JAERI-Research
97-085

FISSION GAS RELEASE FROM ROCK-LIKE FUELS, PuO_2 - $ZrO_2(Y)$ { OR ThO_2 }- Al_2O_3 - MgO AT BURN-UP OF 20MWd/kg

November 1997

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編集兼発行　日本原子力研究所
印　　刷　いばらき印刷㈱

Fission Gas Release from Rock-Like Fuels, PuO_2 - ZrO_2 (Y) { or ThO_2}- Al_2O_3-MgO at Burn-up of 20MWd/kg

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(Received October 23, 1997)

Two types of fuels, that is, a 20w/o PuO_2 incorporated into ThO_2-Al_2O_3-$MgAl_2O_4$ and a 23w/o PuO_2 incorporated into ZrO_2(Y) -Al_2O_3-$MgAl_2O_4$ were fabricated into a disk form (outer diameter by 3mm x thickness by 1mm) by JAERI's self-established development technique. The two were provided for steady-state irradiation test at the Japan Research Reactor 3M (JRR-3M) up to the average burn-up of 20 MWd/kg (27 MWd/kg in peak) to understanding fuel behaviour. Post-irradiation examination (PIE) was carried out and the followings were revealed.

(1) Despite of low irradiation temperature<1000℃, there occurred significant fission gas release (FGR) which could not be explained by diffusion mechanism. A possible explanation obtained from fuel microstructural study is direct escape of FP gas from fuel matrix to gas gap via open pores.

(2) A nuclide Cs migrated from fuel matrix to plenum region. Its amount was roughly 20% of totally produced. This was partially due to slight temperature gradient across disk fuel and partly due to low Cs retentiveness in fabricated fuel matrix. A magnitude of Cs-137 migration was smaller in ThO_2-Al_2O_3-$MgAl_2O_4$ disk fuel than that in ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ disk fuel.

(3) Gas bubble swelling rate estimated by total porosity increase was 14% for ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ disk fuel and 11% for ThO_2-Al_2O_3-$MgAl_2O_4$ disk fuel each per 10MWd/kg.

(4) Slight bonding between disk fuels and spacers (Nb-1w/oZr) and that between disk fuels and sheath (Nb-1w/oZr) occurred. The mechanism was attributed to mutual diffusion mainly between Al compound from fuel and Nb one from spacer or sheath.

Keywords: Rock-like Fuel, Fission Gas Release (FGR), ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$, ThO_2-Al_2O_3-$MgAl_2O_4$, JRR-3M, Post-irradiation Examination (PIE), Diffusion Theory, Cs Migration, Gas Bubble Swelling, Bonding, Dimensional Stability.

燃焼度20MWd/kgにおけるPuO_2-ZrO_2(Y)〔またはThO_2〕-Al_2O_3-MgO 岩石型燃料からのＦＰガス放出

日本原子力研究所東海研究所安定化プルトニウム燃料・燃焼法研究特別チーム
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(1997年10月23日受理)

日本原子力研究所は独自に開発した燃料製造技術を用い２種類の燃料を製造した。すなわち一つは20w/oPuO_2にThO_2-Al_2O_3-$MgAl_2O_4$を混ぜたものであり、もう一つは23w/oPuO_2にZrO_2(Y)-Al_2O_3-$MgAl_2O_4$を混ぜたものであり、燃料の形態として外径３㎜肉厚１㎜のディスク〔円板〕状を採用した。この２種類の燃料につき、通常運転下での燃料ふるまいを研究する目的で試験研究炉(JRR-3M)を用い平均燃焼度20MWd/kg(最高27MWd/kg)まで照射を実施した。照射後試験(PIE)にてFPガス放出ふるまい等につき以下の事柄を見いだした。

(1)低い照射温度（<1000℃)にも拘わらず、著しい割合のFPガス放出(FGR)が起こっており、拡散理論では説明の付かないものであった。燃料の微細組織を研究した結果、FPガスが燃料マトリックスから開気孔を通じて直接ギャップ空間に放出されたと考えられた。

(2)セシウム(Cs)が燃料マトリックスからプレナム領域まで移行していた。その量は、生成量の約20%程度であると考えられる。この原因の一つは、使用した円板型燃料の半径方向温度分布がわずかであるが一定でなかったためであり、もう一つは製造段階からこの燃料はセシウム保持能力が弱かったためであろうと考えられる。Cs-137の移行率は、ThO_2-Al_2O_3-$MgAl_2O_4$燃料の方がZrO_2(Y)-Al_2O_3-$MgAl_2O_4$燃料より小さかった。

(3)気孔率の増加量から推定したガスバブルスエリング率は、10MWd/kgにつきZrO_2(Y)-Al_2O_3-$MgAl_2O_4$燃料で14%またThO_2-Al_2O_3-$MgAl_2O_4$燃料で11%であった。

(4)円板燃料とその間に挟み込まれたスペーサ(Nb-1w/oZr)および円板燃料と被覆材(Nb-1w/oZr)との間に僅かな界面癒着が起こった。これは、燃料側からのアルミニウム(Al)化合物とスペーサおよび被覆材側からのニオブ(Nb)化合物の相互拡散に基づくものである。

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1. Introduction

To use a stockpile of weapons-grade plutonium (Pu), several options have been considered to date. Hence, use of mixed oxide fuel (MOX) for thermal reactors (LWR, UO_2-5w/oPuO_2) and further for fast reactors (FR, UO_2-20w/oPuO_2). Regarding fuels, fabrication and irradiation experiences were accumulated much, especially by using test/research reactors. Results of many studies relating to in-pile fuel behaviours were published elsewhere[1-2].

The Japan Atomic Energy Research Institute (JAERI) initiated an original concept of fuel as "once-through type fuels from nuclear warheads in form of the rock-like oxide (ROX) fuel for LWR [3] ". The term "once-through" means once-through usage of plutonium fuel coming out from various sources. In ROX fuel, a utilization of Pu obtained either from reprocessed spent LWR fuel or from nuclear warheads is considered. It has a potential advantage to burn Pu completely in LWR without considering further reprocessing. It become an effective tool to consume nuclear warheads stockpiled now of order of 100 tons both in USA and Russia[4].

A new fuel concept developed in JAERI is principally to incorporate PuO_2 into chemically stable inert matrices and to become stable during and after use in the existing LWR. It can be disposed directly after use in LWR [3].

As a candidate of ROX fuel inert matrices, a two phase mixture of ZrO_2(Y) (fluorite structure) and $MgAl_2O_4$ (spinel structure) was chosen due to their physic-chemical properties of the stable mineral and ceramics. A fissile material PuO_2 can be easily incorporated into ZrO_2(Y) due to resembled fluorite structure. Therefore, a fuel component will be PuO_2 - ZrO_2(Y) - $MgAl_2O_4$. As an another candidate of ROX fuel is chosen thus incorporation of PuO_2 into ThO_2 (fluorite structure). In the latter, a fuel will be PuO_2 - ThO_2 - $MgAl_2O_4$. In the two types of ROX, a small amounts of Al_2O_3 was added to enhance thermal conductivity of the fuels.

After finishing the fabrication of two ROX fuels, a capsule irradiation in the Japan Research Reactor 3M (JRR-3M) was carried out in order to increase an understanding of in-core behaviour. Subsequently, post-irradiation examination (PIE) was done.

The principal objective of the present paper is to study a fission gas release (FGR) behaviour of the two ROX fuels. Fuel dimensional stability relating to bonding was also mentioned. Additionally, a comparison between ROX and conventional LWR fuel was made for recommendations for further studies to be performed.

2. Fabrication of Fuels

An original Pu metal was purchased from UKAEA, England in 1970 containing 94.3 w/o Pu-239 + 0.4w/o Pu-241 as shown in **Table 1**. Prior to getting plutonium dioxide (PuO_2) powder, impurities (0.78w/o) and Am-241 were removed by ion exchanger [3]. Refined PuO_2 powder was blended mechanically together with stabilized zirkelite (ZrO_2(Y)) powder or with thorianite (ThO_2) powder. Other elements such as spinel ($MgAl_2O_4$) and corundum (Al_2O_3) were also added to. They are known widely as component of geologically stable minerals, like a rock [4].

As described previously one of main purpose of addition of the latter elements into fuel matrices was intended to increase thermal conductivity. Hence, ZrO_2(Y) component has a poor thermal conductivity to the magnitude of 0.0047 cal/s/cm^2℃ at 100℃, while $MgAl_2O_4$ and Al_2O_3 components have an excellent thermal conductivity respectively to the magnitude of 0.036 cal/s/cm^2℃ and 0.072 cal/s/cm^2℃. The additives had one order of larger magnitude of thermal conductivity than the former. The properties changed less at elevated temperatures. Regarding fissile components, thermal conductivity of ThO_2 (0.025 cal/s/cm^2℃ at 100℃) is similar to that of UO_2 or PuO_2 [5].

Hereafter, a disk fuel made of PuO_2 - ZrO_2(Y) - Al_2O_3- $MgAl_2O_4$ is abbreviated either as ZrO_2(Y)- Al_2O_3 - $MgAl_2O_4$ fuel or simply as PuO_2 - ZrO_2 fuel. While, a disk fuel made of PuO_2 - ThO_2 - Al_2O_3 -$MgAl_2O_4$ is abbreviated either as ThO_2-Al_2O_3-$MgAl_2O_4$ fuel or simply as PuO_2 - ThO_2 fuel. In the following text, the two different types of fuels will be denoted frequently either as the two specimens or as the two fuels.

During a fabrication the two fuels, powders were pressed first into the form of pellets to have preliminary sintering at temperature of 1400℃ for 14 hours (hereinafter abbreviated as 1400℃ /14h) under atmospheric conditions. Subsequently, pellets were fractured mechanically into small pieces in order to make accurate weighing of Pu amounted to about 40 mg to each specimen. The fragments was pressed again into the

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form of disk for resintering at 1400℃/8h under reduced (8% H_2 + 92% Ar) conditions. The disk after resintering had common dimensions by 3mm in diameter and 1mm in thickness. A detail composition of each disk pellet is shown in the second raw of **Table 1.**

In **Photo. 1**, as-fabricated microstructures taken by scanning electron microscopy and distribution of metal elements involved in the compounds of the two disk fuels are shown. They were taken from center position of the disk fuels. **Photo. 1(a)** is for PuO_2 - ZrO_2 fuel and **Photo. 1(b)** is for PuO_2 - ThO_2 fuel, respectively. In the former, as a whole there observed a good correlation between Zr and Pu distributions along trace line, however, partly (especially at left-hand side in the photograph) there observed poor correlation between them. This poor correlation may be attributed to formation of hibonite described below. In the latter, a correlation between Th and Pu was comparatively good along trace line. With respect to the present ROX fuel, use of reduced condition at resintering stage seems to be doubtful because as seen in the photo.1 a part of fuel matrix was yielded unnecessary compound such as hibonite (metal, Pu, O) at Zr and Th depleted region in the fuel matrix. This may be affected with FGR behaviour during irradiation.

