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A Comprehensive Swelling Model of Silicide Dispersion Fuel for Research Reactor

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연구로용 우라늄실리사이드 분산형 핵연료의 팽윤모델

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Abstract

One of the important irradiation performance characteristics of the silicide dispersion fuel element in research reactors is the diameteral increase resulting from fuel swelling. This paper, will attempt to develop a physical model for the fuel swelling, DFSWELL, by analyzing the basic irradiation behaviours and some experimental evidences. From the experimental evidences, it was shown that the volume changes in irradiated U_3Si–Al were strongly dependent on temperature and fission rate. The quantitative–amount of swelling for silicide fuel is estimated by considering temperature, fission rate, solid fission product build–up and gas bubble behavior. The swelling for the silicide fuel is comprised of three major components :

i) a volume change due to the formation of an interfacial layer between the fuel particle and matrix.

ii) a volume change due to the accumulation of gas bubble nucleation

iii) a volume change due to the accumulation of solid fission products

The DFSWELL model which takes into account the above three major physical components predicts well the absolute magnitude of silicide fuel swelling in accordance with the power histories in comparison with the experimental data.

요 약

硏究用 原子爐의 分散形 核燃料에 대한 노내 조사 거동의 주요 특성중의 하나는 核燃料心 팽윤에 기인된 핵연료봉 직경 증가이다. 본 논문에서는 分散形 우라늄실리사이드 核燃料에 대한 노내 조사거동과 실험 증거들을 분석함으로써 그 핵연료의 팽윤에 대한 물리적 해석모형인, DFSWELL 전산 모형을 개발하였다. 문헌에 보고된 실험 증거들로 부터 노내에서 U_3Si-Al 核燃料心의 부피변화는 온도와 핵분열율에 따라 크게 영향을 받는 것으로 나타났다. 分散形 우라늄 실리사이드 核燃料에 대한 정량적 膨潤量은 주어진 온도, 핵분열율, 핵분열고체생성물 축적 및 핵분열기체 기포거동을 고려함으로써 평가될 수 있다. 연구로의 分散形 우라늄실리사이드 核燃料의 膨

潤 현상은 다음과 같은 세가지 현상으로 귀결된다.

ⅰ) 核分裂氣體生成物 기포 생성/축적에 의한 부피변화

ⅱ) 固體 核分裂生成物의 축적 및 상 변화에 의한 부피변화

ⅲ) 核燃料 粒子와 기지사이의 공유층에 대한 부피변화

상기 세가지의 물리적 현상을 고려하는 본 DFSWELL 전산 모형의 출력이력 조건에 따른 절대 예측치들은 실험 결과와 비교할 때 分散形 우라늄실리사이드 核燃料의 조사후 膨潤 실측치와 잘 일치한다.

1. Introduction

As a research reactor operates, the fission chain reaction in the intermetallic fuel affects the fuel microstructure in several ways. At the same time, the fuel element is subjected to the temperatures and, there exist temperature gradients along the radial direction in the fuel element under time varying conditions. Consequently, the material properties, such as plasticity, elastic and creep constants, material phase, etc., would change. In reactor irradiation experiments with intermetallic uranium compounds, Hofman showed[1] that an enormous increase in gas bubble growth occurs when a compound becomes amorphous during irradiation. To date, no a physical model for uranium–silicide fuel has been reported. This paper represents an attempt to develop a physical model to efficiently predict the swelling of the silicide fuel according to power history. To develop a physical model, it is necessary to analyze the irradiation behaviours of uranium silicide fuels. As described in section 2, one of the important irradiation performance characteristics of a silicide dispersion fuel element in research reactors is its diameteral increase resulting from fuel swelling. By analyzing the irradiation behaviours in the literature on experimental data for silicide dispersion fuel, a analytical swelling model is derived. The objective of this report is to provide an analytical swelling model by studying the basic irradiation behavior of uranium silicides which were reported by AECL(Atomic Energy of Canada Limited) and ANL(Argonne National Laboratory).

