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THERMAL PERFORMANCE PREDICTION OF FUEL PIN CONTAINING U-9Mo/Zr-ALLOY PRODUCED BY CAPILLARY IMPREGNATION TECHNIQUE FOR HIGH BURNUP PWR

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ABSTRAK

PREDIKSI KINERJA TERMAL PIN BAHAN BAKAR U-9Mo/PADUAN Zr YANG DIHASILKAN OLEH TEKNIK IMPREGNASI KAPILER UNTUK REAKTOR AIR TEKAN BERDERAJAT BAKAR TINGGI. Akhir-akhir ini, telah dikembangkan sebuah jenis paduan Zr baru yang mempunyai titik leleh 650-860°C. Berdasarkan paduan-paduan matriks Zr baru, telah dikembangkan pin bahan bakar U-9Mo berdaya muat uranium tinggi dengan teknik impregnasi kapiler. Pin ini mempunyai konduktivitas termal dari 18 sampai dengan 22 W/m/K yang lebih tinggi jika dibandingkan dengan pin pelet U-9Mo. Makalah ini menyajikan fabrikasi dan analisis kinerja termal dalam reaktor air tekan berderajat bakar tinggi. Proses fabrikasi meliputi pencampuran serbuk atau granul (butiran) U-9Mo dengan serbuk paduan Zr baru yang mempunyai titik leleh rendah, pengisian campuran ke dalam tabung kelongsong yang salah satu ujungnya sudah ditutup, pemanasan pin dengan suhu di atas titik leleh paduan Zr selama satu jam, pendinginan alamiah, dan perlakuan panas pada suhu 300K selama ½ jam. Analisis termal mempertimbangkan distribusi pori dan suhu, dan pengaruh derajat bakar tinggi pada konduktivitas pelet. Rasio difusivitas termal bahan bakar jenis baru dengan bahan bakar konvensional dijadikan sebagai faktor koreksi untuk konduktivitas termal bahan bakar jenis baru. Hasilnya menunjukkan penurunan suhu pelet yang cukup signifikan sepanjang radius pin hingga 1000K pada posisi yang paling panas. Analisis ini memberikan perkiraan rendah karena konduktivitas celah dianggap menurun dengan 2% gas hasil belah dilepas. Kenyataannya, penggunaan suhu rendah dan penurunan konduktivitas termal oleh pembentukan porositas akan semakin jauh menurun nilainya. Analisis juga menunjukkan bahwa bahan bakar jenis baru ini mempunyai sifat-sifat termal yang sangat baik yang mampu melampaui batas 65 MWD/kg-U yang merupakan batas bahan bakar komersial yang ada saat ini. Perpanjangan derajat bakar berarti lebih sedikit bahan bakar baru diperlukan untuk menghasilkan listrik, melestarikan sumber uranium alam, dan kemudahan di dalam penanganan operasi bahan bakar untuk setiap energi yang dihasilkan.

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KATA KUNCI : met-met fuel, capillary impregnation, thermal performance, PWR fuel.

ABSTRACT

THERMAL PERFORMANCE PREDICTION OF FUEL PIN CONTAINING U-9Mo/Zr-ALLOY PRODUCED BY CAPILLARY IMPREGNATION TECHNIQUE FOR HIGH BURNUP PWR. *In recent years, a novel class of zirconium alloys having the melting temperature of 650-860 °C has been developed. Based on novel zirconium matrix alloys, high uranium content fuel pin with U-9Mo has been developed using capillary impregnation technique. The pin has a thermal conductivity ranging from 18 to 22 (w/m/K). It is comparably higher than U-9Mo pellet pin. The paper presents the met-met fabrication and thermal performance analysis in high burn-up PWR. The fabrication consists of mixing U-9Mo powder or granules and a novel Zr-alloy powder having low melting point, filling the mixture in a cladding tube in which one of its end has been plugged, heating the pin to above the melting temperature of Zr-alloy for an hour, natural cooling and heat treating at 300 K for ½ hr. The thermal analysis takes into account the pore and temperature distribution and high burn up effect on pellet conductivity. The thermal diffusivity ratio of novel to conventional fuel has been used as a correction factor for the novel fuel thermal conductivity. The results show a significant lowering of pellet temperature along the radius until 1000 K at the hottest position. The analysis gives an underestimate since the gap conductivity has been treated as to decrease by 2% fission gas released, which is not real since the use of lower temperature, and also decreasing thermal conductivity by porosity formation will much lower the value. The analysis shows that the novel fuel has very good thermal properties which are able to pass the barrier of 65 MWD/kg-U, the limit to today commercial fuel. The burn-up extension means less fresh fuel is needed to produce electricity, preserve natural uranium resource, easier fuel handling operational per energy produced.*

