

ADVANCED FABRICATION TECHNIQUE AND THERMAL PERFORMANCE PREDICTION OF U-MO/ZR-ALLOY DISPERSION FUEL PIN FOR HIGH BURNUP PWR

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Abstrak

In recent years, a novel class of zirconium alloys having the melting temperature of 990-1160 K has been developed. Based on novel zirconium matrix alloys, high uranium content fuel pin with U-9Mo has been developed according to capillary impregnation technique. The pin shows its thermal conductivity ranging from 18 to 22 W/m/K that is comparably higher than UO₂ pellet pin. The paper presents the met-met fabrication and thermal performance analysis of the fuel in typical PWR. The fabrication consists of mixing UO₂ powder or granules and a novel Zr-alloy powder having low melting point, filling the mixture in a cladding tube that one of its end has been plugged, heating the pin to above 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 to pellet conductivity. The thermal diffusivity ratio of novel to conventional fuel has been used as correction factor for the novel fuel conductivity. The results show a significant lowering pellet temperature along the radius until 1000 K at the hottest position. The analysis underestimates since the gap conductivity has been treated as decreased by 2 % fission gas released that is not real since the use of lower temperature, and also decreasing thermal conductivity by porosity formation will much lower. The analysis shows that the novel fuel has very good thermal properties which able to pass the barrier of 65 MWD/kg-U, the limit to day 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.

Keywords: met-met fuel, capillary impregnation, thermal performance, PWR fuel. PACS:

1. 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 PWR fuel assemblies such as 14 x 14, 15 x 15, 16 x 16, 17 x 17 and 18 x 18 square of fuel rods array. Oxide fuel has been used in most commercial power plant. However, its thermal conductivity is very low, that limits its performance. The performance is limited by many phenomena as pellet crack, densification, swelling, resulting cladding stress and corrosion. Higher grain size 5% pore needs dopant for higher creep fuel, and Helium fill gas for higher gap conductance.

