations and Discussion

Fuel swelling models for the dispersion fuel of (TRU-Zr)-Zr fuel have been developed according to the deveolpment program of the DIMAC code. To evaluate the predictive capability of DIMAC with a mechanistic swelling model, benchmark calculations by the DIMAC code were performed and the results were compared with results by the DIFAIR code[2,14], which was already verified. The major input data were prepared as shown in Table 1. The same input data for both cases, such as fuel meat and cladding dimensions, linear power and the burnup, were used so that the swelling levels of (TRU-Zr)-Zr fuel comparable with those of silicide fuel for a

research reactor. Fig. 2 shows a comparison of the swelling components due to the buildup of solid fission products and the accomodation of gaseous fission products, between (TRU-Zr)-Zr and U_3Si-Al fuels. The swelling amount due to the buildup of solid fission products is not only much less than that of gaseous fission products, but it is also directly proportional to the burnup. As mensioned in the foregoing section, the density of solid fission products by fast fission is slightly higher than that by thermal fission. Hence, a value of 1.26 was chosen as the swelling rate constant for

Table 1. The major input data for swelling calculation

Parameters \ Fuel Type		U_3Si-Al Dispersion Fuel	(TRU-10Zr)-Zr Dispersion Fuel
Fuel slug	Fuel dia. (mm)	5.18	5.18
	Composition	61wt%U_3Si-39wt%Al	45wt%(TRU-10Zr)-55wt%Zr
	Density(g/cm^3)	5.3	9.16 (TRU-10Zr : 18.37)
	Fissle Density(g/cm^3)	3.10	3.7
Thermal conductivity of Fuel particle (kW/mK)		0.0193	0.01633
Cladding (mm)	Inside dia	5.28	5.28
	Outside dia	6.68	6.68
	Thickness	0.7	0.7
LHGR (kW/m)		13.5	13.5
Young's modulus of fuel meat of fuel meat (GN/m^2)		88.6	280
Young's modulus of fuel cladding (GN/m^2)		70	172.6
Linear power(kW/m)		13.5	
Thermal conductivity of cladding (kW/mK)		0.2168	0.025
Thermal expansion coefficient of fuel meat ($\mu m/m \bullet K$)		22	17
Thermal expansion coefficient of fuel cladding ($\mu m/m \bullet K$)		24.6	6.1
Surface tension ($J\ m^{-2}$)		0.225	0.5
Poisson's ratio of fuel meat			
Poisson's ratio of fuel cladding			

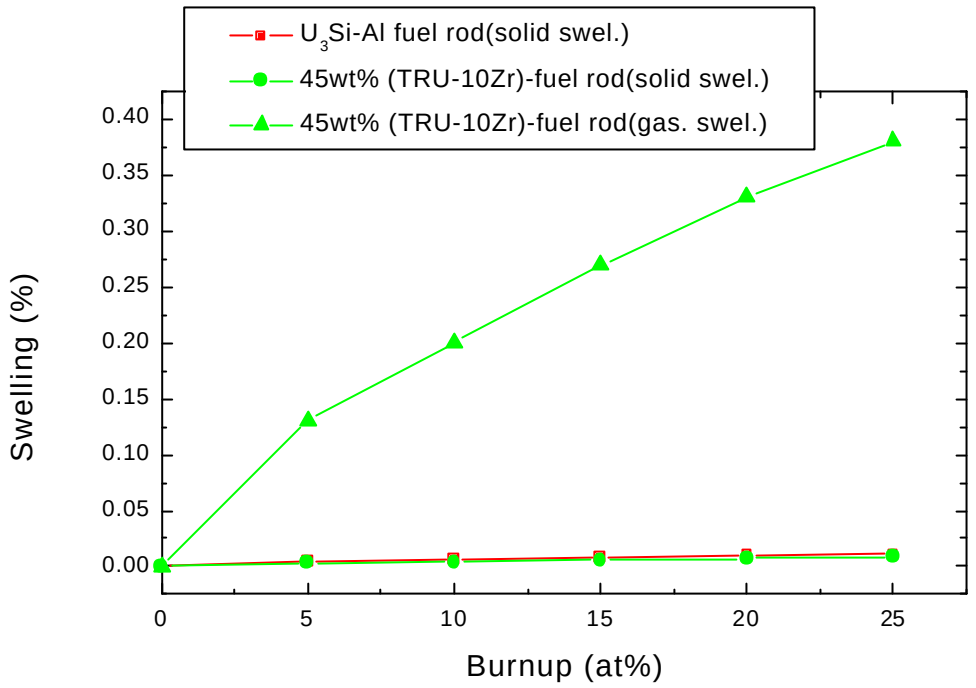

Fig.2 Comparison of swelling components due to solid fission product build-up and gaseous fission product accommodation