FREE TERMS : *met-met fuel, capillary impregnation, thermal performance, PWR fuel.*

I. INTRODUCTION

The PWR/VVER core comprises an array of square/hexagonal fuel assemblies which are similar in mechanical design, but different in fuel enrichment. The enrichment of the various groups of fuel assemblies is varied in order to flatten the power profile in the core. The typical core is cooled and moderated by light water at a pressure of about 155 MPa; nominal full power inlet and outlet temperatures are about 288 and 327 °C, respectively. In general, there are five types of PWR fuel assemblies which are 14 x 14, 15 x 15, 16 x 16, 17 x 17 and 18 x 18 square of fuel rods array.

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Oxide fuel has been used in most commercial power plants. However, its thermal conductivity is very low, which limits its performance. The performance is also limited by other phenomena such as pellet cracking, densification, swelling, resulting cladding stress and corrosion. Higher grains size with 5% pore need a dopant for higher creep fuel, and helium fill gas for higher gap conductance.

Sintered UO_2 exhibits a very stable fuel in reactor core and has been used as LWR and BWR fuel for more than 40 years, with continuous improvement in burn-up from only 28 MWD/kg-U up to 62 MWD/kg-U.

With respect to the total fuel cycle costs, a decrease of nearly 50% over the entire burn-up range considered. Depending on the commercial boundary conditions, a one percent reduction of the fuel cycle costs corresponds to up to one million Euros per GW reactor/year[1]. This represents an enormous saving in fuel cycle costs.

To achieve such progress development of UO_2 pelletized fuel, many fuel and cladding parameters have been studied for improvement of: pellet geometry, pellet grains size, burnable poison, and porosity. Pellet geometry including diameter, height, and chamfer which influences its dimensional change and its mechanical interaction to the cladding.

Met-met and cer-met fuels have been known for high thermal conductivity, but it had been abandoned in early 1960 because of fabrication difficulty. In the first decade of century however, a new met-met fuel has been developed with new matrix alloy and fabrication technique. A novel class of zirconium alloys having a melting temperature of 690-860 °C has been developed[2]. It is found that due to their capillary properties they might be applied in brazing dissimilar materials. Based on novel zirconium matrix alloys, high uranium content fuel comprised of U9Mo has been developed using capillary impregnation technique. The pin shows it is thermal conductivity ranging from 18 to 22 W/m/K, it is much higher compared to 2-4W/m/K of UO_2 pellet.

Dispersive fuel has been used in many MTR, in different shapes: flat plate, curved plate, and cylindrical bar and different fuel such as metallic, compound and alloy. U-Mo in Al-6Si matrix, for lower temperature reactors has been developed more than 10 years in Korea, US, France, Russia, Argentine, etc[3].

Dispersion fuel of UN in Al matrix has been developed for outer space power reactor[4]. Development of dispersion fuel for conventional PWR which

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has been abandoned at early commercial PWR, in recent years come back as a low melting point novel Zr alloy has been invented.

The objectives of the present paper are to evaluate an advanced fabrication of met-met fuel pin and to predict the pin thermal performance. The related study is intended to provide some considerations in fuel development for Indonesia.

Fuel Pin Fabrication based Capillary Impregnation

The novel met-met fuel rod is fabricated by capillary impregnation. The fabrication consists of preparation of fuel granules, preparation of matrix powder and fabrication of cladding with welded up plug, as represented in Figure-1. Fuel and matrix granules are fabricated by melting-solidification and granulation and fabrication of matrix, and loading the powders into the cladding, by vibration, then capillary impregnation quality control and then sealing the fuel pin top end.