Usually in LWR, UO_2 pellet was sintered at 1700℃/4-7h to control the amounts of open and closed pores <5%[11]. Open pores in a fuel pellet are connecting to fuel surface while closed pores are confined within a fuel having no direct pass to fuel surface. Pores of order of 5% consisted of almost closed pores. During sintering, temperature combined with time is influential to formation of open and closed pores, hence initial fuel density. For present case, however, an increase of sintering temperature under atmospheric conditions was strongly limited below 1400℃ by reason from a capacity of furnace. As a result, a disk fuel having excess open pores to the magnitude of 18% was fabricated. Unfortunately those excess open pores were taken the role to enhance FGR of the two fuels during irradiation. This will be discussed in detail in subsequent section 4 and Appendix 1.

Melting point (hereinafter abbreviated as m.p.) of the disk fuel was not measured in the present study. For ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ fuel or simply PuO_2 - ZrO_2 disk fuel, it. was predicted by a reference composed of 10%UO_2 - 18%ZrO_2 - 27%MgO - 45%Al_2O_3 (all in mol%) and obtained as about 1860℃. For ThO_2-Al_2O_3-$MgAl_2O_4$ fuel or simply PuO_2 - ThO_2 disk fuel, it was predicted by a reference composed of 20%ThO_2 - 40%$MgAl_2O_4$ - 40%Al_2O_3 (all in mol%) and obtained as about 1960℃ [6]. The m.p. of the two fuels seems

to be ranged around that levels(1860℃-1960℃). The most recommendable value obtained from the Author's recent work(to be published by separate report) is 1910℃. It is worthy of mentioning that in spite of high m.p. of original ZrO_2 (2770℃) and additives Al_2O_3 (2050℃) as well as $MgAl_2O_4$(2140℃), m.p. of the two disk fuels were reduced due to formation of new eutectic phases after resintering.

The m.p. of the two disk fuels as high as 1910℃ seems to be one of shortcomings if one compares it with that of UO_2 or MOX fuel (m.p., about 2800℃). As an option, a use of PuO_2 - ZrO_2 only two phases compounds fuel instead of PuO_2 - ZrO_2 - $MgAl_2O_4$ - Al_2O_3 four compounds fuel may be possible if one wants strongly to have a high m.p. In that case, however, one should be necessary to compensate a thermal conductivity of it at least as the same level as conventional UO_2 fuel, because as mentioned previously ZrO_2 has a poor thermal conductivity. In the present study, a combination of oxide fuels having rather high m.p. (PuO_2-ZrO_2) with those having excellent thermal conductivity ($MgAl_2O_4$-Al_2O_3) was selected sacrificing the reduction of melting point. It is recommended to make more effort to find out a fuel having not only high m.p. but also excellent thermal conductivity than the two fuels presented here.

The first loaded two fuels were, however, proven to be advanced in proliferation resistance and environmental safety due to complete burn of Pu resulting in immunity of reprocessing[7].

3. Capsule Irradiation

3.1 Temperature of disk fuel

As shown in Fig. 1, a total of five disks in a fuel was fabricated. Each disk fuel (3mm by outer diameter (O.D.) and 1mm by thickness(t)) was sandwiched by disk spacer (3.5mm O.D. and 0.5mm t) and loaded into sheath (instead of cladding we prefer to use the term), both were made of 99w/o Nb-1w/oZr alloy. Fuel column in the sheath contained 5 disk fuels (8mm in length) and 6 spacers. Fuel sheath contained a fuel column, aluminum wool and a suppression rod having a center hole diameter by 1mm at the top of fuel column where was known as plenum region. Practically the suppression rod did not have function to prevent the disk fuels from axial and radial movement during the course of irradiation. A center hole drilled into the suppression rod can permit migration

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of FP from disk fuels to plenum region, if it occurs.

The sheath was filled with pure helium (He) at ambient pressure. Calculated plenum volume from the drawing shown in Fig. 1 was about 0.4 ml. The sheath was assembled into a fuel pin made of Nb alloy and was filled with pure He under ambient pressure. The fuel pin had O.D. by 10mm and length by 89mm. Prepared fuel pins in this study were total three for PuO_2-ZrO_2 fuels and three for PuO_2-ThO_2 fuels, respectively. The fuel pin was assembled into fuel container made of Nb alloy having O.D. by 10mm and length by 88.5mm prior to loading into an irradiation capsule.

In the irradiation capsule named as BRF-24H as shown in **Table 2**, two fuel pins denoted as No.1 (#1), which contained PuO_2-ZrO_2 disk fuels and No.4 (#4), which contained PuO_2-ThO_2 disk fuels were positioned in bottom of capsule. A fuel temperature at the place was designed to be 800℃. Two fuel pins denoted as #2 and #5 were placed at the middle (hereinafter abbreviated as mid.) location, hence 15 cm above from bottom. The remainder two fuel pins #3 and #6 were placed at the top location, hence 30 cm above from the bottom. Designed fuel temperatures were 1000℃ for the former and 600℃ for the latter.

A temperature of disk fuels was closely monitored by two alumel-chromel type thermocouples (T/C's) having O.D. by 1.6 mm. T/C's were attached directly to the bottom of fuel container of which span was about 7mm from the disk fuel. They were additionally attached to the container wall spanned by 5mm from the disk fuel. In experiment, a fuel temperature at mid position (1,000℃) was carefully controlled and monitored by means of mixed(nitrogen and helium) gas flow apparatus installed into the capsule.

As T/C did not touch directly with disk fuel, a temperature drop from disk to T/C was necessary by calculating heat conductance between the two. Obtained results were 107, 116 and 93℃ depending upon disks located at bottom, mid and top, respectively. Temperatures at the disk fuels were finally decided as a sum of temperature drop obtained from calculation and monitored temperature at T/C location.

Averaged disk fuel temperatures through experiment are shown in **Table 3.** A small error band within plus minus 5% at mid location is a result of careful control of temperature done at this point and a large error band to the magnitude of around 18%

at top location is due to a unavoidable control rod movement in the core. A maximum temperature at mid location was around 990℃.

The authors tried to estimate a linear heat generation rate(LHGR) corresponding to aforementioned temperature of 990℃ but found to be very hard due to capsule irradiation involving a disk type fuel. Then LHGR of the present study was estimated by using data obtained from past PWR in-core experiment [8] carried out at Halden Boiling Water Reactor. A 17x17 PWR fuel rod consisted of UO_2 fuel with diametral gap by 0.17mm and prepressurized to 1.5 MPa had fuel centerline temperature as a function of LHGR at beginning of life (BOL). An extrapolated LHGR from the data base, hence changing diametral gap from 0.17mm to 1.0mm used in the present study at fuel centerline temperature of 990℃, was about 21 kW/m in average (peaking factor, 1.0). It is worthy of mentioning that 21 kW/m is a typical LHGR in conventional PWR. It may be said from viewpoint of fuel temperature that present study has a similarity to that of conventional standard PWR fuel.

In the capsule, three containers for PuO_2-ZrO_2 and those for PuO_2-ThO_2 were prepared and they were designed to have well vertical alignment. At any horizontal section, a span between two centerlines of each container was about 12mm. The narrowest gap between two containers was about 2mm. All of the gap space was filled with molybdenum metal as a thermal insulator. In the course of irradiation there was no sign of fuel failure.

3.2 Determination of Burn-up

The capsule irradiation was started since January 1995 and terminated on December 1995, resulting in an effective full power hours (EFPH) of 2.33 $x10^3$ which was equivalent to 97 effective full power days (EFPD). A thermal neutron flux monitored by Co-Al alloy at three axial different locations is shown in **Table 3.** The axial power shape was cosine curve having a peaking factor of 1.18.

Burn-up of disk fuels determined by Nd-148 method is simple and described elsewhere[9]. Obtained burn-up for PuO_2-ZrO_2 fuel is 27.90 fission per initial metallic atoms (FIMA(%)) at mid. location having a peaking factor of 1.18 and 20.75 FIMA(%) at top having a peaking factor of 1.00. While, that for PuO_2-ThO_2 fuel is 27.95 FIMA(%) at peaking factor of 1.18.. From fuel morphology point of view, the present two fuels may

be categorized into a dispersion type fuels, hence PuO_2 fissile material is dispersed in the matrix composed ZrO_2, Al_2O_3 and $MgAl_2O_4$. A unit conversion procedure from FIMA to MWd/kg per fissile Pu was made taking into consideration of an initial plutonium composition (mol%). Then resultant burn-up according to this concept was 20 MWd/kg in average and 27 MWd/kg in peak irrespective of the two types of fuels. To check the validity of the present burn-up conversion method, a steady state/accident computer code " FPRETAIN" developed by JAERI[21] was used making input as close as most realistic. Calculated value was closed to that mentioned above. Regarding burn-up determination procedure on dispersed type fuels it should be necessary more discussions for finding out a suitable conversion factor from FIMA to MWd/kg which has been used widely in conventional LWRs.

One difficulty met during burn-up analysis should be introduced here. At first one could not dissolve sampled three specimens due to nature of rock-like oxide fuels, that is, the specimens had a very strong resistance against dissolution by nitric acid (HNO_3) used widely to melt spent LWR fuels. For dissolution of ROX fuel, a solution of aqua regina (HNO_3:HCl = 1:3) combining with small volume of HF was prepared. The solutions with specimens were heated up to 150℃ and hold continuously for 40h. Three specimens were, however, not dissolved. Then one stirred a solution a specimen was fragmented into small pieces due to melt of Al_2O_3 component in a specimen. Another 20h holding with a small volume of fresh aqua regina was requested until three specimens were dissolved completely.

