MODEL DEVELOPMENT OF UO2-ZR PLATE-TYPE FUEL BEHAVIOR AT EARLY PHASE OF SEVERE ACCIDENT AND MOLTEN FUEL MEAT RELOCATION

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ABSTRACT

According to former study on oxygen diffusion, Nb-Zr solid reaction and UO_2-Zr solid reaction, models of oxidation, solid reaction in fuel meat and relocation of molten fuel meat are developed based on structure and material properties of UO_2-Zr plate. The new models can supply theoretical elements for safety analysis of core assembled with dispersion plate-type fuel under severe accident.

KEYWORDS
UO_2-Zr plate-type fuel, Severe accident, Core behavior at early phase, Relocation of molten fuel meat

1. Introduction

As an advanced type of nuclear fuel, plate-type fuel has some advantages, such as high heat conductivity, high heat flux and deposition of radical production. However, new properties of structure and materials affect safety properties during severe accident.

The irradiation experiments on UO_2-S.S. plate fuel performed at 1950s-1960s in US indicated that plate fuel swells under high fuel cost[1,2]. The research also gave data about impact of different parameters on swelling and limited fuel cost and fuel temperature upon which fuel could swell. Swelling process under high fuel cost in plate-type fuel is also studied in research report of RERTR[3]. The heat stability experiments performed by RERTR (Reduced Enrichment Research and Test Reactor) on plate fuel showed that solid reaction between fuel matrix and fuel particles in fuel meat will change thermal and physical properties of fuel meat and take impact on fuel failure. Some summary reports of fuel performance at high temperature by INL[4], CEA[6] and IAEA[5] also indicated the impact of solid reaction on fuel failure of plate-type fuel.

The fuel molten experiments of UAl plate-type fuel performed in SRS(Savannah River Site) [7]showed that UAl fuel will drain with molten fuel meat and heat source relocation process could take different severe accident properties of power relocation, melting of control rods and re-criticality because of the high enrichment of uranium in plate-type fuel.

Thus, such special phenomenon and behaviors of plate-type fuel affect severe accident properties and risks, and studies on mechanism and models are limited even though experimental research is enough. Thus, studies on mechanism and models of special phenomenon are investigated in the paper according to some related experiments.

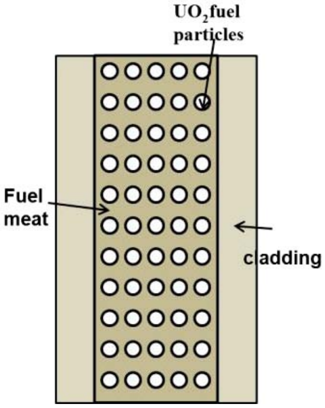


Figure 1 Structure of UO_2-Zr plate

2. Model development

UO_2-Zr plate-type fuel consists of Zircaloy cladding, zirconium fuel meat with dispersed UO_2 fuel particles. As showed in fig.2, during the heat treatment experiment, UO_2-Zr plate type fuel has some special phenomenon: fuel swelling, solid reaction in fuel meat, oxygen diffusion from UO_2 fuel particles and fuel meat melting process at low temperature. Fuel swelling will destruct integral structure of the fuel, take a new fuel failure mode at the lower temperature than cladding oxidation. Solid reaction in fuel meat will change thermal and physical properties of fuel meat; Relocation process of molten fuel materials with UO_2 fuel particles and fuel meat take impact on sequence of core degradation and re-criticality properties. The study on models and mechanism of such phenomenon is the key to research severe accident properties of UO_2-Zr plate-type fuel. Model development of every special phenomenon will be discussed respectively.