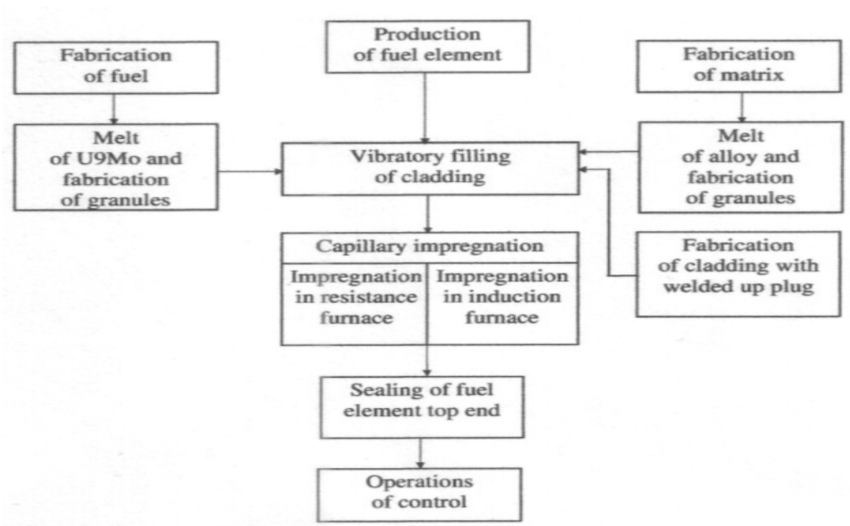

Figure-1. Process Fabrication Flow Diagram for Pin Fuel of met-met type[2].

Fig.2 shows cross sectional pin filled with fuel granules(2) and zirconium alloy matrix (3) powder in zircaloy cladding (1) by vibro-loading technique(b-left), and Fig.3 (b-right) shows when melting-solidification Zr alloy powder have taken place by capillary impregnation where formation of '*bridges*' (4) between matrix alloy coats on fuel granules and between granules and cladding after heating and cooling, and pores (5). The pore should accommodate fission gases and solid swelling during irradiation/burning in reactor. Fig.3(c) shows typical macrograph of met-met (U-9Mo /Zr-alloy) fuel pin fabricated by capillary impregnation method containing U9Mo granules 0.8 mm average diameter, pore ~ 18%v. Geometrics conditions: Cladding diameter 5.8 mm, thickness 0.5 mm, fuel granules 0.6-1.2 mm, matrix granules 0.15-0.5 mm in (a) and 0.06-0.2 mm in (b).

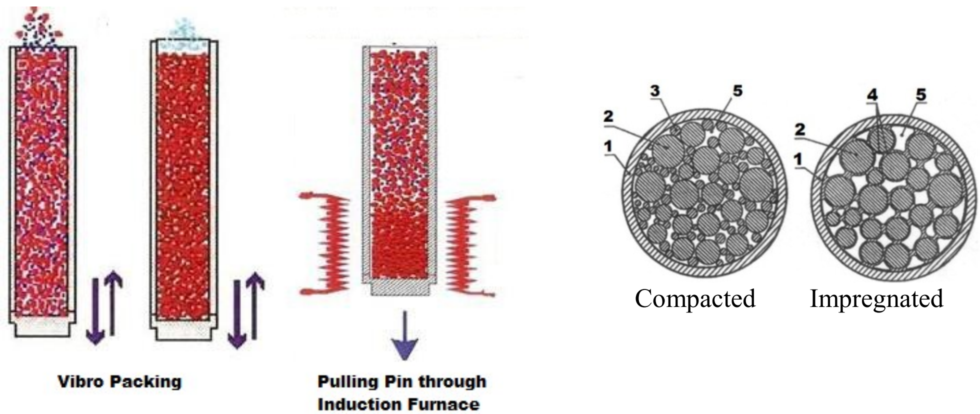

Figure-2. Schema of vibratory filling and Capillary Impregnation Technique (left) and schematic cross-section pre and post impregnation (right)[5].