4. PIE, Results and Discussion

After irradiation to 97 EFPD, the capsule was unloaded from JRR-3M and cooled further 90 days in the core pit prior to transportation to the Department of Hot Laboratory, JAERI. At the Hot Laboratory, six containers were disassembled from the capsule to take out six sheaths from six containers. Then six fuel pins were dismantled from all sheaths and provided for visual inspection. No damage was observed in all fuel pins.

4.1 Dimensional Stability

In Fig. 2, representative X-ray photographs taken from the two type of disk fuels loaded

be categorized into a dispersion type fuels, hence PuO_2 fissile material is dispersed in the matrix composed ZrO_2, Al_2O_3 and $MgAl_2O_4$. A unit conversion procedure from FIMA to MWd/kg per fissile Pu was made taking into consideration of an initial plutonium composition (mol%). Then resultant burn-up according to this concept was 20 MWd/kg in average and 27 MWd/kg in peak irrespective of the two types of fuels. To check the validity of the present burn-up conversion method, a steady state/accident computer code " FPRETAIN" developed by JAERI[21] was used making input as close as most realistic. Calculated value was closed to that mentioned above. Regarding burn-up determination procedure on dispersed type fuels it should be necessary more discussions for finding out a suitable conversion factor from FIMA to MWd/kg which has been used widely in conventional LWRs.

One difficulty met during burn-up analysis should be introduced here. At first one could not dissolve sampled three specimens due to nature of rock-like oxide fuels, that is, the specimens had a very strong resistance against dissolution by nitric acid (HNO_3) used widely to melt spent LWR fuels. For dissolution of ROX fuel, a solution of aqua regina (HNO_3:HCl = 1:3) combining with small volume of HF was prepared. The solutions with specimens were heated up to 150℃ and hold continuously for 40h. Three specimens were, however, not dissolved. Then one stirred a solution a specimen was fragmented into small pieces due to melt of Al_2O_3 component in a specimen. Another 20h holding with a small volume of fresh aqua regina was requested until three specimens were dissolved completely.

4. PIE, Results and Discussion

After irradiation to 97 EFPD, the capsule was unloaded from JRR-3M and cooled further 90 days in the core pit prior to transportation to the Department of Hot Laboratory, JAERI. At the Hot Laboratory, six containers were disassembled from the capsule to take out six sheaths from six containers. Then six fuel pins were dismantled from all sheaths and provided for visual inspection. No damage was observed in all fuel pins.

4.1 Dimensional Stability

In Fig. 2, representative X-ray photographs taken from the two type of disk fuels loaded

at mid location (990℃) are shown. Relocation of the two disk fuels occurred. The relocation may cause a slight temperature gradient across disk fuel that was not expected before study. It is noticeable that one of main purpose of disk fuel usage is to give uniform temperature distributions across fuels, however, it did not occur practically.

As to the case of axial gamma scanning, one was for PuO_2-ZrO_2 fuel pin done without container and the other for PuO_2-ThO_2 fuel pin done with container. The two fuel pins were taken from mid location (990℃). In measurements, five different nuclides were selected and the results are shown in **Fig. 3.** We were interested in comparing magnitude of gamma activities between the two disk fuels so that Zr-95 and Rh-106 were selected here for this purpose. A magnitude of Zr-95 was greater than that of Rh-106 in PuO_2 - ThO_2 fuel pin because the measurements were made with container made of Nb - 1w/oZr alloy. For PuO_2-ZrO_2 disk fuel a magnitude of gamma activity from Zr-95 was found to be smaller than that of Rh-106 because the measurements were made without container.

As shown in **Fig. 3(a)**, nuclides Rh-106 and Cs-137 from PuO_2-ZrO_2 fuels clearly indicated five gamma peaks at arbitrary distance between 10 and 30mm, where gamma activities were traced from pin bottom (figure left) to top direction. The gamma peaks were exactly corresponded to the disk fuels. Almost the same magnitudes in gamma activity either at each disk fuel locations or at each spacer locations implied that disk fuels in this case burned uniformly, except one disk fuel at the most top location which showed relative low gamma activity than others. The equal span observed among the disk fuels implied that relocation or deformation occurred little.

In **Fig. 3(b)**, nuclides Rh-106 and Cs-137 for PuO_2-ThO_2 fuel also indicated five gamma peaks at arbitrary distance between 10 and 30mm, where gamma activities were traced from cladding bottom (figure left) to top direction. A magnitude of gamma activity was roughly equal at each disk fuel location but not at spacer location. It implies that disk fuels in this case burned almost uniformly but relocated to some degree of magnitude. A span between the disk fuels was not the same implying an occurrence of relocation, too. Th relocation occurred in large magnitude at the bottom location.

Disk fuels could relocate because of a large fabricated gap by 1mm (c.f. 0.17mm in PWR) and having no axial constraint such as a plenum spring. According to gamma scanning

data a magnitude of relocation in PuO_2-ThO_2 disk fuel was greater than that in PuO_2-ZrO_2. As a comparison, fuel microstructures obtained from fuel #2 (PuO_2-ZrO_2, mid., 990℃) and #5 (PuO_2-ThO_2, mid.,990℃) were prepared. By using pictures taken from local cross sectional area we examined numbers of cracks and degree of local deformations occurred. The result showed that relocations were relatively large in PuO_2 - ZrO_2 than in PuO_2-ThO_2 fuel . This was contrary to that obtained from gamma scanning. We consider that priority for judgement should be addressed to the gamma scanning data because observations obtained from the latter is very local and not representing the whole tendency. It should be remarked here that PuO_2-ThO_2 fuel relocated greatly than that of PuO_2-ZrO_2 one.

Due to the occurrence of bonding (see, section 4.4) it is very hard to discuss about a dimensional stability between the two disk fuels. During visual inspections we observed many incipient and plate-through cracks in both disk fuels. This implied that radial temperature profile across disk fuel should not uniform. As a cause of incipient cracking one may point out a possibility of residual stress occurred during fabrication stage. According to previous study by UO_2 fuel it is known that plate-through crack could occur by temperature gradient>100℃ between adjacent points[10] in the fuel matrix. However, plate-through cracks occurred in the present fuels despite of a slight temperature gradient<50℃, as shown in Table 2. This phenomenon might be one of characteristics of the disk fuels used.

To increase knowledge about dimensional stability, more studies taking the following factors into consideration are recommended to perform in future. Hence, (a) Fuel form: use pellet instead of disk, (b) Diametral gap: use normal(0.17mm) instead of large (1mm), (c) Cladding: use zircaloy or SUS instead of Nb-1w/oZr, and (d) Plenum structure : use plenum spring instead of non-constraint suppression rod.

At present it is concluded that due to limited data base further discussions about a dimensional stability of the two disk fuels should be terminated.

4.2 Release of Fission Product (FP)

In conventional UO_2 fuel used in LWR, there occurred a migration and release of FP from fuel matrix to gas gap. For example, Cs-137 usually tends to migrate from center to fuel edge according to temperature gradient resulting in forming reversed parabolic

curve at the EOL[11]. Frequently, deposited Cs at the fuel edge acts as aggressive chemical species for stress corrosion cracking (SCC) of zircaloy cladding[12]. Hence, a study on release of chemical aggressive species like Cs is important from fuel reliability point of view.

In the present study, whether or not Cs could migrate from disk fuel to pin plenum was most concerned. From gamma scanning data, we found that there occurred Cs migration across the disk fuels. Only one exception was PuO_2-ThO_2 disk fuel came from bottom location (766℃) which showed no trace of Cs migration.

A typical Cs migration is shown in **Fig. 3(a)**. Released Cs-137 and Cs-134 deposited much at aluminum wool part, namely more cold part toward pin plenum. Released rate of Cs-137 was amounted to be about 20% of total produced. Comparison between Fig. 3(a) and (b), it can be said that release of Cs-137 from PuO_2-ThO_2 fuel was smaller than that from PuO_2-ZrO_2 one. The former has a relative strong resistance against FP migration than the latter though we cannot make clear explanation about its mechanism. Within this experimental scope, no other nuclide except Cs was released from the disk fuels to plenum.

In conventional PWR, Cs migration across fuel pellet will occur along parabolic temperature gradient down to pellet edge, where temperature is about 400℃. So, at the fuel center region Cs tended to be depleted much but accumulated significantly at the fuel edge region. In the present case, an occurrence of Cs migration implied a slight temperature gradient across disk fuel.

4.3 Release of Gaseous FP

A gaseous FP release from the two disk fuels was studied by PIE data obtained from gas puncturing. Target gases interested in studying here were xenon (Xe) and krypton (Kr) isotopes because of their poor thermal conductivity and relative high fission yield of about 20% to the whole gas production. Released gases were collected from fuel pin plenum. For simplification, five disk fuels were counted as a pellet having outer diameter by 3mm and length by 5mm. In the following it was denoted as " equivalent pellet ".

During gas puncturing, a top plenum part of each fuel pin was drilled mechanically in

order to trap gaseous FP into evacuation bottle of which volume and pressure were known. The collected gases were expanded into gas chromatograph to determine a volumetric percentage of individual collected gases. Existing ratio of Xe and Kr radioisotopes were also studied.

Results are shown in **Table 3.** In addition to Xe and Kr, nitrogen and oxygen from air, Ar from welding, and He from initial filler gas were detected. A total volume of collected gas was about 0.4ml coincided well with initial calculation. After knowing pressure and volume of collected gases, one obtained the plenum volume of fuel pin at EOL. Comparison of each plenum volume of fuel pin led that fuel pin No.1(#1, PuO_2 - ZrO_2, bottom, 766℃) had a plenum volume of 0.24ml, this was almost half volume of others. Fuel fabrication report, however, mentioned that an initial plenum volume of #1 was as same level as others. This leads that puncturing data is suspicious. Hence, we omit pin #1 data from the subsequent discussions.

By using a simplified diffusion model[13], fission gas (Xe+Kr) release from an equivalent pellet i.e., from five disk fuels was tried to estimate by the following equation.