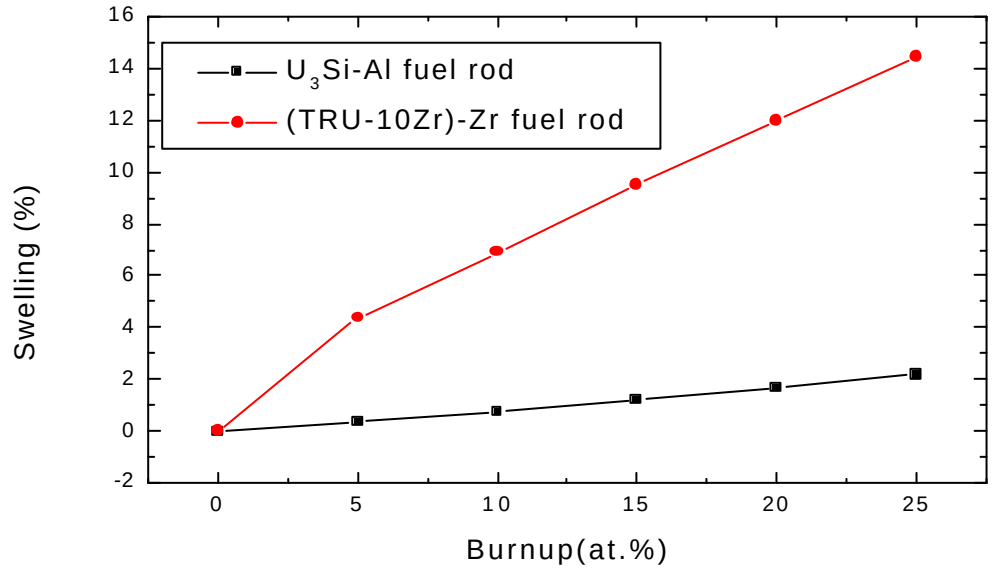

Fig. 3 Fuel swelling comparison between (TRU-10Zr)-Zr and U_3Si-Al fuel rods

(TRU-Zr)-Zr fuel due to the buildup of solid fission products. When it compared with that of silicide fuel under thermal reactor operation conditions, there is no meaningful difference. The swelling rate for (TRU-Zr)-Zr fuel shows slightly lower values. Fig. 3 shows a comparison of the total swellings predicted by the DIMAC and DIFAIR codes. The swelling was calculated for both dispersion fuels of (TRU-Zr)-Zr and U_3Si-Al, with repect to burnup. As shown in Table 1, the linear power was 13.5 kW/m for both cases. The swelling rate of (TRU-Zr)-Zr fuel is much higher than that of silicide fuel, as predicted by the DIFAIR code.

5. Conclusion and Recommendations

The conceptual design of blanket fuel for HYPER has been performed. The in-reactor performance computer code for blanket fuel has also been developed. The present status of DIMAC code deveolpment is in the second step which emphasizes on fuel swelling calculation. In this paper, mechanistic swelling models have been developed and the models were installed into the DIMAC program. The preliminary calculation results predicted by DIMAC were compared with those of silicide fuel predicted by DIFAIR. The swelling levels for (TRU-Zr)-Zr dispersion fuel were much higher than those of silicide fuel. Some material properties which are different with those of silicide fuel were found. While the thermal expansion and swelling rates for U_3Si fuel particles are much less than those of the alumium matrix, the thermal expansion and swelling rates for TRU-Zr fuel particles are much higher than those of the zirconium matrix. These differences may cause the high swelling rate of (TRU-Zr)-Zr fuel. Otherwise, the zirconium matrix may restraint the swelling of TRU-Zr fuel particles. In this case, the stress is not only getting larger as the burnup increases, but is also applied for the matrix during a long residence time. Hence, dispersion fuel performance may be limited by swelling. Fission gas released from the TRU-Zr particles accumulates around the particles and at high burnup the gas pressure causes the metal matrix to swell, as would a heavy-wall pressure vessel. The maximum strain produced may be limited in accord with the reduced ductility of the neutron-embrittled matrix. Therefore, some experimental tests including inpile and outpile experiments are need for clarifing the integrity and the material properties of the inter-metallic fuel core.

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