II. METHOD

The analysis has been accomplished by bi-dimensional axy-symmetric approach and the outer boundary was the outer surface of cladding. It has been given same temperature for both fuel system, and burn-up of 80000 MW.D/kg. The effective thermal diffusivity has been used in computation of novel fuel, where as for computation on pellet in cladding rod fuel type the computation has taken into account of gap, and pellet that is depend on temperature, the distribution of both pore size and volume fraction since this has strong influence. The algorithm used for analyzing the met-met fuel is same as one used for analyzing pellet fuel rod containing partly tungsten metal network[4].

The temperature distribution has been obtained by applying a simple model of steady-state heat transfer of fuel rod at particular level of high burn-up. Calculation has been done by typical models of pore distribution and gap conductance at high burn-up. The model takes into account thermal properties dependent of pellet to temperature, pore, and burn-up. The procedure consisted

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of the following steps. Pore distribution at evaluated high burn-up has been modeled by fitting typical measured pore distribution at high burn-up and by Halden model. ANS recommendation has been chosen for temperature dependent of pellet conductivity, and for cladding thermal conductivity by. Pore contribution to pellet thermal conductivity is taken into account as multiplication factor for 95% TD fresh fuel. The outer boundary condition has been fixed temperature of outer surface of cladding for both two fuels type. The temperature of cladding surface has been determined without taken into account thermal conductivity of CRUD. The gap conductance has been calculated as given parameter since it depends on fuel design. The radial power distribution is modeled as a linear combination function obtained by fitting of typical experimental data of power depression. Rod heat transfer in the axial direction has been omitted. The radial space is discretized into nr element of linear LaGrange type. The heat transfer equation in the fuel pellet has been approached by a combination of finite element and finite differences of Saturn-FS1[4]. Mathcad has been used for implementation of the algorithm on a personal computer.

The dependent of pellet thermo-physical properties has been modeled by U-9Mo/Zr of 75% TD, MATPRO properties. The pore distribution has been modeled by curve fitting of experimental measurement. Correction factor is chosen for taking into account the porosity effect and its distribution, a combination piece wise using an empiric curve fit.

The thermal conductivity of non-irradiated fuel is obtained by logarithmic fitting of Savchenko data, with 0.999 coefficient of correlation[2]:

$$ThC(Tk) = 25.29.\ln(Tk)-148.1 \qquad (1)$$

where ThC is the fuel thermal conductivity and Tk is temperature in K.

The irradiation effect on thermal conductivity is taken into account by correction factor of pore that depends on pore and a coefficient depending temperature. The measured thermal conductivities were normalized to the values of 96.5%TD (TD: theoretical density) by using the Loeb's equation:

$$\lambda_n=\lambda_m(1-0.035\varepsilon)/(1-\varepsilon P) \qquad (2)$$

where: λ_n is the thermal conductivity normalized to that of 96.5%TD; λ_m, the measured thermal conductivity; ε, the parameter which express the effect of

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pore shape on the thermal conductivity of pellets; is P, the porosity evaluated from the sample density. The parameter pore factor is expressed as follows[3]:

$$\varepsilon = 2.6\text{-}5\times10^{-4}(T(K)\text{-}273.15), \tag{3}$$

$$P = 1 - TD \tag{4}$$

The last two models may be unified as correction factor of Waisenak

$$Fp = (1\text{-} \varepsilon.P) / (1\text{-}0.05 * \varepsilon) \tag{5}$$

$$T=Tc \tag{6}$$

$$K = K_{95\%TD} \cdot (1-\beta p)/(1-0.05\cdot\beta) \qquad (W/mK) \tag{7}$$

$$\beta = 2.58 - 0.58\times10^{-3}.T \tag{8}$$

MatPro v 9.0 model of temperature dependent of thermal conductivity of fresh/un-irradiated UO_2 fuel has been chosen[6].