FGR (%) = { Released amounts of Xe+Kr from an equivalent pellet} / {Produced amounts of Xe+Kr in an equivalent pellet} x100

= {A total volume of Xe+Kr gases obtained from gas puncturing / y x BU x Wf} x 100 ----------/1/

where, y: Fission yield of Xe+Kr (cc/MWd)

BU: Average burn-up(MWd/kgPu) of equivalent fuel determined by Nd-148 method, see Table 3,

Wf: Weight of fissile materials at BOL(kg Pu)

As shown in Table 3, calculated values regarding pins #2 - #6 are of order of 100%. The reader should be recognized that it is impossible to consider such significant FGR to be caused by thermal diffusion at temperature<1,000℃, even though at accumulated burn-up of 20MWd/kgPu. Further, if recoil and knock-out mechanism dominated the equivalent pellet, FGR should be < 1% as usually seen in conventional PWR fuels[13]. Consequently, we concluded that FGR from present disk fuels was neither dominated by diffusion nor recoil/knock-out mechanism. Therefore calculation made by above equation /1/ was invalid.

To find out a cause of such significant FGR in the disk fuels, a fuel microstructural

study was made. As-polished fuel microstructures taken from irradiated pin #2 (PuO_2-ZrO_2, mid, 990℃) and pin #5 (PuO_2-ThO_2, mid, 990℃) are prepared and shown in Fig. 4, where pictures from as-fabricated fuels were also included for comparison. It is worthy of mentioning that significant open pores in the fuel matrix are existed not only from BOL but also EOL. A ratio of area occupied by open pores having a diameter>0.2 micron against the whole area of photograph was defined as porosity shown by percentage. Measured porosity at EOL was 46% for pin #2 and 39% for pin #5. In Appendix 1, details about porosity distribution change in the course of irradiation are described separately.

It is widely known that in conventional PWR a UO_2 fuel pellet mostly had closed pores to the magnitude of <5%[14]. This was result of high sintering temperature (1700-1750℃) and sintering time(4-8 hours) under reduced condition. A main purpose of porosity control in PWR is to prevent the fuel from significant densification at BOL.

A significant FGR from disk fuels may occur partly due to marked open pores in a fuel matrix fabricated during sintering stage and partly due to gaseous FP produced by fission. As mentioned above all fuels had porosity ranged from 39% to 46% so that fission gases could possibly migrate from fuel matrix to gas gap via pass to fuel surface formed by open pores. From a viewpoint of fuel reliability, one should avoid a significant FGR as observed in the present study because of contamination of gas gap and increase of plenum pressure. These are affecting fuel reliability significantly.

From present study we learned that control of open pores in the fabrication stage was of importance in comparison with UO_2 pellet in LWR. Reduction of open pores may be possible by increasing sintering temperature and time to the same level as used in conventional PWR fuel fabrication. The oxidized condition instead of reduced one for fabrication is preferable to avoid formation of the hibonites (Pu_2O_3 . $11Al_2O_4$). The compound seems to have a potential to enhance significant FGR though its mechanism is not revealed to date.

4.4 Swelling

Swelling in UO_2 pellet occurs by fission gas bubble formed in the fuel matrix. If UO_2 accompanies no external constraint such as pellet-cladding mechanical interaction (PCMI), swelling rate will be of order of 1-20% volumetric change per 10 MWd/kg

depending on pellet temperature history[19]. We called such swelling as "gas bubble swelling". While, If UO_2 accompanies external constraint, swelling rate will be of order of 1% volumetric change per 10 MWd/kg[19]. We called it as "solid swelling". Swelling could be monitored, for example, by continuous measuring of fuel column change as a function of burn-up. Occasionally, a change of pellet bulk density before and after irradiation was used.

T. Ohmichi reviewed recently a swelling rate of present types of fuel and found that gas bubble swelling occurred with range from 2 to 10%[20] by irradiation up to 10^{22}n/cm^2 . The worst case was attributed by non-crystallization(or amorphization) of aluminum compounds such as Al_2O_3 or $MgAl_2O_4$. While ZrO_2(Y) causes less swelling.

We estimated a gas bubble swelling rate of the two disk fuels by means of total porosity change in the fuel matrix because an increase of total porosity in fuel matrix was directly connected to decrease of fuel bulk density. As shown in Table A1, a total porosity in the disk fuel was determined directly from fuel microstructure. Consequently, a total porosity in PuO_2-ZrO_2 fuel increased from 18.4% to 45.5% and that in PuO_2-ThO_2 fuel increased from 17.4% to 38.7%, respectively. Net increase during irradiation was about 27% for the former and 21% for the latter per 20 MWd/kg in average burn-up. In other words, average swelling rate is 14% for PuO_2-ZrO_2 fuel and 11% for PuO_2-ThO_2 fuel per 10MWd/kg. Observed swelling was gas bubble one because of no constraint of disk fuel from external. Observed gas bubble swelling in the two fuels was larger not only than those reviewed by Ohmichi[19] but also than those by UO_2[20]. The main cause might be attributed to significant increase of porosity in fuel matrix during irradiation.

At present there is no clear reason why total porosity increased 2-3 times at burn-up of 20 MWd/kg. Two possibilities were considered. One is agglomeration and redistribution of original pores. From Table A2, occurrence of pore agglomeration and redistribution are implied. The other is growth and accumulation of fission gas bubbles during irradiation. Unfortunately, in Fig. 4, we cannot distinguish between agglomerated /redistributed pores and fission gas bubbles yielded during irradiation.

Recommendations: In order to clear this, one of best way is to measure change of fuel column length continuously. Then we can also evaluate the fuel densification behaviour.

4.5 Bonding

In LWR either a fuel rod irradiated to extended burn-up region(>50MWd/kgU) or one irradiated with high temperature(>1,800℃) is known to cause bonding, which is generally dominated by mutual diffusion of uranium compound diffused from fuel and zirconium compound diffused from zircaloy cladding. At the interface of pellet and cladding, formation of (U, Zr) O_2 compounds is observed mostly. When it happened, fuel axial and radial displacements are limited to further movement. At steady- state operation, a bonding at the extended burn-up region is tended to be significant in BWR than in PWR due to use of Zr liner at inside of cladding[15,16]. Bonding should be taken into consideration from fuel reliability point of view, however, fuel failure directly related to bonding was not reported to date.

During reactivity initiated accident (RIA) condition, a significant bonding shall occur irrespective of a power history of each fuel rod due to strong contact between cladding inside and hotter pellets. Because of fuel relocation occurred locally during RIA, bonding also occurred locally at inside of cladding. Due to unbalance of heat transfer occurred between bonded and non-bonded cladding region, thickness of cladding was drastically changed from place to place. During quenching phase, cracks were initiated mostly from thinner part of brittle cladding[17]. Hence, fuel bonding takes important role on fuel failure during RIA.

In the present study, bonding occurred either between disk fuels and Nb-1%Zr sheath or between disk fuels and Nb-1%Zr spacers. The main components of the two disk fuels were PuO_2-ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ and PuO_2-ThO_2-Al_2O_3-$MgAl_2O_4$. The reaction occurred with stress free conditions among them. A representative feature taken from bonding part occurred in specimen #6 (PuO_2-ZrO_2, mid.,990℃) is shown in **Fig. 5(a)**.

Around point (P) shown in Fig. 5(a), an analysis by scanning electron microscope(SEM) was made to understanding detail condition at bonded area. Observations were made along transversal trace line shown in the figure.

- Bonding occurred slightly between disk fuel edge and Nb-1%Zr spacer. At edge of fuel all compounds except aluminum was depleted in the range of about 10 microns. Thus, aluminum compound was diffused from fuel and reacted with Nb-1%Zr spacer. Bonded layer consisted of Al and Nb was only a few microns. This kind of light bonding might be due to little external compressive stress between disk fuel and spacer.

- Except bonded region, there is little correlation between Pu and Zr, where Pu was dispersing almost uniformly irrespective of another compounds. A magnitude of correlation between Pu and Zr observed in as-fabricated condition(see, Photo.1(a)) was almost disappeared at this place.
- A niobium(Nb) was diffused out from spacer to fuel matrix and deposited at zirconium rich region. It is notable that amounts of niobium in the disk was decreased significantly from original state of Nb-1%Zr due to this Nb migration.
- Usually, a fission product(FP) will be a main cause of bonding, especially at extended burn-up stage[18]. In the present study, however, we didi not study the role of FP on formation of bonding.
- For further study, one should be taken the followings into consideration: An occurrence and magnitude of bonding should be checked when one changes sheath from Nb - 1%Zr to zircaloy or stainless steel cladding. A magnitude of bonding should be studied when fuel pellet is used instead of disk fuel.

5. Conclusion

Plutonium incorporated rock-like oxide fuels (ROX) were fabricated by JAERI with self-established development technique. In the present study, two types of fuels were used. One had a composition of 23w/oPuO_2-17w/oZrO_2-56w/o-Al_2O_3-4w/oMgO (abbreviated here as ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ or simply as PuO_2-ZrO_2 fuel). The other has a 20w/o PuO_2-29w/oThO_2-48w/o Al_2O_3-3w/oMgO (abbreviated as ThO_2-Al_2O_3-$MgAl_2O_4$ or simply as PuO_2-ThO_2 fuel). They were fabricated in form of disk for a feasibility study. The five disk fuels consisted of an active column length by 5mm and fuel diameter by 3mm.

Steady-state capsule irradiation was carried out at JRR-3M to the average burn-up of 20MWd/kgPu or 97 EFPD as a function of three different fuel temperatures, that is, 766℃ (bottom), 990℃(mid) and 706℃(top) depending upon axial location in the capsule. The following remarks were found mainly through post-irradiation examination (PIE).

(1) Despite of relative low temperatures <1000℃, a fission gas release (FGR) from the tested disk fuels was of order of 100% which was too large to understand by diffusion mechanism. The possible mechanism revealed is FP (Xe and Kr) gas escape directly from fuel matrix to gas gap via free pass consisted of open pores existed markedly.

(2) Retentiveness of Cs-137 in the tested fuel matrix was found to be low under present experimental conditions. Hence, amounts of about 20% of totally produced Cs-137

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was migrated from fuel matrix to non-fueled region due mainly to slight temperature gradient occurred across the disk fuel. The total amounts of Cs-137 released were smaller in ThO_2-Al_2O_3-$MgAl_2O_4$ fuel than in ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ fuel.

(3) Gas bubble swelling rate estimated by total porosity increase during irradiation was 14% for ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ fuel and 11% for ThO_2-Al_2O_3-$MgAl_2O_4$ fuel each per 10MWd/kg.