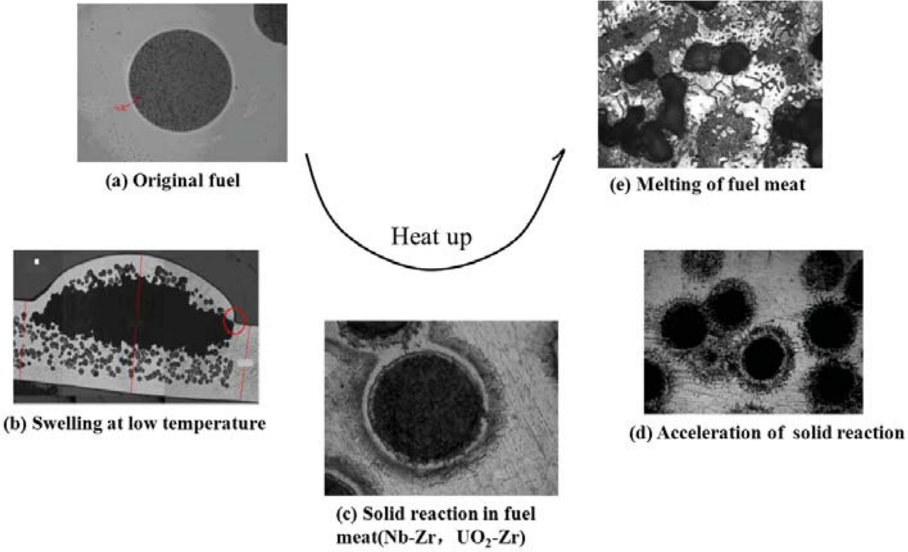


Figure 2 Fuel behaviors of UO_2-Zr plate at high temperature

2.1 Model development of oxygen diffusion

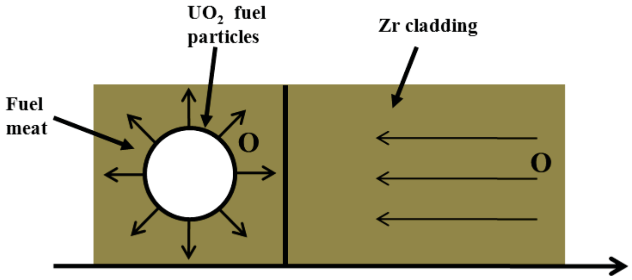


Figure 3 Oxygen diffusion process of UO_2-Zr dispersion plate fuel

As showed in fig.3. Dispersion plate-type fuel has some special oxidation properties: at high temperature, fuel cladding will react with steam and due to metallurgy connect between fuel meat and cladding, oxygen can transfer across the interface; UO_2 fuel particles with high oxygen concentration will diffuse oxygen to fuel meat with low oxygen concentration at high temperature. The study on oxidation can research oxygen absorption, heat release and composition exchange in fuel meat which can supply data for fuel failure research. Due to complex structure of dispersion plate-type fuel, diffusion method is chosen to investigate oxidation. According to mass conservation, oxygen diffusion equations can be constructed as follows.

Oxygen conservation equation in metal plate:

$$\int_V \frac{\partial (c)}{\partial t}\mathrm{d}v = \int_S D_m \nabla c \mathrm{d}s + \int_S Q_S \mathrm{d}s - \underline{\int_V \frac{\mathrm{d}\delta}{\mathrm{d}t}\frac{\partial (c)}{\partial x}\mathrm{d}v} \qquad (1)$$

Oxygen conservation equation in oxidized plate:

$$\int_V \frac{\partial (c)}{\partial t}\mathrm{d}v = \int_S D_{oxi} \nabla c \mathrm{d}s + \int_S Q_S \mathrm{d}s - \underline{\int_V k\frac{\mathrm{d}\delta}{\mathrm{d}t}\frac{\partial (c)}{\partial x}\mathrm{d}v} - \underline{\int_V \frac{x}{k}\frac{\mathrm{d}k}{\mathrm{d}t}\frac{\partial (c)}{\partial x}\mathrm{d}v} \qquad (2)$$