2. Physical Processes for Dispersion Fuel Core Swelling

The most important part of developing a physical model of any sort is the identification of the controlling physical processes. So far, some contributing mechanisms have been suggested for the swelling of silicide dispersion fuel. Several key physical processes are occurring simultaneously as damage is sustained in dispersion fuel. Even though the current models of the swelling behavior for silicide fuel do not adequately explain the observed swelling phenomena, the limited amount of experimental evidences acquired so far have led to the following summary :

(i) U_3Si–Al and U_3Si_2–Al are thermodynamically unstable. The type and amount of reaction products (uranium aluminides) formed, obviously varies with concentration and temperature, and can be predicted from the ternary phase diagram. The uranium aluminides UAl_2, UAl_3 and UAl_{4+x} dissolve substantial amounts of silicon at higher temperatures, the solubility is however, in the case of UAl_2 and UAl_3, temperature dependent.[2]

(ii) From the result of FZZ–909A experiment, post–irradiation metallography revealed that interfacial layers around reacted silicide particles had reached a thickness of 7.5 μm near the fuel core periphery and 25 μm around particles near the fuel core centre. Again "reacted silicide" particles had coalesced.[3]

(iii) The swelling observed in most specimens was clearly associated with a reaction between the fuel particles and the aluminum matrix, the alu-

miunm having reacted with particle surfaces and also penetrated along networks of grain boundaries to attack sub–grains throughout the U_3Si. The end point of such a reaction would occur when all the U_3Si (and the precipitates of U_3Si_2 contained within the U_3Si particles) had been consumed to form $UAl_3Si_{1/3}$.Rates of reaction would be reduced eventually (as observed) due to the impedence, offered to diffusing aluminum atoms, by the $UAl_3Si_{1/3}$ surrounding the U_3Si particles and growing in thickness.[4,5]

(iv) The swelling is comprised[6] of three major components ;

- a volume change due to the transformation to a higher silicon phase as a result of uranium burnup,
- volume increase due to the accumulation of non–gaseous fission products,
- volume increase due to fission gas accumulation.

The volume change due to uranium burnup will be calculated using the most current uranium–silicon equilibrium phase diagram and measured densities of the phases involved. The amounts of non–gaseous fission products were determined with the aid of published fission yield data for U–235. These contributions to volume change were estimated by evaluating their solubility in the fuel and the tendency to form compounds with each other and with uranium and silicon. 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 the fuel swelling is due to the formation of fission gas bubbles.

(v) Nucleation of fissin gas bubble in U_3Si fuel was apparently very uniform as compared to the USi*Al fuel. But the fission gas bubble sizes were larger near the fuel core periphery than at the fuel core centre in both cases.[7,8] It would be considered likely that this result was due to the fact that the local fission rate along the radial direction within the fuel core is different. Since the thermal neutron flux originates outside the fuel element in the moderator, the flux decreases as one moves towards the center of the fuel due to neutron absorption in the fuel. As irradiation proceeds, the original fissile atoms are depleted in the fuel, fission products build up and new fissile atoms are formed.

(vi) 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. Apparent fission rate dependence is due to a delay in development of larger bubbles when fission rate increases.[6]

(vii) The aluminum having reacted with particle surfaces formed interfacial layers around the silicide particles in the fuel core. The interfacial layers were thinner[3,7,8] near the fuel core periphery than at the fuel core centre. It should be noted that the density of the interfacial layer is lower than that of U_3Si particle. This experiment shows that one of the important contributing parameters for forming reaction products (uranium aluminides) is temperature. The reacted areas near the fuel particles in USi * Al fuel were much larger than those of U_3Si fuel. The mobility of the fission gas bubble in U_3Si must be rather low as compared with USi*Al fuel, so that each bubble has a limited capture range for fission gas. Metallography[7] also confirmed that there was no evidence of back–diffusion of uranium or silicon into the aluminum matrix.