(4) Due to a large diametral gap (1mm) between disk fuels and sheath made of Nb-1w/oZr materials and also due to no axial constraint such as a plenum spring, the disk fuels were cracked and relocated. According to gamma scanning data a magnitude of relocation in ThO_2-Al_2O_3-$MgAl_2O_4$ fuel was greater than that in ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ fuel.

(5) Bonding occurred slightly in the two disk fuels. The location is between disk fuel and spacer made of Nb - 1w/oZr or between disk fuel and sheath made of Nb-1w/oZr. The main mechanism revealed was a mutual diffusion of Al compound from fuel matrix and Nb compound from spacer or sheath.

Acknowledgment

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Appendix 1 Porosity Change during Irradiation

Changes of pore diameter and porosity by irradiation were studied by PIE. Specimen from ThO_2-Al_2O_3-$MgAl_2O_4$ or simply PuO_2 - ThO_2 disk fuel irradiated at 990℃(mid) and one from ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ or simply PuO_2-ZrO_2 disk fuel irradiated at 990℃ (mid) were prepared and provided for analysis by two-dimensional image analyzer. Measurements were addressed to fuel center locations. The results are shown in Table A1.

Table A1 Results of porosity measurement

Specimen	Average pore diameter(micron)		Total porosity(%)		Swelling(%)
	Pre-irrad.	Post-irrad.	Pre-irrad.	Post-irrad.	per 10MWd/kg
PuO_2-ThO_2	2.3±3.2	4.4±6.4(1)	17.4	38.7	10.7
PuO_2-ZrO_2	2.5±4.5	4.4±5.2	18.4	45.5	13.6

Note: (1) Error band was obtained statistically.

It is found from the table that average pore diameter was increased by two times through irradiation. Total porosity in the two disk fuels was increased approximately from 17% to 39% for PuO_2-ThO_2 and from 18% to 46% for PuO_2-ZrO_2 fuel, respectively. To study the reason of it, a porosity distribution as a function of pore diameter was measured and shown in Table A2.

Table A2 Porosity as a function of pore diameter

Pore diameter(micron)	< 1	1-10	<10	max. observed	Area in measure. ($x10^4$ um^2)
PuO_2-ThO_2					
Pre-irradiation(%)	53(375)	34(239)	13(93)	100	2.3
Post-irradiation(%)	38(183)	51(245)	11(50)	39	5.0

PuO_2-ZrO_2					
Pre-irradiation(%)	61(396)	26(168)	13(85)	220	2.3
Post-irradiation(%)	20(105)	74(397)	6(30)	51	5.0

Note: Figures in parentheses are numbers of pores counted.

The followings are revealed from the table.

(1) There was no marked difference in PuO_2-ThO_2 and PuO_2-ZrO_2 for pore distribution at pre-irradiation stage.

(2) Pores smaller than 1 micron decreased porosity and their numbers significantly after irradiation. The tendency was similar to pores having diameter greater than 10 micron. The tendency was completely reverse in pores having diameter from 1 to 10 microns.

The similar pore distribution observed between two different disk fuels seems to be attributed to similar fabrication process. Agglomeration of pores might be addressed markedly to pores>10 micron. Effect of irradiation on change of porosity seems to be addressed to pores having diameter<1micron as well as pores having diameter between 1 and 10 micron. This is coincided well with those observed in UO_2 fuel[18].

A presence of significant submicron pores(having diameter<1micron) at EOL should be derived from fission gas bubbles. For the case of UO_2 fuel, a densification occurred usually from the beginning of irradiation accompanying with annihilation of as-fabricated submicron pores.

Consequently, the presence of significant amounts of pores in the disk fuel matrix was a main cause of significant FGR and swelling.

Table 1 Characteristics of Fabricated Disk Fuels

Composition of Pu isotopes (at%)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
	<0.01	94.3	5.3	0.4	<0.01

Specimen (1)	
Powder	23w/oPuO_2-17w/oZrO_2(Y)[a]-56w/oAl_2O_3-4w/oMgO
After resintering	PuO_2-ZrO_2(Y)-Al_2O_3(corundum)-$MgAl_2O_4$ (spinel)
Written in the text as	ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ fuel or simply PuO_2-ZrO_2 fuel
Specimen (2)	
Powder	20w/oPuO_2-29w/oThO_2-48w/oAl_2O_3-3w/oMgO
After resintering	PuO_2-ThO_2-Al_2O_3(corundum)-$MgAl_2O_4$ (spinel)
Written in the text as	ThO_2-Al_2O_3-$MgAl_2O_4$ fuel or simply PuO_2-ThO_2 fuel

Specifications					
Disk Fuel		Fuel Sheath		Fuel Pin	
Fuel form	Disk	State	Zr-1w/oNb	State	Pt tube
Outer diameter(O.D.)	3mm	O.D.	6mm	Enriched fuel column	5mm
End form	Flat	Inner Diameter	4mm	Diametral gap	1mm
Length	1mm			Fill gas pressure	0.1MPa
L/D[b]	0.3			Gas composition	100% helium(He)
				Plenum volume	about 0.4ml

Note:(a) ZrO_2(Y), Stabilized ZrO_2 composed of 88.8%ZrO_2-11.0%Y_2O_3-0.2%Gd_2O_3(mole percent)
(b) L/D, Ratio of disk length to diameter

Table 2 Temperatures of Disk Fuels During Capsule Irradiation at JRR-3M

Pin No.	Fuel Type[1]	Weight of 5 disk fuels(g)	Location in Capsule[2]	Target Temp.(℃)	T/C reading (℃)[3]	Temp. drop (℃)[4]	Estimated temp. (℃)[5]
#1	PuO_2-ZrO_2	0.147	B	800	659±31	107	766±31
#2		0.154	M	1000	874±12	116	990±12
#3		0.161	T	600	613±49	93	706±49
#4	PuO_2-ThO_2	0.179	B	800	659±31	107	766±31
#5		0.154	M	1000	874±12	116	990±12
#6		0.160	T	600	613±49	93	706±49

Note:(1)ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ fuel or simply PuO_2-ZrO_2 fuel, ThO_2-Al_2O_3-$MgAl_2O_4$ fuel or simply PuO_2-ThO_2 fuel

(2)B: Bottom location, M: Middle location, and T: Top location

(3)Small error band at mid. location was due to intentional temperature control at thermocouple(T/C) made by mixed flow of helium and nitrogen gas. Large error bands at bottom and top were due to frequent change of neutron flux made by control rod movement

(4)Temperature drop from disk fuel to T/C was estimated by using heat conductance of 15,000 kcal/m^2h℃ between two points.

(5) Sum of T/C reading and temperature drop. Obtained figure is lower than that of target temperature at bottom (5%), middle(1%) but higher at top(18%).

Table 3 Irradiation Conditions in JRR-3M and Results of Gas puncturing in PIE

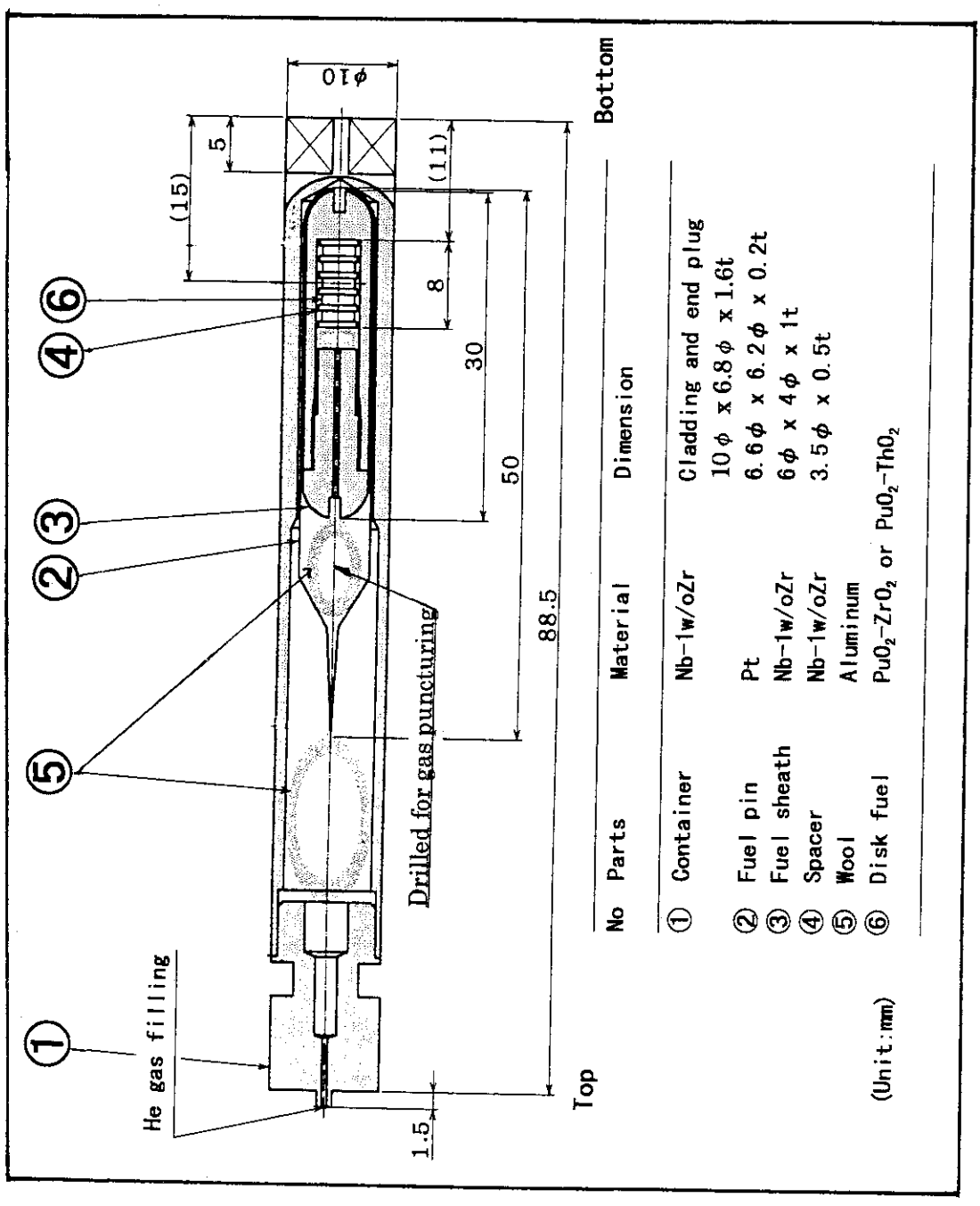
Pin/Location	Estimated temp.(℃)	Thermal flux (n/cm^2s)	Peaking factor	Burn-up[1] (MWd/kg)	Results of Gas Puncturing Vol(ml)[2]	FP gas(vol%) Kr	Xe	Released Kr+Xe(ml)	FGR(%)
(1)ZrO_2(Y)-Al_2O_3-$MgAl_2O_4$ or simply PuO_2-ZrO_2 fuel									
#1 /Bottom	766±31	5.3×10^{13}	0.88	-	0.22	0.1	1.9	4.4×10^{-3}	-[3]
#2 /Mid	990±12	7.1×10^{13}	1.18	26.8	0.37	2.5	42.8	0.17	-[4]
#3 /Top	706±49	5.9×10^{13}	1.00	19.9	0.36	1.9	35.2	0.13	-[4]
(2)ThO_2-Al_2O_3-$MgAl_2O_4$ or simply PuO_2-ThO_2 fuel									
#4 /Bottom	766±31	5.3×10^{13}	0.88	-	0.38	1.8	32.4	0.13	-[4]
#5 /Mid	990±12	7.1×10^{13}	1.18	26.8	0.45	2.0	35.8	0.17	-[4]
#6 /Top	706±49	5.9×10^{13}	1.00	-	0.38	2.6	45.1	0.18	-[4]