For: $0 < T < 1650\ ^{o}C$ etc

The (qv(r)) is volumetric power density profile according to radial coordinate, be modeled as polynomial eq.10 that is fit of typical power distribution.

$$qv(r) = p\ .\ v(r) \tag{9}$$

For high burn-up constant p and variable v are vectors of:

$$p = \begin{pmatrix} 0{,}373 \\ 0{,}22 \\ 0{,}410 \end{pmatrix} \quad \text{and} \quad v(x) = \begin{pmatrix} 1 \\ x^{12} \\ x^{24} \end{pmatrix} \tag{10}$$

The correlation between linear power density / LHGR qr(r) and volumetric power density qv(r) is:

$$qr(r) = 2.\pi.\int r.qv(r).dr \tag{11}$$

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The analysis is underestimates since the gap conductivity has been treated as decreased by 2% fission gas released, that is not really since the use of lower temperature, and also decreasing thermal conductivity by porosity formation will much lower.

A finite element approach is applied for the radial distribution of fuel temperature. The radial space is discretized into nr element of linear LaGrange type. Fuel temperature in each element is defined as according to SATURN-FS1 algorithm, that gives a solution of pellet temperature as eq.12

$$T_k = T_{k+1} + D_k \cdot \frac{Qv}{\lambda\ (T(r), por(r))} \cdot \left[Qv.R.\{D_{nR-i} \cdot (AA_i + A_{nR-i}.pm) + BB\}\right] \qquad (12)$$

where Q, R, A, B, AA, BB, F and G are numerical variable mentioned some where[4, 7]

III. RESULT AND DISCUSSION

Table 1 shows the fuel specification of metal alloy and Figure 3 shows plot of thermal diffusivity data from room temperature to 1500 ^oC for UO_2 pellet (noted UO_2 square) and for UO_2 pellet containing tungsten network (noted UO_2 diamond). The data for has been obtained by using the same measurement method.

Table 1. Metal alloy fuel specification

Designation	Fuel composition	Volume fraction of fuel, %	Volume fraction of matrix, %	Porosity of meat, %	Density of loaded granules, %	Uranium content, g/cm^3
1	2	4	5	6	7	8
C-15	U-9Mo+Zrl0Fel0Cu	62.08	15.18	22.75	77.26	9.53
C-26	U-9Mo+Zr8Fe8Cu	64.56	18.04	17.39	82.60	9.91
C-27	U-9Mo+Zr-8Fe-8Cu	9.15	64.21	20.10	15.69	84.31

The volume fraction of composite fuel of U-9Mo / Zr-8Fe-8Cu: fuel / matrix / pore = 64.56 / 18.04 / 17.39. Its thermal conductivity has been measured at 4 different temperatures. The correlation has been fitted by cubic polynomial, and has been used for the analysis. Figure 3 shows the conductivity of the met-met fuel compared to UO_2 pellet 95% theoretical density both in fresh condition. The figure shows that although the met-met fuel has greater porosity (17.39%) than UO_2 (5%) the thermal conductivity of met-met fuel other than higher it increases rapidly with temperature, contrary the UO_2 pellet conductivity is very low and decreases with temperature.

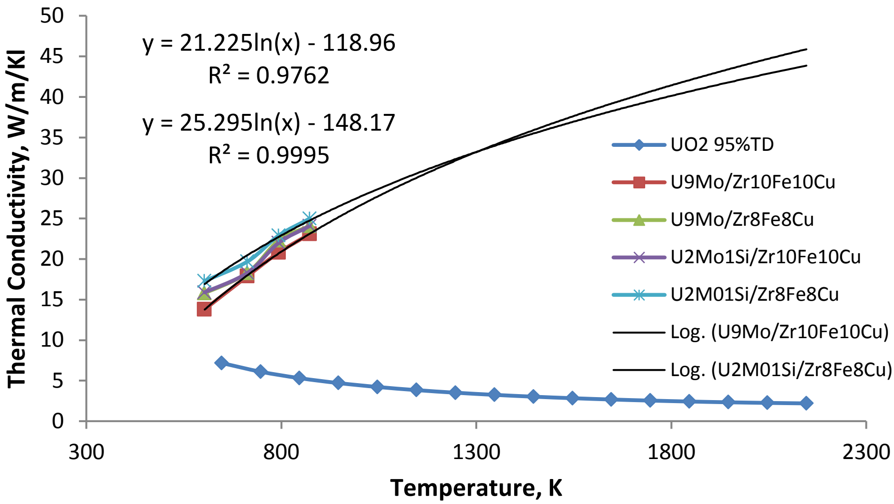

Figure-3. Thermal Conductivity of met-met U-Mo/Zr-Fe-Cu[5] and UO_2 pellet[6].