(viii) The fine particles had essentially completely reacted forming the less dense UAl_3 while only thin interfacial layers formed around the coarser

silicide particles. The larger surface area provided by the fine particles undoubtedly led to the increased reaction and contributed significantly to the observed core swelling. Small fission gas bubbles up to 8 μm in diameter were contained in the silicide kernels, but few fission gas bubbles had been retained in the aluminide.[9]

(ix) During heat treatment of Al–USiAl particles above a temperature around 200 ℃, the aluminum diffuses into the USiAl particles along grain and particle boundaries forming UAl_3 and eventually UAl_4. The stresses resulting from the formation of these new phases cause cracking of the individual U_3Si grains permitting aluminum to diffuse along the cracks and form more UAl_x. The aluminum apparently diffuses into the USiAl particles along unstable paths such as grain boundaries or subparticle boundaries to form the new Al–U compounds.[10]

(x) In reactor irradiation experiments with intermetallic uranium compounds, Hofman[1] showed that an enormous increase in gas bubble growth occurs when a compound becomes amorphous during irradiation. Fission gas bubbles have recently been shown to exhibit extremely high growth rates at relatively low temperatures in certain uranium compounds. 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.[11] The ion bombardment and in situ TEM experiments of Birtcher et al.[12] on U_3Si underscore the general nature of the effect. The microhardness of U_3Si (measured after irradiation) increased slightly after prolonged neutron irradiation, during which the compound became amorphous and experienced excessive swelling.[11]

From the foregoing description, the volume changes in irradiated U_3Si–Al were shown to be strongly dependent on temperature and fission rate.

The quantitative–amount of swelling for silicide 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 silicide 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 the transformation to a higher silicon phase as a result of uranium depletions as burnup increase and the accumulation of solid fission product swelling

$\left\{\frac{\Delta V}{V}\right\}_{il}$: the fractional volume change due to thermal–chemical reaction between fuel particle and matrix.

3. The Model Description

3.1 The Modelling of the Volume Change due to the Formation of the Interfacial Layer Between the Fuel Particle and Matrix

As per the foregoing description in chapter 2, the aluminum having reacted with particle surfaces formed interfacial layers around the silicide particles in the fuel core.

Even though there are many features for the interfacial layer, the major feature may be explained by the following simple model. The basis reaction between the aluminum matrix and U_3Si particles is[4]

$$U_3Si + 9\ Al \longrightarrow 3\ UAl_3Si_{1/3}. \quad (1)$$

It is infered that the nine aluminum atoms that

react with each U_3Si molecule leave behind in the aluminum matrix nine vacancies.

The vacancies are created at the U_3Si/matrix interface and apparently do not migrate away to the free surface, but condense to form pores with volumes which will be assumed to be equal to the sum of the volumes of the vacancies.

If the radius of silicide particle is R and the thickness of the interfacial layer is L, the initial volume of the interfacial layer, V_{IL}, is given by

$$V_{IL}=\int_{rb}^{R}4\pi r^2\,dr \qquad (2)$$
$$=\frac{4}{3}\cdot\pi\cdot(R^3-r_b^3),$$

where, r_b is (R−L), and V_{IL} is the initial volume of interfacial layer per a silicide particle. On the other hand, the number of silicide particles, N_{sp}, per unit volume of fuel meat, is given by

$$N_{sp}=\frac{\delta_m\cdot F_{sp}}{M_{sp}}, \qquad (3)$$

where, δ_m is the density of fuel meat, and F_{sp} is weight fraction of U_3Si, and M_{sp} is the weight of a silicide particle.

Considering the basis reaction as shown in Eq.(1), the weight of Al required to react with all the U_3Si per unit volume of fuel meat, W_{al}, is given by

$$W_{al}=\frac{243}{742}\cdot V_{IL}\cdot\delta_{sp}\cdot N_{sp}, \qquad (4)$$

where δ_{sp} is the density of U_3Si, 15.58, and 243 is nine times the atomic weight of Al, and 742 is the molecular weight of U_3Si. And the Al-weight per the unit volume of Al–U_3Si, WT_{al}, is given by

$$WT_{al}=\delta^{m}\cdot F_{al}, \qquad (5)$$

where, F_{al} is weight fraction of Al in Al–U_3Si fuel meat.