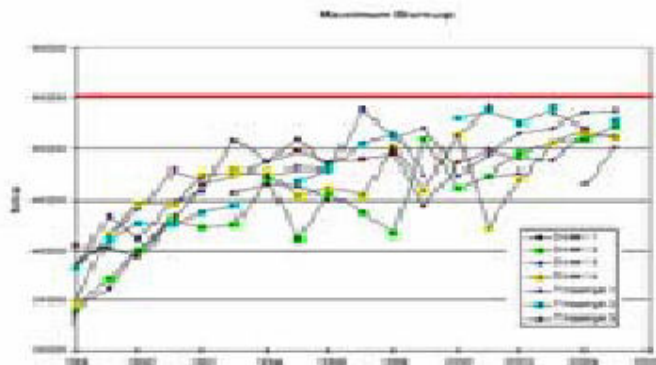


Figure 1. Increasing discharge burn-up Belgian NPP

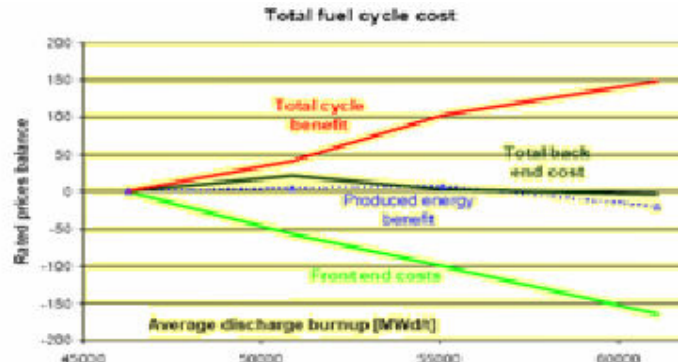


Figure 2. Benefit of increasing fuel burn-up of OECD countries

Sintered UO₂ exhibits very stable in reactor core and has been used as LWR and BWR fuel for more than 40 year, with continuous improvement from only 28 MWD/kg- U climbed to 62 MWD/kg-U. Figure 1 shows history of fuel development in term of maximum discharge burn-up of Belgian nuclear power reactors (1988 – 2006). The energy extracted from fresh fuel has been doubled during 20 years. Meanwhile, Fig.2 shows the economic benefit of increasing discharge burn-up is presented in figure 2. It shows fresh fuel or front cycle cost decreases linearly with burn-up as needed fresh fuel decrease with increasing discharge burn-up. With respect to the total fuel cycle costs, the figure shows a decrease of nearly 50% over the entire burn-up range considered. As one percent reduction of the fuel cycle costs corresponds, it depends on the commercial boundary conditions, to up to one million euro per 1300 MWe reactor/year [1]. This avoidance represents an enormous saving in fuel cycle costs. Met-met and cer-met fuel has been known for high thermal conductivity, but it had been abandoned in early 1960 because of fabrication difficulty. In the millennium 3 however, a new met-met fuel has being developed by new matrix alloy and fabrication technique. A novel class of zirconium alloys having the melting temperature of 690-860°C has been developed [2]. It is found that thanks 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 according to capillary impregnation technique. The pin shows its thermal conductivity ranging from 18 to 22 W/m/K; it is much higher compared to 2-4W/m/K of UO₂ pellet. To achieve such progress development, many fuel and cladding parameters has been studied for improvement of: pellet geometry, pellet grains size, burnable poison, and porosity. Fuel geometry including diameter, height, and chamfer affects the growth of reaction.

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. Some examples are metal, alloy, inter metallic as: U₃O₈, UAl_x, U₃Si, U₃Si₂, U-Mo in aluminum matrix, for lower temperature reactor [2]. Dispersion fuel of UN in Mo tungsten or matrix has been developed for outer space power reactor [3].

Development of dispersion fuel for conventional PWR which has been abandoned at early commercial PWR, in recent years come back as a low melting point novel Zr alloy has been invented. The objective of the present paper is evaluating an advanced fabrication of met-met fuel, providing thermal performance evaluation of the novel product and process. The related study is intended to provide some considerations in fuel development for Indonesia. The paper includes long development and utilization in commercial plant, computing / prediction of the new fuel thermal performance and its comparison to the fresh fuel for the fuel of existing technology. The geometry of the analysis is simplifies to bidimensional axe-symmetric problem and the outer boundary was the outer surface of cladding. Conventional UO₂ pelletized fuel rod. The fuel rods are plugged and seal welded at the ends to encapsulate the fuel pellets, and are pressurized with helium to minimize clad creep down and improve heat transfer inside the rod. The fuel pellets are right circular cylinders fabricated from slightly enriched uranium dioxide powder that has been compacted by cold pressing and then sintered to the required density. The nominal density of fuel pellets is 95% of the theoretical density. This density has been found to provide adequate void volume to keep irradiationinduced swelling to reasonable values, simultaneously providing a stable structure resistant to densification. The ends of each pellet are dished slightly to allow for greater axial expansion at the center of the pellets. In some designs, the pellets are also chamfered at the ends to improve resistance to pellet chipping during pellet and rod manufactures and to reduce the potential for stress-induced failures. To avoid overstressing the cladding or the seal welds, pellet-to-clad clearances and gap plenums are provided within the rods to accommodate fission gas released: 1) fission gas released from the fuel; 2) differential thermal expansion between the cladding and fuel pellets; and 3) fuel pellet swelling during burn-up. Shifting of the fuel pellets within the cladding during handling or shipping prior to core loading is minimized by an internal stainless steel helical spring that bears on the top of the fuel pellet stack.

Advanced novel fuel pin fabrication. The novel metmet 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 flow-chart represented in figure-3(a). 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.

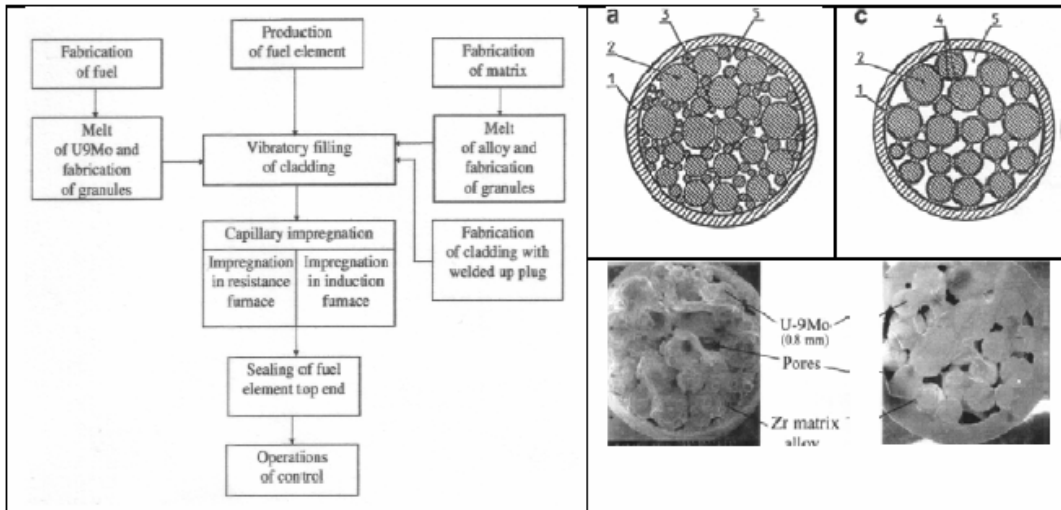


Figure-3. Fabrication by capillary impregnation method of dispersive fuel pin(a), Schematic cross-section of before (b left) and after impregnation (b-right), and Macrograph of fractured met-met fuel pin of 3.b (c)[2]