Note:(1) Megawatt gays per kilogram of heavy metal by initial plutonium(Pu). Burn-up analysis was made to #2, #3 and #5. Corresponded FIMA(%) was 27.9, 20.8 and 28.0 for #2, #3 and #5, respectively. EFPD was 97.

(2) Vol(ml): a total volume of sampled gas. Detected gases adding to Kr and Xe were N_2(<0.1%) and O_2(<0.1%) from air, Ar(<0.1%)from welding and the others.

(3) This data was omitted due to suspicious of data reliability.

(4) FGR of the specimens #2-#6 was of order of 100% if one used diffusion theory in calculation. Details on FGR, see section 4.3 in the text.



No	Parts	Material	Dimension
①	Container	Nb-1w/oZr	Cladding and end plug 10φ x 6.8φ x 1.6t
②	Fuel pin	Pt	6.6φ x 6.2φ x 0.2t
③	Fuel sheath	Nb-1w/oZr	6φ x 4φ x 1t
④	Spacer	Nb-1w/oZr	3.5φ x 0.5t
⑤	Wool	Aluminum	
⑥	Disk fuel	PuO_2-ZrO_2 or PuO_2-ThO_2	

(Unit:mm)

Fig. 1 Longitudinal section of the test specimen used in JRR-3M irradiation. From inside to outside of the capsule, a specimen made of disk fuel, spacer, fuel sheath, fuel pin and container.

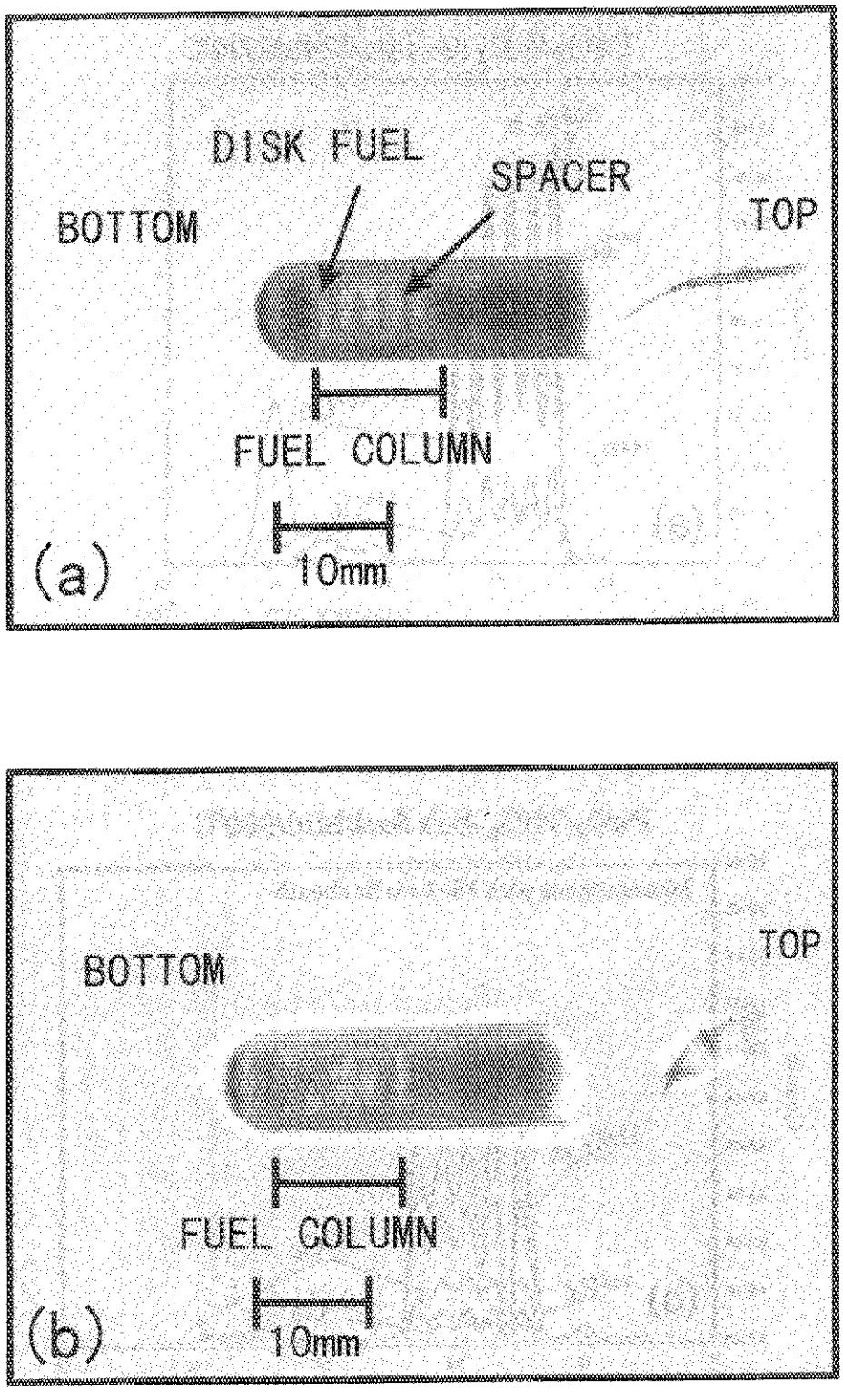


Fig. 2 X-ray photograph for irradiated disk fuel taken at burn-up of 27MWd/kg · Pu .
(a) PuO_2-ZrO_2 disk fuel/mid location/990℃
(b) PuO_2-ThO_2 disk fuel/mid location/990℃
The latter relocated greatly than the former.

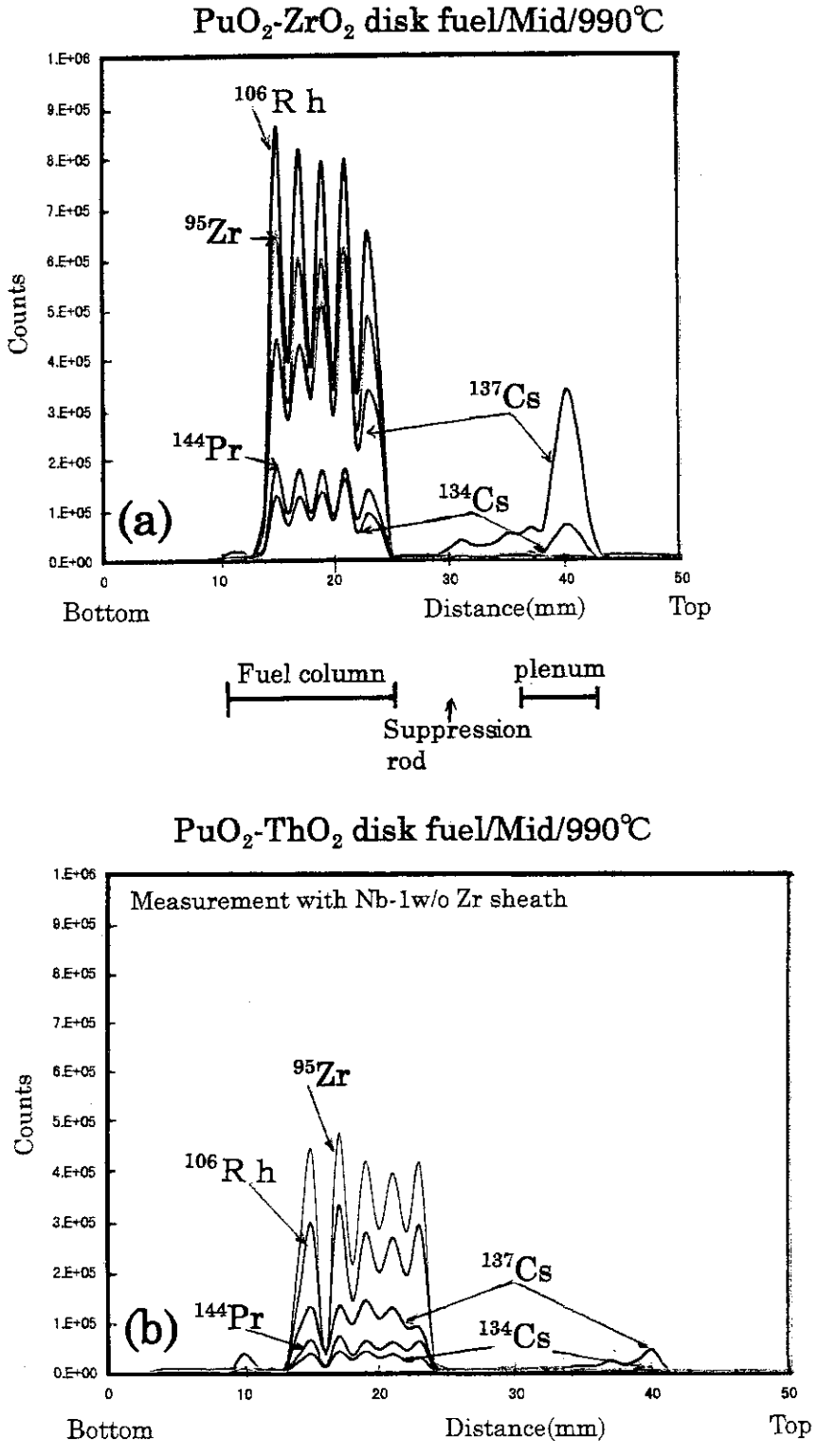


Fig. 3 Results of axial gamma scanning represented by selected five nuclides. (a) PuO_2-ZrO_2 disk fuel /mid./990℃ where scanning was carried out without a container. (b) PuO_2-ThO_2 disk fuel/mid. /990℃, scanned with a container. BU=27MWd/kg