Hence, the increased volume due to the formation of the interfacial layer, for the unit volume of fuel meat, VT_{IL}, is given by

$$VT_{IL}=\frac{(V_{ILT}\times\delta_{sp})+W_{al}}{\delta^{il}}+\frac{WT_{al}-W_{al}}{\delta_{al}}+$$
$$\frac{(M_{sp}-(V_{IL}\cdot\delta_{sp}))\cdot N_{sp}}{\delta_{sp}}+V_{ip}, \qquad (6)$$

where,

$V_{ILT}=V_L\cdot N_{sp}$, δ_{il} is the density of $UAl_3Si_{1/3}$, 6.8,

δ_{sp} is the density of U_3Si, 15.58,

δ_{al} is the density of Al, 2.7,

V_{ip} is the initial pore volume per unit volume of fuel meat, $(1-(V_{sp}\cdot N_{sp}+VT_{al}))$,

V_{sp} is the volume of a silicide particle, and

VT_{al} is the volume of Al-matrix per unit meat volume, WT_{al}/δ_{al}.

Also, the unit volume of fuel meat, V_{uc} is given by

$$V_{uc}=V_{sp}\cdot N_{sp}+\frac{WT_{al}}{\delta_{al}}+V_{ip}=1 \qquad (7)$$

Therefore the fractional volume increase due to the formation of interfacial layer, for the unit volume of fuel meat, is given by

$$\left(\frac{\Delta V_{uc}}{V_{uc}}\right)_{il}=\frac{VT_{IL}-V_{uc}}{V_{uc}}$$
$$=VT_{IL}-1. \qquad (8)$$

3.2. The Modelling of the Volume Change due to the Accumulation of Solid Fission Products

The swelling due to solid fission products is caused by the incorporation of solid fission products in the solid lattice and is, therefore, proportional to the number of fissions which have taken place in the fuel. Also, the volume change due to the transformation to a higher silicon phase as a result of uranium burnup is proportional to the fission rate. Therefore, these two contributions to swelling are combined and yielded as a linear

function of fission density.

The volume change for a silicide particle, f_{sp}, due to solid fission is given by:

$$f_{sp}=C\cdot 10^{-3}\cdot B_a, \tag{9}$$

where, C is a coefficient ranging from 1.26 to 1.86 which were drived from the experimental data[5,13], and B_a is the atomic % burnup. Therefore, the fractional volume change due to the formation of solid fission product, for unit volume of fuel meat, is given by

$$\left(\frac{\Delta V_{uc}}{V_{uc}}\right)_{sp}=\left\{\left(\frac{WT_{al}}{\delta_{al}}\right)+V_{ip} +(V_{sp}\cdot N_{sp})\cdot(1+f_{sp})\right\}-1. \tag{10}$$

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

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 particle. The procedure for calculating the bubble size distribution consists of dividing the bubbles into equal size ranges on a logarithmic scale and averaging properties over the ranges as per Poeppel[14,15].

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 functions of temperature and pressure. If n_i is the number of gas atoms in the bubble with the radius, r_i is given by the following expression [16,17]

$$r_i=\left\{(3kT/8\pi\gamma)n_i\right\}^{1/2}. \tag{11}$$

Hence, the mean radius, $\bar{r}_i$, at a region which is ranged from r_i to r_{i+1} is given by

$$\bar{r}_i=\left\{(3kT/8\pi\gamma)\left((n_i+n_{i+1})/2\right)\right\}^{1/2}. \tag{12}$$

where γ is the surface tension of U_3Si, k, the Boltzmann constant, and T, the Kelvin temperature.

