# IRRADIATION PLAN OF DUPIC FUEL AT HANARO

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Abstract

DUPIC fuel is planned to start irradiation at HANARO in January 2000. Before the campaign of DUPIC fuel irradiation, preliminary irradiation of simulated DUPIC fuel started in July 1999. A mini-element of five pellets was designed and analyzed in the condition of HANARO. The performance of mini-element was carried out with modified material properties of the DUPIC pellet. In addition, the design of open-basket type capsule was analyzed. All those results satisfied the criteria of HANARO. According to the results of this irradiation, irradiation condition at HANARO will be fixed and the design of the fuel and capsule will be proved.

## 1. Introduction

The concept of direct use of spent pressurized water reactor (PWR) fuel in Canada deuterium uranium (CANDU) reactor is a dry processing technology to manufacture CANDU fuel from spent PWR fuel material without separating fissile materials and stable fission products from the spent fuel. The concept was proposed and termed Direct Use of Spent PWR Fuel in CANDU Reactors (DUPIC) fuel cycle by Korea Atomic Energy Research Institute (KAERI) and Atomic Energy Canada limited (AECL) in participation of USA in 1991.

The DUPIC fuel cycle builds upon CANDU reactor fuel cycle flexibility and the synergism between PWRs and CANDU reactors. Spent PWR fuel typically contains 0.9 wt % fissile uranium and 0.6 wt % fissile plutonium, which exceeds the natural uranium fissile content of 0.711 wt %. The neutron economy of a CANDU reactor is sufficient to allow for DUPIC fuel to be used in a CANDU reactor, which is originally designed for natural uranium fuel, even if the neutron absorbing fission products originating from the spent PWR fuel remain in the DUPIC fuel. The concept of the DUPIC fuel is similar to

Atomics International Reduction and Oxidation (AIROX), which was developed in the middle of 1950s in USA. However, it was mainly to reuse the spent PWR fuel in PWR or fast breeder reactor (FBR) with addition of enriched materials.

There are two conventional nuclear fuel cycles. (1) once-through direct disposal fuel cycle, in which the spent PWR fuel is not recycled but treated for the final disposal, and (2) recycling fuel cycle, in which the fissile elements such as uranium and plutonium are separated from the fission products by wet processes and refabricated into mixed oxide (MOX) fuel for PWRs or FBRs. The DUPIC fuel cycle is suggested as one of the alternatives. In the DUPIC fuel cycle, the spent PWR fuel, having a nominal burnup of 35 MWd/kgHE (megawatt-day/kg-heavy elements), would be processed into DUPIC fuel for the additional burnup of about 15 MWd/kgHE through irradiation in a CANDU reactor, which is about twice the burnup of natural uranium CANDU fuel.

The DUPIC fuel cycle offers several benefits to countries with both PWR and CANDU reactors. The benefits include the additional energy produced in a CANDU reactor, efficient natural uranium utilization, and a significant reduction in spent fuel arisings through proliferation resistant way of recycling the spent fuel.

## 2. Fuel Performance of DUPIC

A premise in the DUPIC fuel cycle development is to make a maximum use of existing CANDU reactor design to burn the DUPIC fuel. The inherent features of the CANDU reactor, such as the excellent neutron economy and on-power refuelling result in an unsurpassed degree of fuel cycle flexibility, enabling DUPIC fuel to be utilized safely and economically in existing reactors. The detailed reactor physics assessment such as the maximum channel and bundle power, channel power peaking factor, power distribution, and the reactor regulating system, etc. have shown that the DUPIC fuel can be fuelled in the current CANDU-6 reactors without significant modifications. However, the detailed assessment on the handling of the radioactive DUPIC fuel at reactor fuelling system may be required.

In fuel performance viewpoint, the main characteristics of DUPIC fuel is its initial content of fission products as impurities. Since the thermal conductivity is, therefore, expected to be lower, the centerline temperature of the DUPIC fuel pellet would be higher than the standard CANDU fuel, which causes adverse effects on the fuel behaviors such as fission gas release, swelling and thermal expansion of pellets. Also, the burnup of DUPIC fuel is twice the standard CANDU fuel, the behavior of the high burnup DUPIC pellet and the pellet-cladding-interaction needs to be further evaluated through the irradiation tests at research reactors.

## 3. Irradiation Plans

To verify the performance of DUPIC fuel, KAERI has several irradiation plans of DUPIC pellets. Then follow the irradiations of DUPIC elements and bundles. Four irradiations of DUPIC pellets are scheduled shown as Table 1.

The first irradiation of DUPIC pellet is scheduled to start in July 1999. The main purpose is "Proof of Concept of the Design". It includes the capsule design verification and establishment of irradiation conditions at HANARO loading simulated DUPIC pellets. Therefore power is normal and discharge burnup is short. Only limited instuctions like thermal behavior will be estimated from the results of post irradiation examination because non-instrumented capsule is used.

Next irradiation starts in the early 2000. Main difference from the first irradiation is to load lead-DUPIC pellet with normal discharge burnup and linear power. As the DUPIC fuel is radioactive, the remote handling verification is one of the main purposes. The thermal behavior is compared with the results of previous irradiation.

From the third irradiation, semi-instrumented capsule will be used to get the informations of the irradiation behavior of DUPIC pellets. Thermocouple and other instruments will be mounted into the pellets and collect the in-situ thermal information. In next irradiation, pressure sensor will be attached in addition to the thermocouple.