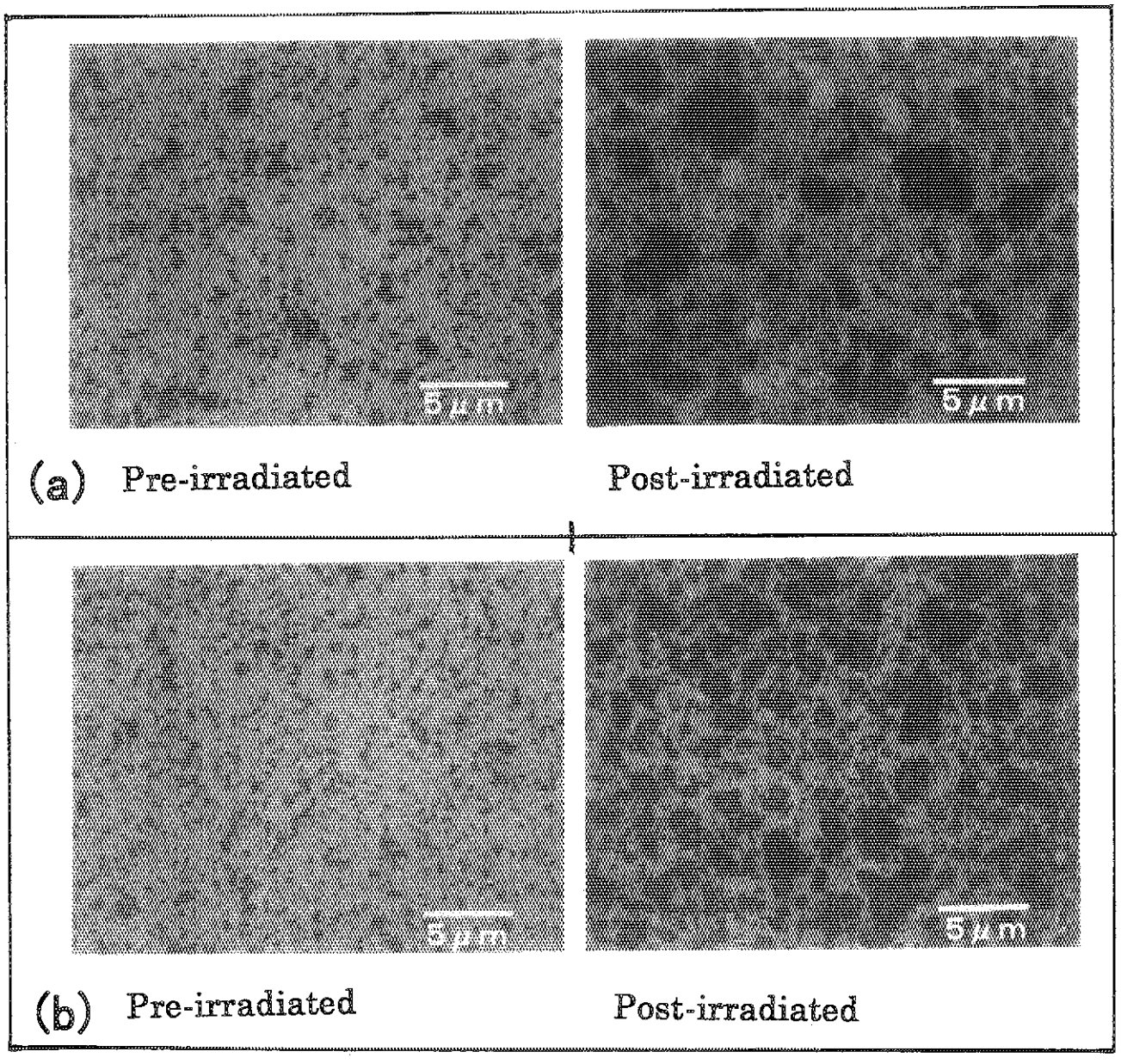


Fig. 4 As-polished microstructure of the tested disk fuel specimens.
(a) PuO_2-ZrO_2 fuel: (Left: pre-irrad. 18% porosity, Right: post-irrad. /mid. / 990℃, 46% porosity)
(b) PuO_2-ThO_2 fuel (Left: pre-irrad. 17% porosity, Right: post-irrad. /mid. / 990℃, 39% porosity)
Burn-up at sampled locations was 27MWd/kg

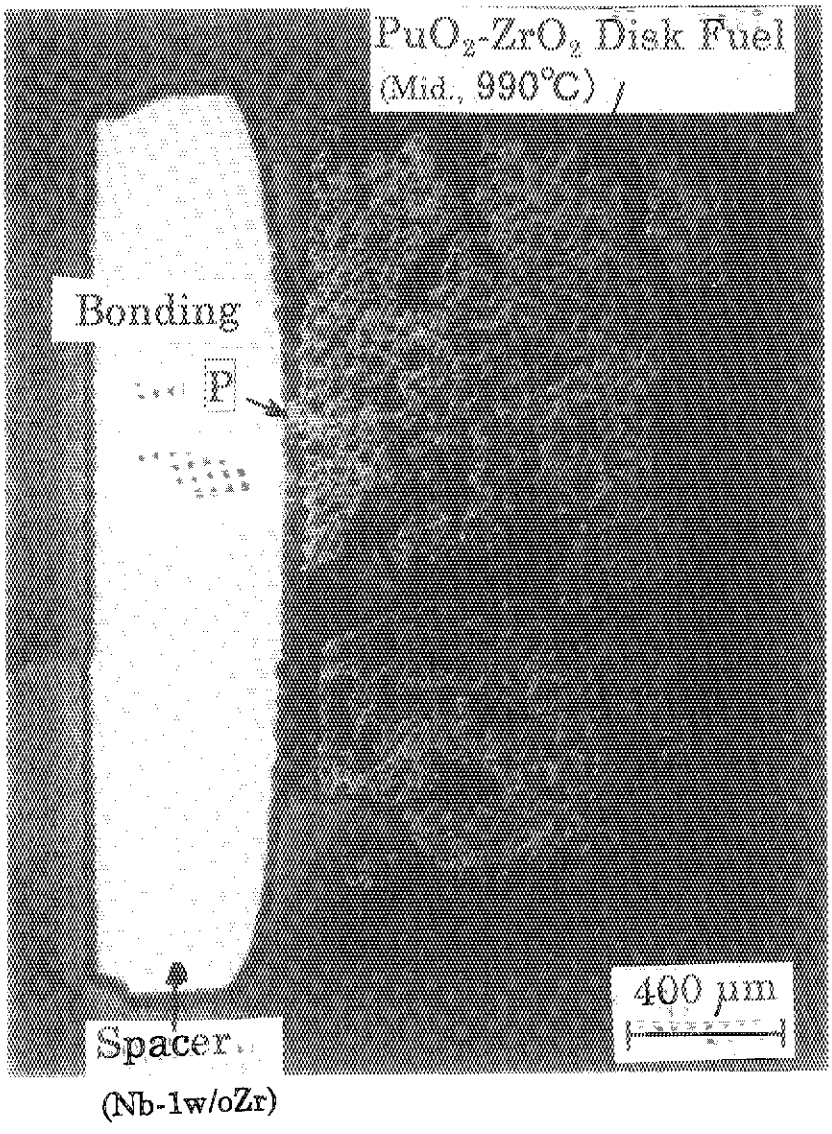


Fig. 5(a) Condition of bonding occurred between PuO_2-ZrO_2 fuel (mid.,990℃) and spacer made of Nb-1w/oZr at average burn-up of 20 MWd/kg. Longitudinal section at bonded location. Analysis by scanning electron microscopy was made at location "P".