An equivalent volume diffusion coefficient for the surface diffusion migration of a bubble is given[18] by

$$D_b=0.301\ D_s\ \alpha_o^4(\bar{r}_i^{-4}+\bar{r}_j^{-4}), \tag{13}$$

where r_i and r_j are the radii of bubbles i and j, α_o is the lattice constant. U_3Si has a primitive tetragonal structure[2] with:

i) $T\leq 1038$ K; $A=6.02\times10^{-8}$cm, $c=8.69\times10^{-8}$cm,
ii) 1038 K$\leq T\leq$1198 K; cubic $a=4.34\times10^{-8}$cm.

And D_s is the surface diffusion coefficient[19] [cm^2/sec, $5.4\times10^{-5}\exp(-108000/RT)$].

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

$$m=0.310\cdot C_F\cdot q'''\cdot t, \tag{14}$$

where, $C_F=3.1209415\times10^{10}$, t is irradiation time (sec),

q'''=heat generation rate per unit fuel particle.

Based on the multiple bubble size distribution[18] according to the random migration of the bubbles, the average number of bubbles per unit fuel particle at given bubble size range, $\bar{f}_i$ is given by

$$\bar{f}_i(n_{i+1}-n_i)=0.23m\cdot\tau^{-4/5} \int_{n^i}^{n_{i+1}}\left[\exp(-A(n\tau^{-2/5}-0.5))\right] \left[\sinh\sqrt{B(n\tau^{-2/5})-0.5}\right]dn. \tag{15}$$

Hence, using $\bar{f}_i$ from Eq. (15), a balance equation can be written as follows

$$m=C_s \sum_{\text{size range}} \bar{f}_i \cdot \frac{(n_i+n_{i+1})}{2}, \quad (16)$$

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

$$f_i=\bar{f}_i \cdot C_s. \quad (17)$$

Eq. (15) can be solved for the bubble size distributions incrementally by using the Gaussian quadrature formula[20] which approximate the definite integral by the expression

$$\int_{-1}^{1} f(x)\,dx = w_O\, f(X_O)+W_1\, f(x_1) + W_w\, f(x_2)+\cdots\cdots+W_n\, f(x_n) = \sum_{i=0}^{n} W_i\, f(x_i), \quad (18)$$

where w_o, $w_1 \cdots w_n$ are the weighting coefficient and x_o, x_1, $\cdots x_n$ are the associated points.

Finally, the swelling due to the accumulation of gas bubbles is calculated. The volume increase per fuel particle due to the accumulation of gas bubbles is calculated by considering the bubble size distribution, f_i, using

$$V_b=\frac{4\pi}{3} \sum_{\text{size range}} (\bar{r}_i)^3 f_i, \quad (19)$$

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

$$\left(\frac{\Delta V_{uc}}{V_{uc}}\right)_{gb} = \frac{((V_{sp}+V_b)\cdot N_{sp}+VT_{a1}+V_{ip})-V_{uc}}{V_{uc}} = ((V_{sp}+V_b)\cdot N_{sp}+VT_{a1}+V_{ip})-1, \quad (20)$$

where, VT_{a1} is the volume of Al-matrix per unit meat volume, WT_{a1}/δ_{a1},

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

V_{sp} is the volume of a fuel particle which is treated as a equivalent sphere of uniform size characterized by a single equivalent radius, and

V_{ip} is the volume of the initial pore per unit volume of fuel meat, $[1-(V_{sp}\cdot N_{sp}+VT_{a1})]$.

4. Model Application

In order to verify the present swelling model, it is necessary, first of all, to compute the temperature distribution depending on the power history in the fuel element. Therefore, a program[21] which computes the temperature distribution was used in this study, and the present swelling model was incorporated into the program. This incorporated code is referred to as DFSWELL hereafter. DFSWELL code predicts the swelling behavior of silicide fuel by considering the volume change for unit silicide particle and unit volume of the fuel meat, depending on temperature, fission rate, solid fission product build-up and gas bubble behavior. The swelling model for the silicide fuel is comprised of three major components :

- a volume change due to the formation of an interfacial layer between the fuel particle and matrix
- a volume change due to the accumulation of gas bubble nucleation
- a volume change due to the accumulation of solid fission products.