Figure 3(a) shows schematic section representation of fuel element fabrication by capillary impregnation method. Figure 3(b) shows cross sectional pin filled with fuel granules(2) and zirconium alloy matrix (3) powder in Zr alloy 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%. Geometrics conditions: Cladding diameter 5.8 mm, thickness 0.5 mm, fuel granules 0.6-1.2 mm, matrix granules mm in (a) and 0.06-0.2 mm in (b).

2. METHOD OF THERMAL PREDICTION

The analysis has been accomplished by bidimensional axe-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 80 MWd/ton. 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 [3]. The analysis takes into account the correction factor for pellet conductivity related to both pore and temperature distribution and high burn up effect. The results (47 kW/m; 40%burn-up) show temperature decrease in all position of fuel. The thermal conductivity changes of irradiated UO₂ pellets may be related to different phenomena such irradiation-induced point defects, fission products and irradiation-induced micro bubbles. 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 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 UO2 of 95% 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 fitting. The thermal conductivity of non-irradiated fuel is obtained by logarithmic fitting of Savchenko data, with 0.999 coefficient of correlation:

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

where ThC is the dispersive 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:

$$\tilde{\epsilon}n = \tilde{\epsilon}m(1 - 0.035 \hat{a}) / (1 - \hat{a} \cdot P) \quad (2)$$

where: $\tilde{\epsilon}n$ is the thermal conductivity normalized to that of 96.5%TD; $\tilde{\epsilon}m$, is the measured thermal conductivity; \hat{a} , the parameter which express the effect of pore shape on the thermal conductivity of pellets; P, the porosity evaluated from the sample density. The parameter pore factor, Fp , is expressed as follows [5]:

$$\hat{a} = 2.6 - 5 \times 10^{-4} (T/K - 273.15) \quad (3)$$

$$P = 1 - TD \quad (4)$$

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

$$Fp = (1 - \hat{a} \cdot P) / (1 - 0.05 \times \hat{a}) \quad (5)$$

$$T = Tc \quad (6)$$

$$K = Kmn(1 - \hat{a} \cdot p) / (1 - 0.05 \times \hat{a}) \text{ (W/m/K)} \quad (7)$$

$$\hat{a} = 2.58 - 0.58 \times 10^{-3} \cdot T \quad (8)$$

MatPro v 9.0 model of temperature dependent of thermal conductivity of fresh/un-irradiated UO2 fuel has been chosen.

The volumetric power density profile according to radial coordinate $qv(r)$ has been modeled as polynomial eq.9 – 10. It is fit of typical power distribution.

$$qv(r) = p \cdot v(r) \quad (9)$$

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

$$p := \begin{pmatrix} 0.373 \\ 0.22 \\ 0.418 \end{pmatrix}, \quad v(x) := \begin{pmatrix} 1 \\ x^{12} \\ x^{24} \end{pmatrix} \quad (10)$$

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

$$qr(r) = 2 \cdot \pi \cdot \int r \cdot qv(r) dr \quad (11)$$

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 (radial nodes number) 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

$$T_k = T_{k+1} + D_k \cdot \frac{Qv}{\lambda \cdot (T(r), por(r))} \cdot [Qv \cdot R \cdot [D_{nR-1} \cdot (AA_1 + A_{nR-1} \cdot pm) + BB]] \quad (12)$$

where Q, R, A, B, AA, BB, F and G are numerical variable mentioned some where [6,7]

3. RESULT AND DISCUSSION

Figure 4 shows plot of thermal conductivity data from room temperature to 1500 oC for UO₂ pellet (noted UO₂ square – red) and for UO₂ pellet containing tungsten network (noted UO₂W diamond-blue). The data for Figure 5a has been obtained by using the same measurement method [8].