Where c is defined to be oxygen concentration, D_m is the oxygen diffusion coefficient in metal plate which is quite different in different phases, D_{oxi} is the oxygen diffusion coefficient in oxidized plate, Q_s is the oxygen diffusion intensity from UO_2 fuel particles, k is the expansion coefficient due to phase change and $d\delta$ is the moving length of interface between metal plate and oxidized plate. The items underlined are intensity of oxygen convection by phase exchange and expansion. Compared with traditional oxygen diffusion equations of normal fuel cladding, such equations have a new item Q_s, thus the key of solving such equations is to investigate detailed expression of Q_s. Oxygen diffusion from UO_2 to Zr fuel matrix consists of three stages: oxygen diffusion in UO_2, coating material and Zr fuel matrix. Studies on oxidation properties of different nuclear materials show that oxygen diffusion ability in these three materials are quite different, and oxygen diffuses much slowly in UO_2 fuel particles than the others. Thus, oxygen diffusion process from UO_2 to Zr fuel matrix is dominated by oxygen diffusion in UO_2. The special oxygen diffusion properties of UO_2 fuel particles, Niobium and Zircaloy make oxygen diffusion simulation in finite spherical coordinate is possible. According to oxygen diffusion coefficient kinetic equation concluded by A.Ya.Kupryazhkin[8], James T. Clenny[9], oxygen diffusion coefficients of three materials differ hugely even reach four magnitude. Coefficient data is listed in Table 1.

Tab.1 oxygen diffusion coefficient($\mu m^2 \cdot s^{-1}$)

Materials	1000 K	1500 K	2000 K
UO_2	2.33×10^{-6}	0.09	23

Coating	2.8	129	874
Zr	1.8	204	2178

According to coefficient data in Table 1, oxygen diffusion in UO_2-Zr fuel meat is dominated by oxygen diffusion in UO_2 fuel particle and oxygen concentration would smooth in Nb and Zircaloy immediately, so intensity of oxygen diffusion at interface of UO_2 and Nb could be estimated conveniently. Oxygen equilibrium equations are listed below,

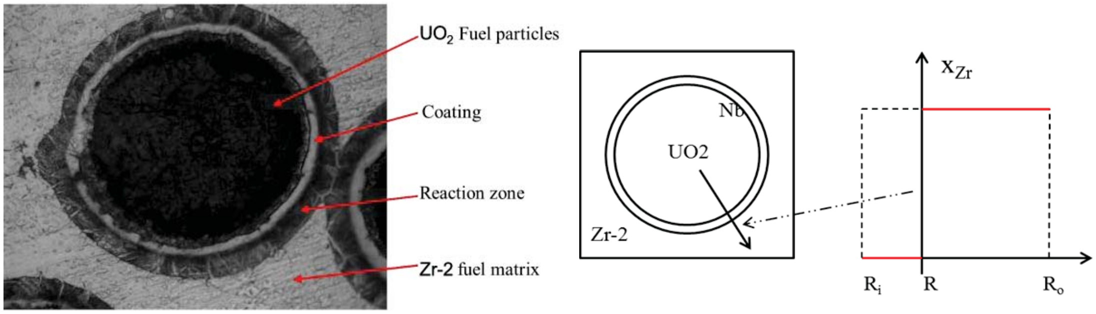
$$\begin{aligned} &\frac{\partial(rc)}{\partial t} = D\frac{\partial^2(ru)}{\partial r^2} \\ &c'|_{r=0} = 0 \\ &c|_{t=0} = c_s \\ &-Dc'|_{r=R} = q" = h(c - c_{equ}) \end{aligned} \tag{3}$$

Where D is oxygen diffusion coefficient of UO_2, h is a transfer coefficient which can be described as a function of atom transition frequency and c_{equ} is equilibrium oxygen concentration in Nb and Zr. The equations can be solved by variable separation approach and results is listed below,

$$\begin{aligned} &y = \frac{1}{r}\sum_{n=1}^{\infty} -\frac{2c_s}{\beta_n}\cos\beta_n R \sin\beta_n r e^{-D\beta_n^2 t} \\ &q" = \frac{2Dc_s}{(1+\lambda^2\beta_n^2)R + a}\sum_{n=1}^{\infty} e^{-D\beta_n^2 t} \\ &\beta_n \approx \frac{n\pi}{R} \end{aligned} \tag{4}$$

So the oxygen diffusion can be estimated by using equation 4.