The number of gas atoms produced in a silicide particle and the multiple-bubble size distribution depending on the power history were calculated in order to estimate the volume change of fuel meat due to gas bubble formation and growth. Here, the Van der Waals gas law is employed to calculate the bubble size. On the other hand, the behaviours of the interfacial layer and solid fission products were estimated. DFSWELL, incorporating these three mechanisms, was programmed as

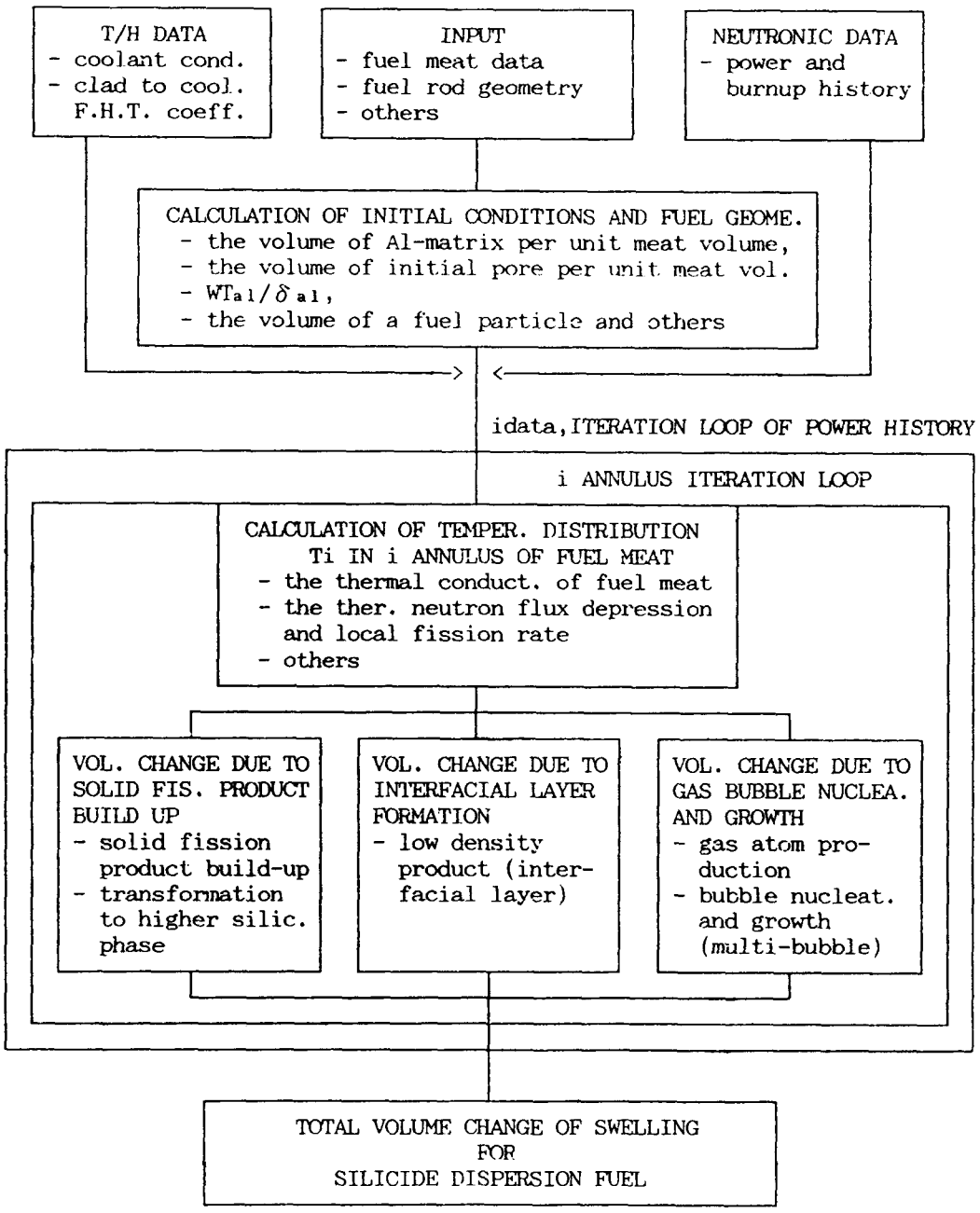

Fig. 1. Calculational Procedure of DESWELL Program

shown in Fig. 1.