Table 1. Metal alloy-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	U9Mo+Zr10Fe10Cu	62.08	15.18	22.75	77.26	9.53
C-26	U9Mo+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 met-met fuel C26 characteristic is composed of U-9Mo alloyed uranium particles dispersed in Zr-8Fe- 8Cu alloyed zirconium matrix having volume fraction of 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 4 shows the conductivity of the met-met fuel compared to UO₂ pellet 95% theoretical density both in fresh condition. The figure shows that although the met-met fuel has greater porosity (17.39%) than UO₂ (5%) the thermal conductivity of met-met fuel is higher. It increases rapidly with temperature, contrary the UO₂ pellet conductivity is very low and decreases with temperature.

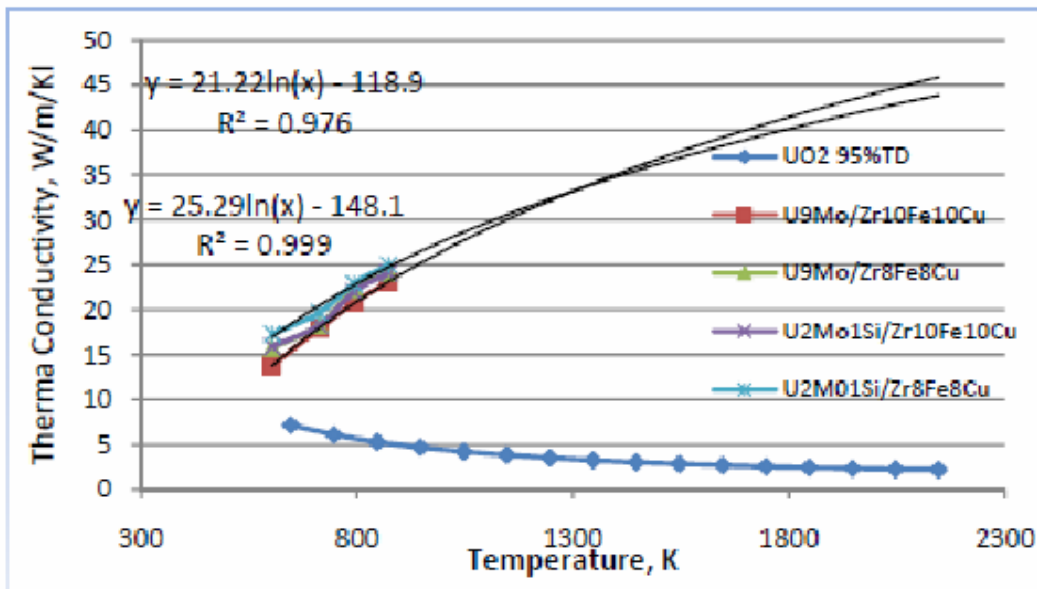


Figure-4. Thermal Conductivity of met-met U-Mo/Zr-Fe-Cu [9] and UO₂ pellet [10]

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 because the pore volume decreases. In view of neutron reactivity, it is needed a more 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 UO₂ 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 Figure 5. Met-met fuel can be loaded ~1.25 higher than UO₂ pellet. The center line temperature of met-met fuel is much lower than UO₂ pellet, it is 380 – 500 oC for metmet and 1500 - 2000 K for UO₂ fuel.

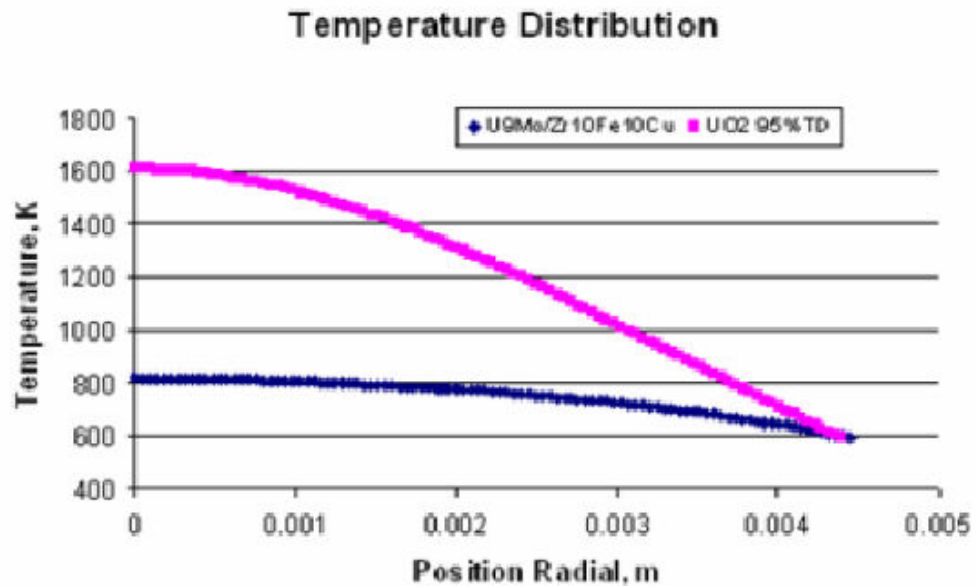


Figure 5. Comparison of radial temperature distributions of UO₂ pellets (square) and U-9Mo/Zr-10Fe-10Cu fuel (diamond), LHGR 47 kW/m, burnup 40 MWd/kg.U