2.2 Model development of solid reaction in fuel meat



(a) **(b)**

Figure 4 Solid reaction in fuel meat (a.Metallography examination of fuel meat; b.Model of solid reaction)

As figured in fig.4a, there are two solid reaction processes during fuel plate being heated-up:

(1) When temperature of fuel plate is higher than 800 centigrade, solid reaction between Zr fuel matrix and Niobium coating becomes obvious, Zirconium and Niobium atoms diffuse into each other and form a new mixed reaction plate.

(2) As Nb-Zr solid reaction goes on, Zirconium atoms concentration in coating plate becomes higher and higher, when Zirconium atoms contact UO_2 fuel particles, Zirconium atoms will diffuse into UO_2 and induce solid reaction between Zr-UO_2. Due to the solid reaction, UO_2 fuel particles are dissolved and volume turns to be smaller. Composition of the mixed reaction plate changes together with solid reaction.

Due to little temperature grads in fuel meat, a unit cell with a single UO_2 fuel particles is chosen to research solid reaction. The solid reaction model in fuel meat is showed in fig.4b. Nb-Zr and UO_2-Zr solid reaction are dominated by atoms diffusion, and solid reaction between Nb and UO_2 is inert, thus the solid reaction model can be build by coupling Nb-Zr solid reaction model and UO_2-Zr solid reaction model at interface between coating and fuel particles.

(1) Nb-Zr solid reaction model

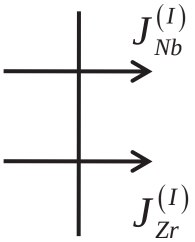


Figure 5 Nb-Zr atoms diffusion across fixed interface

Atoms diffusion intensity is decided by grads of chemical potential, thus atoms diffusion intensity has such expressions:

$$\begin{aligned} J_{Nb}^{(I)} &= -C_{Nb} M_{Nb} \nabla \mu_{Nb}; dC_{Zr} = -\nabla J_{Zr}^{(I)} \mathrm{d}t \\ J_{Zr}^{(I)} &= -C_{Zr} M_{Zr} \nabla \mu_{Zr}; dC_{Nb} = -\nabla J_{Nb}^{(I)} \mathrm{d}t \end{aligned} \tag{5}$$

Then mole concentration exchange of Zr atoms in a unit volume can be deduced by atoms conservation and has expression listed as follows:

$$\frac{\mathrm{d}x_{Zr}}{\mathrm{d}t} = \nabla D_{eff} \cdot \nabla x_{Zr} + \frac{2}{r} D_{eff} \cdot \nabla x_{Zr}$$

$$D_{eff} = x_{Zr} x_{Nb} \left[M_{Zr} \left(\frac{\partial \mu_{Zr}}{\partial x_{Zr}} - \frac{\partial \mu_{Zr}}{\partial x_{Nb}} \right) - M_{Nb} \left(\frac{\partial \mu_{Nb}}{\partial x_{Zr}} - \frac{\partial \mu_{Nb}}{\partial x_{Nb}} \right) \right] \tag{6}$$

Where D_{eff} can be expressed as a function of Gibbs energy. Thus, Nb and Zr atoms distribution in commixed reaction plate can be estimated by solving eq.6.