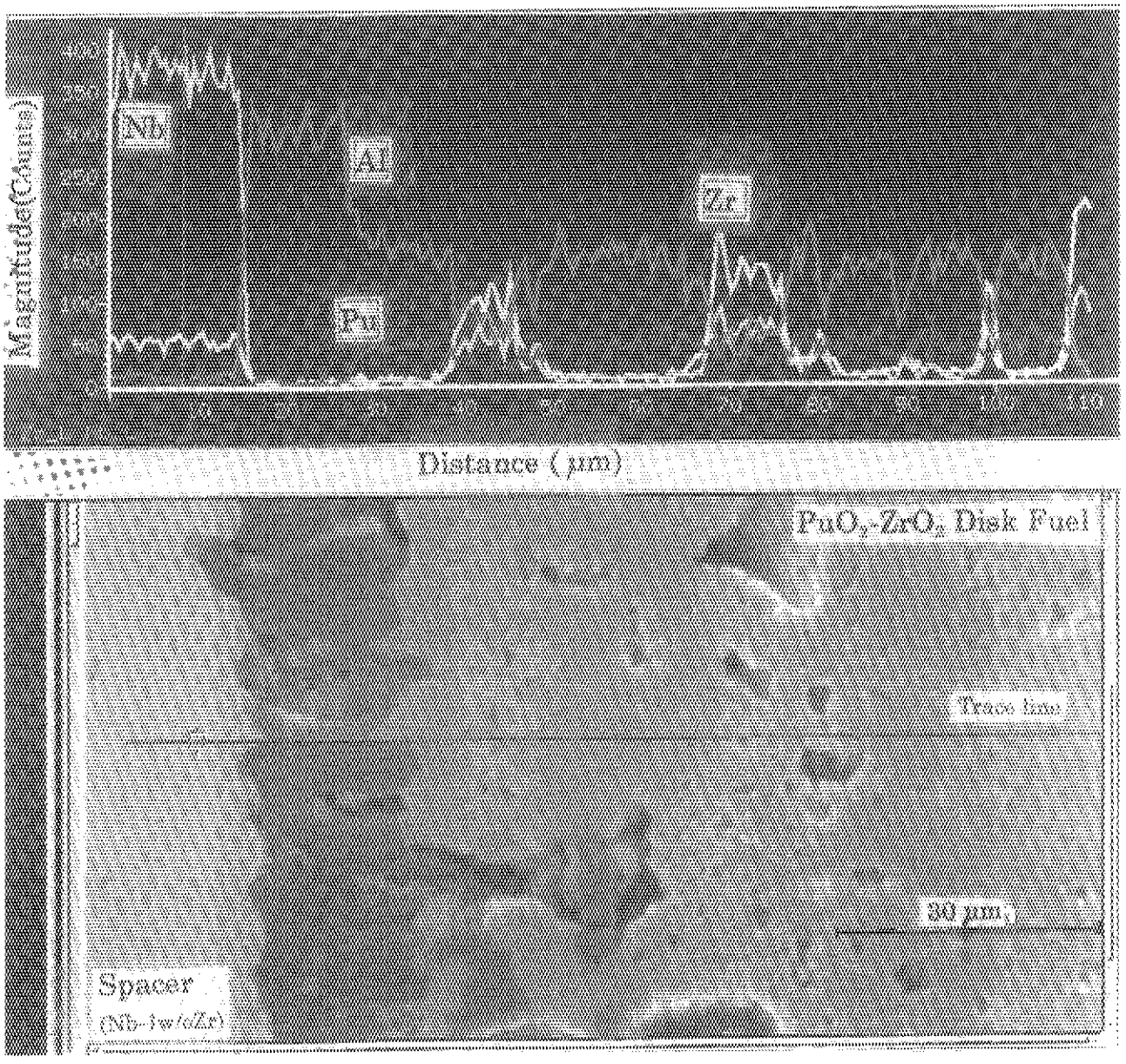


Fig. 5(b) (Bottom): Magnified picture at bonded area obtained from location "P". Along horizontal line individual magnitude of elements Zr, Nb, Al and Pu were measured.
(Top): Results of measurement

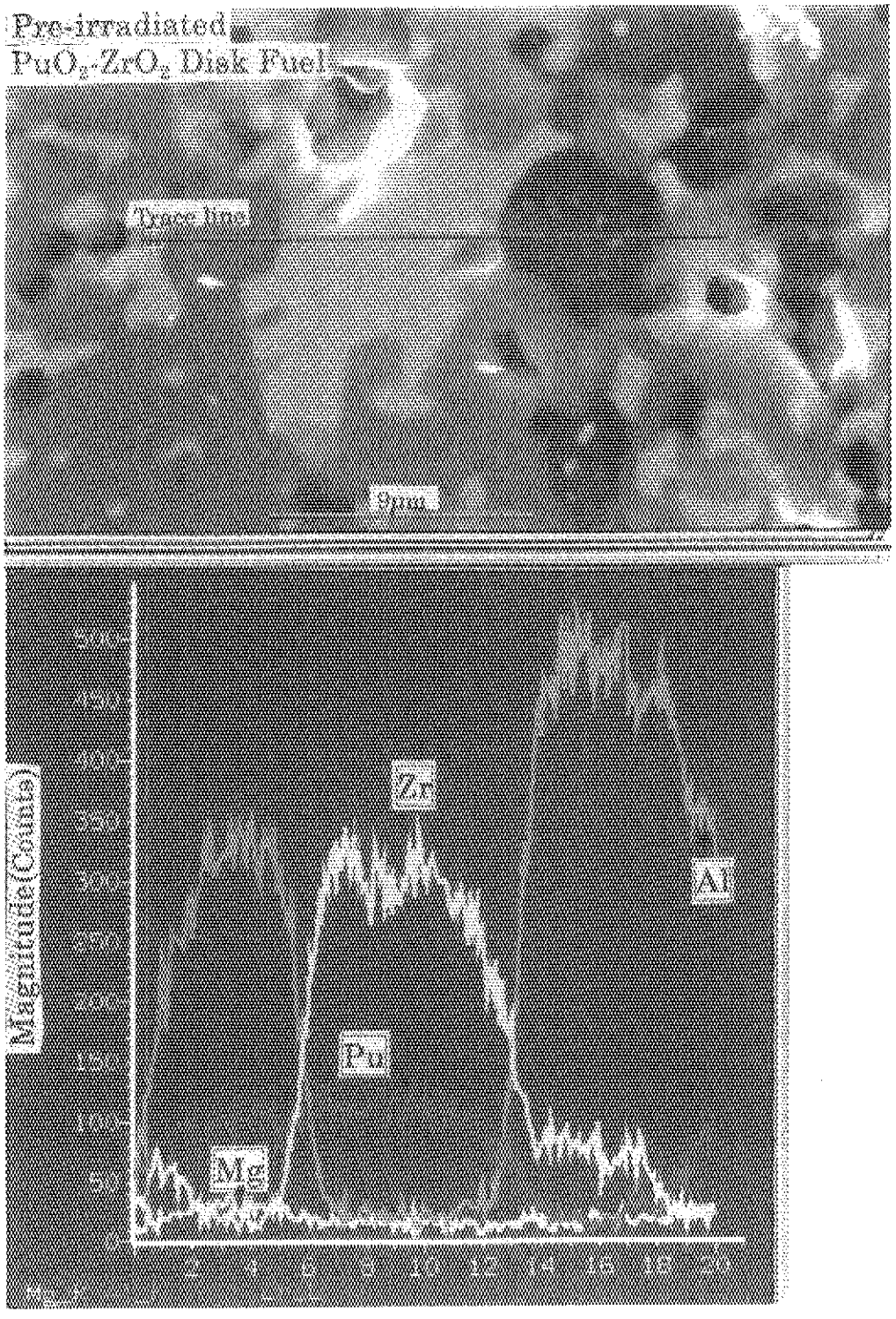


Photo.1(a) (Top) As-fabricated microstructure (SEM image) of PuO_2 - ZrO_2 disk fuel taken from center position. (Bottom) Distribution of metal elements involved in the fuel. Detection was made along the trace line shown in the SEM image.

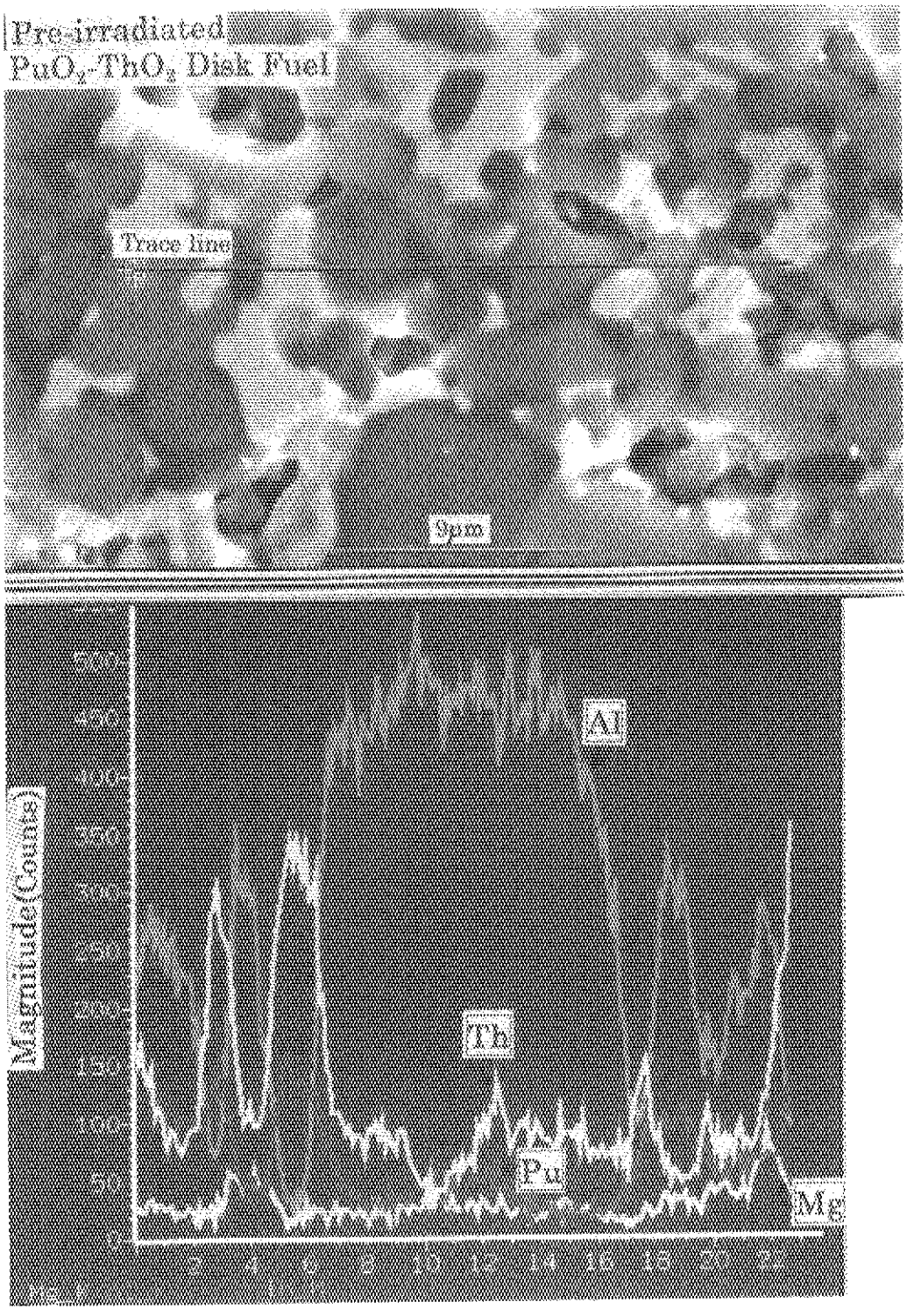


Photo.1(b) (Top) As-fabricated microstructure (SEM image) of PuO_2– ThO_2 disk fuel taken from center position. (Bottom) Distribution of metal elements involved in the fuel. Detection was made along the trace line shown in the SEM image.