5. Comparison with Experimental Data and Discussion

Experimental data are taken from AECL[7,9] in which the silicide fuels for research reactor were irradiated up to 97.5 atomic % burnup and with the linear powers between 28 kW/m and 87 kW/m, as shown in Fig. 2 and Fig. 3.

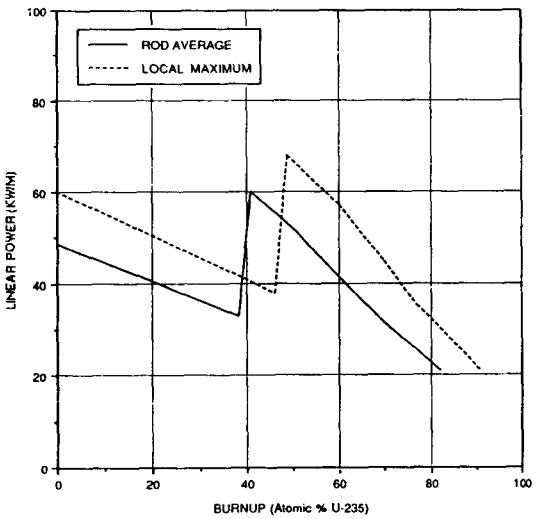

Fig. 2. Typical Power History for LEU Fuel

The irradiation test elements were fabricated into aluminum-clad full-length element about 292 mm long, the fuel core diameter being 5.49 mm and clad wall thickness 0.76 mm. Each element had six cooling fins at 60 ℃ intervals around the cladding, the fin width being 0.76 mm and fin height 1.27 mm. The outside diameter of the cladding was 7.01 mm (exceeding fins).

Fig. 4 shows the results of parametric study on the swelling mechanisms for the silicide dispersion fuel. From this result, the volume change due to the accumulation of the solid fission products is proportional to the burnup and its absolute magitude is relatively less than that of gas bubbles. And, it is apparent that the most dominant parameter of silicide fuel swelling is the volume change due to gas bubble formation and growth.