(2) UO_2-Zr solid reaction

UO_2-Zr solid reaction experiments performed by P.hoffman[10] showed that solid reaction between UO_2 and Zr will form three reaction plates (U-Zr-O, U-Zr, α-Zr) and reaction coefficients of every plate have expressions:

$$\begin{aligned} &(i) \quad K = 0.32 exp(-49000 / RT) \\ &(ii) \quad K = 0.69 exp(-49500 / RT) \\ &(iii) \quad K = 0.70 exp(-44000 / RT) \\ &K = x^2 / t (cm^2 / s) \end{aligned} \tag{7}$$

Where K is reaction coefficient, i , ii , iii are U-Zr-O、U-Zr、α-Zr respectively. Due to spherical geometry of UO_2 fuel particles, the equations should be transferred to be consistent with suit for spherical coordinate. By applying Kuenstlinger equation, the equations can be transferred as follows:

$$K' = \frac{2K}{R_0^2} x_{Zr}; \qquad \frac{\mathrm{d}G}{\mathrm{d}t} = K' \frac{(1-G)^{1/3}}{1-(1-G)^{1/3}} \tag{8}$$

Where R_0 is radius of UO_2 fuel particles, K' is the new reaction coefficient after transformation, x_{Zr} is mole fraction of Zr atoms at interface, G is ratio of reaction volume to total volume of particle. By solving equ.8, radius of UO_2 fuel particles along time can be estimated.

2.3 Model development of relocation process

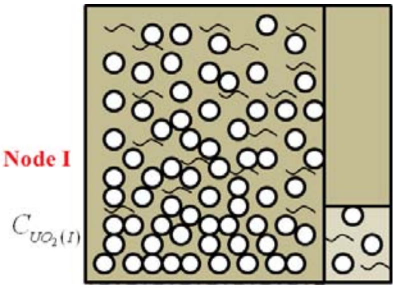


Figure 6 Relocation model of molten fuel meat

As listed in fig.6, fuel meat with lower melting point melts prior to fuel cladding and when there is a crack or rupture, molten fuel meat will drain into flow path of coolant. Because of small volume, the un-dissolved UO_2 fuel particles will drain together with molten fuel meat. The configuration of relocated molten materials is concerned with fuel failure mode. When fuel is failed by fuel swelling under high fuel cost, the crack are generally large, molten materials will drain rapid into flow path and form a molten block across flow path; When fuel is failed by embitterment, the crack are generally small, molten materials will drain into flow path with configuration of semi-spherical. Different relocation configuration has different freezing properties; the model of relocation process for semi-spherical molten materials has been well developed, but little study on model of relocation process for molten block. Because heat transfer rate is much larger than flow rate of molten materials, so flow and heat transfer process can be separated. Heat transfer process can be investigated by Heat balance Integral method proposed by T.R.Goodman [11]:

$$\begin{aligned}
&s\frac{\mathrm{d}s}{\mathrm{d}t}+s^2\frac{\mathrm{d}T_c}{T_c-T_m}+\frac{k_s d(2-c_1)}{2k_c s+k_s d(2-c_1)}=2\frac{\alpha_s}{\alpha_l}\frac{6(1-c_1)}{(2+c_1)}\\
&\frac{2(T_m-T_\infty)}{H-s}=\frac{2}{3}\left(\frac{ds}{dt}(T_m-T_\infty)+\frac{dT_\infty}{dt}(H-s)\right)\\
&\frac{k_s}{k_l dv}\frac{2k_c s(T_m-T_c)}{2k_c s+k_s d(2-c_1)}\frac{-c_1}{s}+\frac{2}{H-s}=\beta\frac{\mathrm{d}s}{\mathrm{d}t}
\end{aligned} \tag{9}$$

Where s is thickness of freezing plate, d is thickness of fuel cladding, H is thickness of molten plate, the subscript c, m, ∞ are cladding, melting point and molten material respectively. Then distribution of freezing plate along axial outface of cladding can be solved by combining simulation of flow and calculation of freezing.