The choice is one that gives coefficient of correlation of 0.999. It is relatively pessimistic for calculated temperature, but it is safer, more conservative. When temperature attains the melting point of the matrix, the thermal conductivity of matrix increases rapidly, also the composite since the pore volume decreases. In view of neutron reactivity, it is needed a detailed analysis in correlation to Doppler effect.

The thermal analysis of fuel comprising 9%w of tungsten network has been carried out by using typical data of UO_2 pellet and the thermal conductivity has been calculated by applying thermal conductivity ratio to correct the new fuel conductivity. Pellets partially containing tungsten network also have been analyzed. The result is presented in Fig.4.

Met-met fuel can be loaded ~1.25 higher than UO_2 pellet. The center line temperature of met-met fuel is much lower than UO_2 pellet, it is 380-500 ^oC for met-met and 1300-1700 ^oC for UO_2 fuel.

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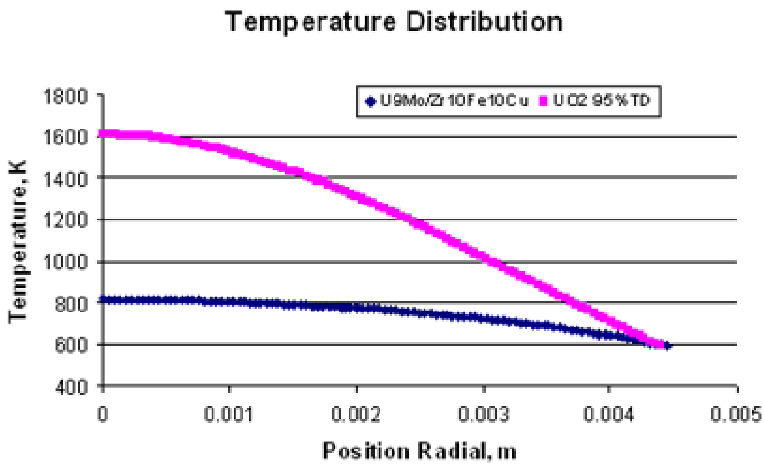

Figure 4. Comparison of radial temperature distributions of UO_2 pellets (square) and U9Mo/Zr10Fe10Cu fuel (diamond), LHGR 47 kW/m, burnup 40 MWd/kgU.

Fig.3 shows plot of thermal conductivity data from Tab.1 from room temperature to 1500 ^{o}C for UO_2 pellet (noted UO_2 square) and for UO_2 pellet containing tungsten network (noted UO_2 diamond). The upper curve in Fig.3 shows the radial temperature distributions of UO_2 pellet for and the lower curve for new pin containing met-met fuel of U-9M0/Zr-10Fe-10Cu in normal operating condition at high burnup. The temperature of fuel pin containing pellets significantly higher than met-met fuel. The difference attains its maximum in center-line of pin, it appear about 800 K.

The maximum elevation temperature of met-met fuel only about 200 K, while pelletized UO_2 fuel elevation temperature is about 1000 K in the same distance of 0.0045 m.

In addition of temperature limit of fuel in fuel safety criteria, a lower pellet temperature reduces the mobility of the fission gases in the fuel and thereby lowers the rate at which fission gases are released. The lower overall heat content of pellets with an increased thermal conductivity improves the fuel assembly performance under accident conditions (LOCA and RIA) by lengthening the time before the fuel assembly is destroyed. A lower central temperature with otherwise identical fuel properties also reduces what is known as the hour-glass effect, which has an adverse effect on the pellet cladding interaction (PCI) properties of a pellet. It seem the potential use of the new pellet that may change the performance of fuel.