Fig. 5 shows the calculated fuel swelling versus

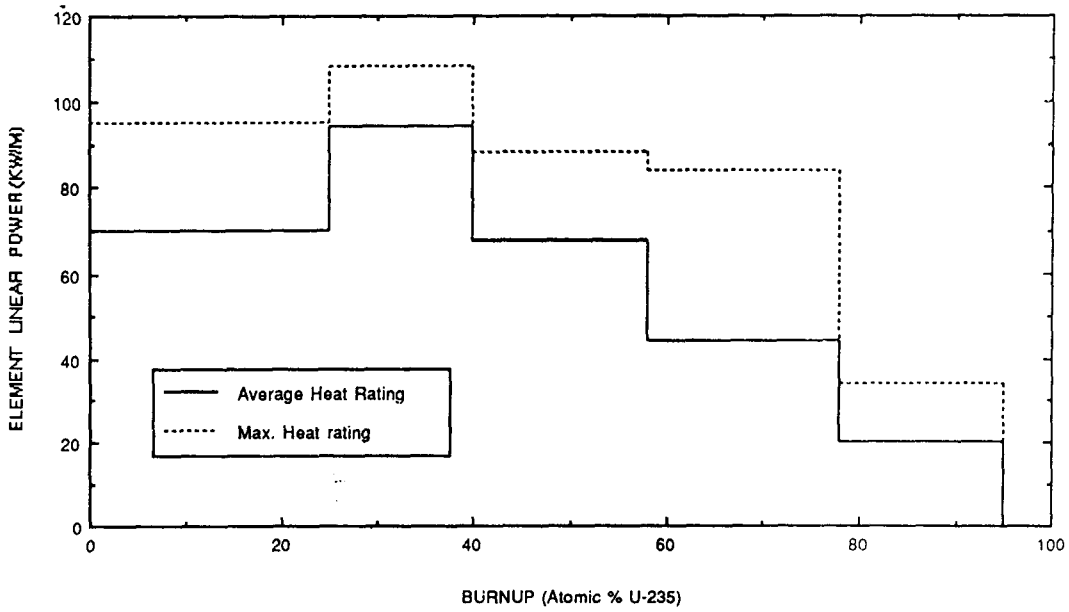

Fig. 3. Power History of Mini-Elements Containing Al-61.5 wt % Uranium Silicide from Experiment EXP-FZZ-918

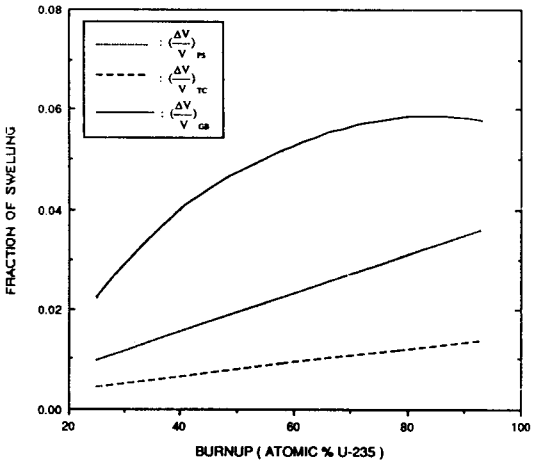

Fig. 4. The Results of Parametric Study on the Swelling Mechanism for EXP-FZZ-918 Fuel Rod

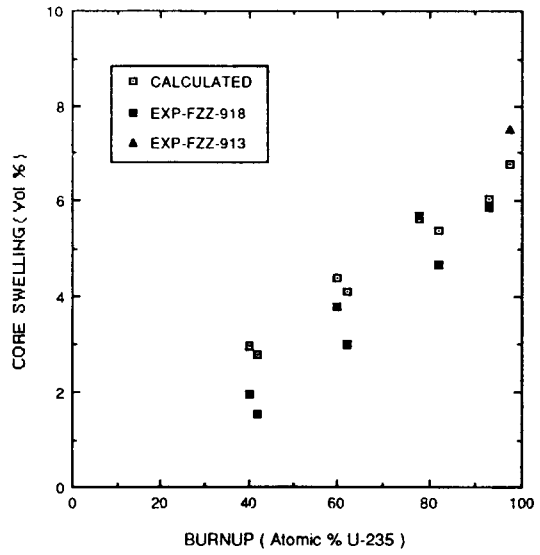

Fig. 5. Comparison of Observed Swelling of LEU Silicide Dispersion Fuel with the Calculated Swelling Using Present Model, as a Function of Burnup

experimental data. In this Figure, the DFSWELL code predicted well the absolute magnitude of the swelling, in comparison with the wide variety of experimental data.

6. Conclusions and Recommendations

One of the important irradiation performance characteristics of dispersion fuel element in research reactors is its diameteral increase resulting from fuel swelling. In this paper, a comprehensive swelling model for the silicide dispersion fuel has been developed by analyzing the basic irradiation behaviours and some experimental evidences.

From the predictions of DFSWELL model and the experimental evidences, it is proposed that the swelling for silicide fuel is comprised of the following three major components :

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

DFSWELL model which takes into account above three major components on the physical phenomena, predicts well the absolute magnitude of silicide fuel swelling depending on the power histories in comparison with the experimental data. It appears that the most dominant parameter of silicide fuel swelling is the volume change due to gas bubble formation and growth.

However, it seems that DFSWELL model tends to overpredict in comparison with experimental results, especially at the region of low burnup. It is therefore recommended that further work for modelling the formation of interfacial layer dependent on the burnup might be required.

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