3. Program development of SCDAP/RELAP5 MOD3.1

The system code for severe accident analysis SCDAP/RELAP5 MOD3.1 has different core component for core modeling, and different core components have different geometry, composition and fuel behaviors during core degradation. It is concluded that all of the core components in S/R code can not be applied directly to analyze UO_2-Zr dispersion plate-type fuel by comparing structure and fuel behavior of UO_2-Zr plate with core components in S/R code, thus program development of S/R code is necessary and important. A new core component named "plate" is modeled to simulate UO_2-Zr plate under severe accident. The new component consists of a geometry block and a fuel behavior block. As showed in fig.7, the new component "plate" includes structures like fuel meat, fuel particles, fuel cladding, freezing plate, molten materials plate, flow path and so on.

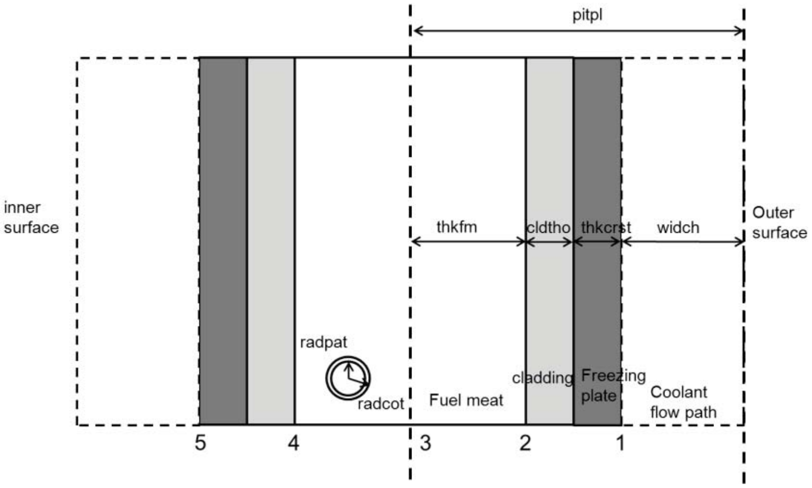


Figure 7 Geometry structure of core component “plate”

Fuel behavior block of the new components “plate” includes oxidation, solid reaction, fuel failure and relocation process. As showed in fig.8, subroutine “PLATEB” is the program of fuel behavior analysis for “plate”, and in this subroutine some subroutines are executed to investigate different fuel behaviors, such as “OXDPLATE”, “FMREAT”, “MESHPL”, “PLATMLC”, “HEATC1”, “MTFUL”, “PLATMLT” and so on. Some important subroutines and their functions are listed in Tab.2.

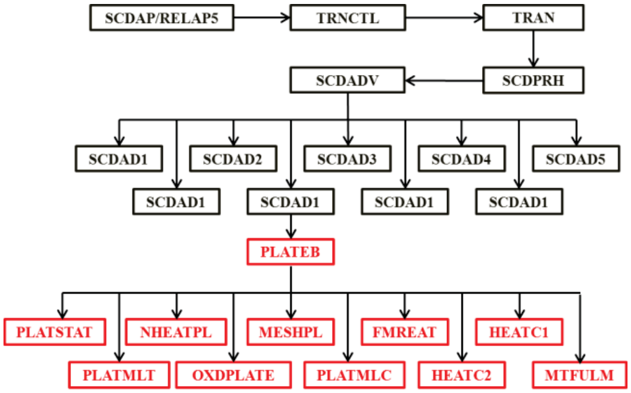


Figure 8 Code structure of subprogram related to “plate”

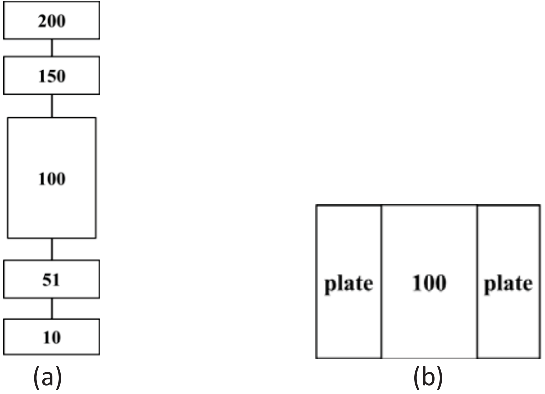
Tab.2 Function of subroutine executed by PLATEB