Figure.2 shows plot of thermal conductivity data from 1 from room temperature to 1500 oC for UO₂ pellet (noted UO₂ square – red) and for UO₂ pellet containing tungsten network (noted UO₂W diamond-blue). The upper curve in Fig.5 shows the radial temperature distributions of UO₂ pellet for and the lower curve for new pin containing met-met fuel of U-9Mo/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 is only about 200 K, while pelletized UO₂ 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 seems the potential use of the new pellet that may change the performance of fuel. Result of applying tungsten network 6-9w% in side UO₂ pellet [11] attains ~ 300 K of lowering maximum temperature of pellet. Result obtained by Tulenko et al. which is 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 K [12]. 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 [13].

Many techniques have been developed to avoid the problem.

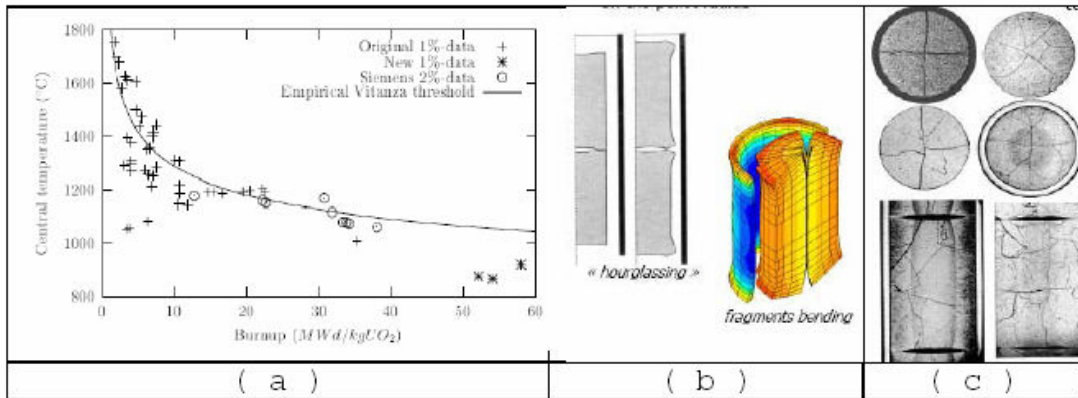


Figure-6. Distribution of Fission gas produced, retained in side and on grain boundary and origin of released gas from the grain [24-15]

Met-met fuel (60%v fuel, 14%v matrix and ~20%v pore has 20-25% higher uranium content than UO₂. 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 Vitansa high burnup appear at temperature above 1800 at low burnup, at about 60 % burnup the threshold temperature of gas release is only ~ 1000 oC. All burnup regime the temperature threshold for gas release (out of pellet) 800 oC as presented in Fig.6(a). The met-met fuel is potentially 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 as presented in Fig.6(b-c) may be avoided by the novel . Gas released from different location tends to diffuse to the lower concentration region that is pellet cladding gap. The addition of fission gas tends to decrease the gap conductance, so the temperature of pellet tends to increase as the gap is the most thermal barrier of heat transfer from pellet to coolant water. Generated fission gas increases sharply in the surface. It is related to the self shielding effect of moderated neutron and Pu reaction by epithermal neutron. The gas diffused in the matrix, precipitated as gas bubble inside the grains. On the rim or outer region where $r > 80\%$ of pellet radius, the precipitated fission gas as bubbles is retained inside the grain lower temperature. The bubbles decrease the rim thermal conductivity. Meanwhile, the novel met-met fuel and cladding is metallurgically bounded, there is no gap between fuel and cladding, and both allow the fuel temperature to be below 1000 oC during normal operation.

4. CONCLUSION

Thermal performance of met-met fuel in novel Zralloy 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 pellet containing tungsten network Met-met fuel permit reduction up to 800 K that is about triple of result by applying tungsten network inside UO₂ pellet, or applying liquid metal bounding between pellets and cladding internal. 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 UO₂ pellet. It is good properties for safety.

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