Result of applying tungsten network of 6-9w% in side UO_2 pellet[9] attains ~300 ^oC of lowering maximum temperature of pellet. Result obtained by Tulenko et al.[10] for improving thermal performance of fuel rod by applying metal liquid bond between pellet and cladding for 6 kW/ft ~2 kW/m power rating is showed a lowering temperature around 350 ^oC[9]. The last two techniques give roughly comparable result. Meanwhile, met-met fuel utilization allows lowering maximum temperature of fuel pin nearly three folds of them.

There are many experimental and theoretical researches on U-Mo alloys as novel fuel for testing material and research reactor. In the first stage U-Mo alloy was dispersed in Al, resulting thicker interaction layer of uranium aluminide which is more porous, lower thermal conductivity, and higher swelling[11,12]. Many techniques have been developed to avoid the problem.

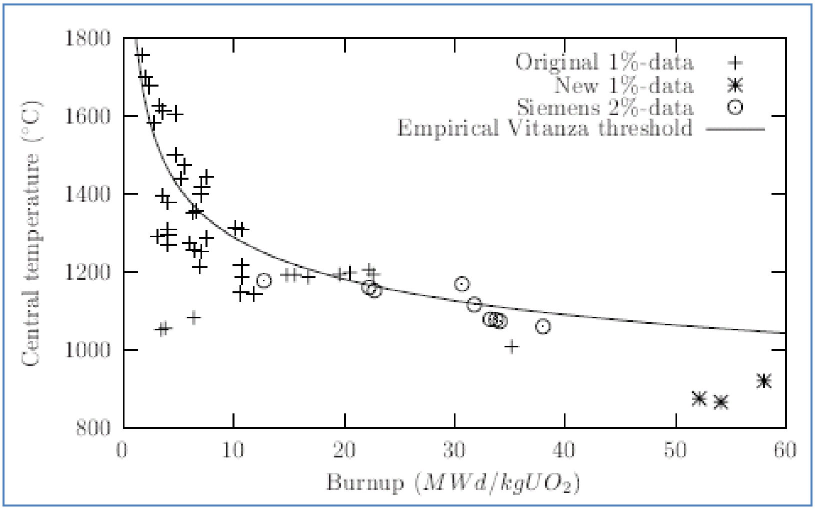

Figure-5. Radial distribution of Temperature of Fission gas start (1-2 %) released[13].

Met-met fuel (60%v fuel, 14%v matrix and ~20%v pore has 20-25 % uranium content than UO_2. The good metallurgical contact between met-met and internal cladding surface allows the fuel serviceable in power transient conditions.

On-set release on fission gas after the Vitanza high burnup appear at temperature above 1800 ^oC at low burnup, at about 60% burnup the threshold temperature of gas release is only ~1000 ^oC. All burnup regime the temperature

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threshold for gas release (out of pellet) 800 ^{o}C as presented in Fig.5. It is potentially the met-met fuel free of releasing fission gas.

Fuel crack and deformation such hour-glassing, fragment bending, radial and axial cracking that occur in current fuel and limits further irradiation may be avoided by the novel fuel.

Gas released from different location tends to diffuse to the lower activity related to lower concentration region that is pellet-cladding gap. The addition of fission gas to tends to decrease, so the temperature of pellet tends to increase. Generated fission gas increases sharply in the surface. It is related to the self shielding effect of moderated neutron and Pu reaction by epy-thermal neutron. The gas diffused in the matrix, precipitated as gas bubble inside and on grain boundary. On the outer pin region of r > 80% the distribution radial may be easily precipitated, grain boundary.

IV. CONCLUSION

Thermal performance of met-met fuel in novel Zr-alloy matrix has been carried out for steady state at high burnup. The thermal conductivity used for analysis is based to Savchenko work, fitting the data and extrapolation. The novel met-met fuel permit reduction up to 800 K that is about triple of result by applying tungsten network inside UO_2 pellet, or applying liquid metal bounding between pellet and cladding.

The maximum temperature of met-met fuel ~800 K is much lower than its melting point of matrix (1200 K). The thermal conductivity of met-met fuel rise with rising temperature is oppositely to UO_2 pellet. It is good properties for safety.

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