Subroutine	function
OXDPLATE	Simulation of Oxidation
FMREAT	Simulation of solid reaction in fuel meat
PLATMLT	Simulation of freezing process
MESHPL	Partition of radial node
NHEATPL	Relocation of decay heat
HEATC1	Calculation of temperature distribution

4. Severe accident analysis of core with UO_2-Zr plate

A simple core assembled with UO_2-Zr dispersion plate-type fuel is chosen to estimate the developed component “plate”. The core consists of a inlet pipe, a outlet pipe and a heat structure stand for core structure. The fuel in the core is UO_2-Zr dispersion plate-type fuel. The node scheme of core in

SCDAP/RELAP5 MOD3.1 is showed in fig.9, where component 10,51 model core inlet,. component 100 with ten axial nodes models core and component 150,200 model core outlet.



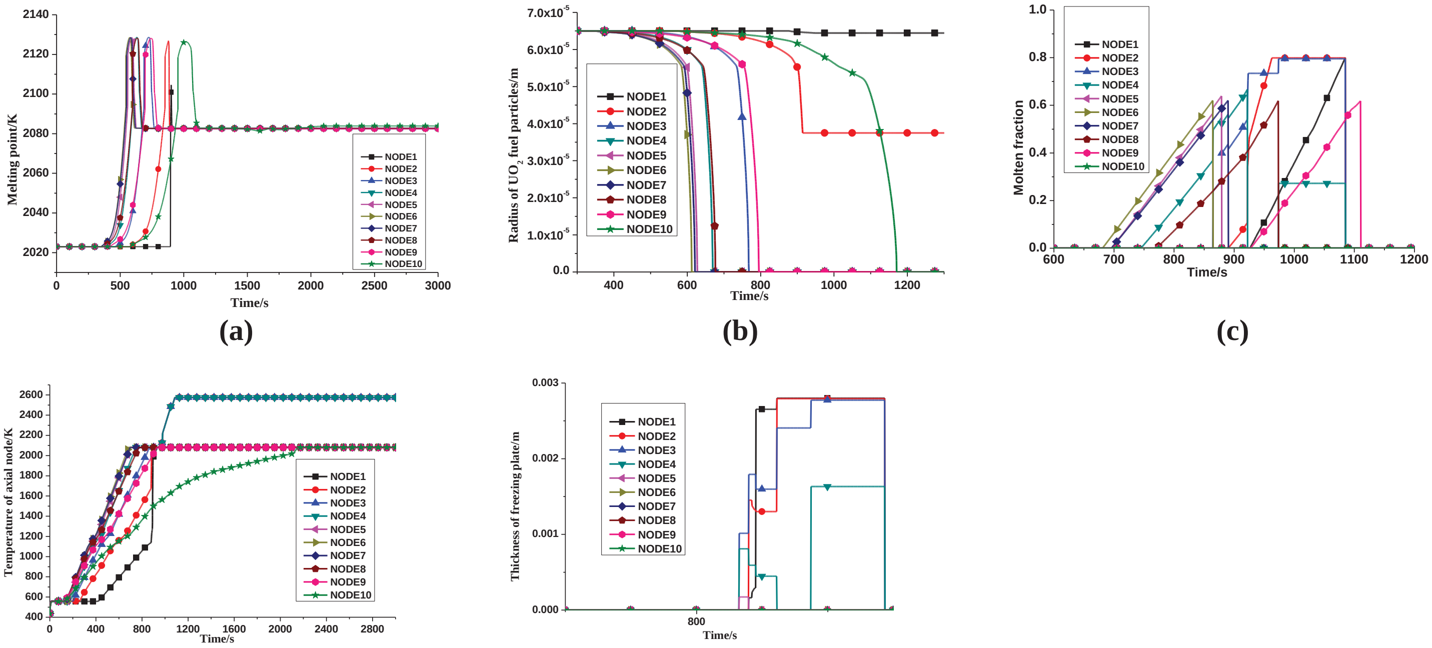
(a) (b)

Figure 9 Nodelization of core in S/R code

The loss of total flow accident is chosen to be the initial incident of severe accident. In S/R code, when flow mass from time dependent junction “51” is 0, a loss of total flow accident occurs. The core is heated up due to loss of heat removal, if there is no other heat removal approaches method injected, severe accident occurs and core degrades.

The calculation results are showed in fig.10. In Fig.10a, the melting points of fuel meat in every node increase at first and then decrease to a value as much as the others. Oxidation and solid reaction are the key reasons for melting point varying so much along time. Oxygen diffusion from UO_2 to fuel meat will increase melting point of fuel meat at early phase, but when UO_2-Zr solid reaction occurs, uranium atoms mixed into fuel meat will decrease melting point. Due to high atoms diffusion ability at high temperature, the change rate of temperature increases along time. In fig.10b, the radius of UO_2 fuel particles in every axial node decreases all over the accident, and decreasing rate is much larger at high temperature. The radius of UO_2 fuel particles turns to be zero at late phase which explains the reason why melting point nearly the same at the end. Thus, it is concluded that fuel meat will melt at temperature nearly 2080K and at that time UO_2 fuel particles in fuel meat have already been reacted.

When fuel meat in an axial node melts, molten materials drain into flow path and relocate at lower place of core. Thickness of freezing plate in every node is showed in fig.10c. Thickness is different between axial nodes and it is quite inconsistent with the core component “ATR” which has already existed in S/R code. Because heat source in fuel meat drains with molten materials, heat source from higher place relocates at lower place and accelerates fuel melting process. As showed in fig.10d, the fuel melting rates (slope of molten fraction) of lower nodes increase because of molten materials relocation of higher nodes. Molten materials freeze at lower place of core and cause a core blockage, then molten materials blocked above will be heated up and melt control rods which have higher melting point. In fig.10e, molten materials relocate and freeze along axial coordinate at early phase, but when there is a core blockage at lower place, molten materials at node 2, 3, 4 are heated up and temperature exceeds the melting point of control rods, then control rods melt and mix into molten materials. It is quite different from the former research on UAl plate-type fuel. the melting of control rods decrease risk of re-criticality during severe accident.



(a) (b) (c)

(d) (e)

Figure 10 Calculation results of the developed S/R code: a) Melting point; b) Radius of fuel particles; c)Molten fraction; d)Temperature; (e) Thickness of freezing plate

5. Conclusion and prospect

Because of structure and composition properties of UO_2-Zr dispersion plate-type fuel, oxygen diffusion from UO_2, solid reaction in fuel meat and molten fuel meat relocation mixed with fuel particles at low temperature occur during core degradation. According to former studies, models of oxidation, solid reaction in fuel meat and relocation of molten fuel meat are developed in the paper. The new models can supply theoretical elements for safety analysis of core assembled with dispersion plate-type fuel under severe accident.

Based on system code SCDAP/RELAP5 MOD3.1, the new models are programmed into a new core component “plate” including special geometry definition and fuel behavior sequence. Some special phenomenon such as dissolving of UO_2 fuel particles into fuel meat before melting of fuel meat, melting of fuel meat at low temperature, a accelerated sequence of core degradation and a core blockage which could induce an molten pool upward is investigated by the developed S/R code. The study in this paper is a preliminary work on severe accident analysis of core assembled with dispersion plate-type fuel, Studies on interactions between different core section and other fuel failure modes will be performed at next stage.

ACKNOWLEDGMENTS

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