## 4. Design and Manufacturing of Simulated DUPIC Fuel for Irradiation at HANARO

### 4.1 Development of capsule

HANARO was newly constructed at KAERI site. Therefore there are several things to solve before the irradiations of DUPIC fuel. Most of all, the development of irradiation test capsule was urgent. KAERI has developed simple basket type non-instrumented capsule for irradiation of DUPIC fuel. The pressure drop and vibration tests were carried out and proved to be compatible with HANARO fuel.

The concept of capsule comes from the design of HANARO fuel. Top and bottom part of the capsule is same as HANARO fuel, the central rod supports the main configuration as same manner. Only the difference is fuel parts. Instead of 18 fuel rods, an aluminum alloy tube is used to connect top and bottom plates. Inside the tube, element assembly of three mini-elements is loaded. In one mini-element, 5 pellets with alumina spacer and plenum are loaded, and the both end of cladding is sealed with TIG welding method. The material of cladding and endcap is type 316L. In Figure 1 and 2, the capsule, the element assembly and mini-element are shown.

4.2 Manufacturing of simulated DUPIC pellets

Several pellets simulating the composition and microstructure of DUPIC fuel were fabricated from resintering powder through the OREOX process of simulated spent fuel pellets, which were prepared from the mixture of stable forms of constituent nuclides. The fission product composition of irradiated $UO_2$ is determined by its starting enrichment and irradiation history. The ORIGEN (Oak Ridge Isotope Generation and Depletation) Code was used to calculate fission product inventories added into $UO_2$ powder. The fourteen elements listed in Table 2 represent major fission products except the volatile elements. The key issues for producing pellets that replicate the phases and the microstructure of the irradiated fuel are to achieve a submicrometre dispersion during mixing and diffusional homogeneity during sintering.

To replicate the complex phase mocrostructure of high-burnup fuel, it is necessary to achieve uniform dispersion and phase equilibrium during fabrication of simulated PWR spent fuel. This implies that the fission products are to be mixed homogeneously on a submicrometre scale, and then heated up to a sufficiently high temperature to achieve diffusion rates that ensure homogeneity on an atomic level. For these reasons, $UO_2$ powder and additives were mixed for 2 hour in turbular and milled 5 times with 150 in RPM for 15 min. in attrition miller. To confirm the homogeneity of additives in the powder typical elements were analyzed by chemical analysis at the three points -top, middle and bottom of bottle- of the mixed and 1st, 3rd and 5th milled powder. The results of chemical analysis represent in Table 3. The concentrations of typical elements of mixed powder slightly vary as sampling point of bottle and the variety of their concentrations after milling is reduced. It can be said that the additives distribute homogeneously in the powder after milling.

In order to increase flowability and packing ratio in a compaction die, the mixed powder was granulated with 1 ton/cm$^2$ pre-compaction pressure. The mixed powder was pressed with 1.30 ~ 1.66 ton/cm$^2$. The green pellets were sintered at 1800 ℃ for 12 hour under flowing $H_2$ with heating rate and cooling rate of 3.5 ℃/min..

Simulated DUPIC fuel can be fabricated by compaction and resintering the powder through the OREOX process of simulated PWR spent fuel. The OREOX process was repeated 3 times at 450 ℃ of oxidation temperature and at 650 ℃ of reduction temperature. During the OREOX process at high temperature, simulated PWR spent fuels were subject to phase transform accompanied with the volume changes. This volume changes pulverized the powder and developed more micro-crack to increase the sinterability of the powder. This powder is called simulated DUPIC powder. Simulated DUIPC fuel can be fabricated using simulated DUPIC powder by the same method as above.

The sintered density of simulated spent fuel pellet, measured by immersion method, increases from 10.227 $g/cm^3$ to 10.257 $g/cm^3$ (95.116 ~ 95.395% of TD) as compaction pressure increases from 1.30 $ton/cm^2$ to 1.66 $ton/cm^2$. Density of simulated DUPIC fuel pellet ranges from 10.365 $g/cm^3$ to 10.398 $g/cm^3$ (96.560 ~ 96.867% of TD). The densities of simulated DUPIC fuel pellets are higher than those of simulated spent fuel pellets.

Microstructure of the pellets represents in Figure 3. Polygonal, equiaxed $UO_2$ grains form simulated fuel matrix. The grain size of simulated DUPIC fuel pellet is larger than that of simulated spent fuel because the sinterability of simulated DUPIC powder through the OREOX process is improved. Small intergranular sintering pores are also present. The shape and size of the grains are typical for $UO_2$ nuclear fuel pellets. The spherical metallic precipitates(about 1 m diameter) of additives distribute in grain boundary. Their components are confirmed by electron probe X-ray microanalysis(EPMA) and the results are shown in Table 4. They consist of Zr, Mo, Ru, Pd, Sr, Rh, Ba, Ce and Te and Mo and Ru are the main components of the metallic phases. These metallic precipitates are of the same size but difference composition as those found in previous investigations of the simulated spent fuel.[1-3] In previous study on microstructural features of the irradiated fuel, the metallic precipitates consist mainly of the -phase(hcp) of the Mo-Ru-Pd-Rh alloy and two other metallic phases were found in small amounts in the residues : a tetragonal intermetallic compound (-phase) of $Mo_{0.7}Ru_{0.3}$, and a cubic-phase alloy, rich in Pd.[4] In this study Zr, Sr, Ba, Ce and Te except Mo, Ru, Rh and Pd are found because of interference from the surrounding matrix.[5,6]

Typical SEM images and characteristic X-ray pictures of simulated spent fuel by electron probe microanalysis are shown in Figure 4. The bright and dark regions in the SEM image are the metallic and oxide precipitates, respectively. The compositions of the oxide precipitates and the oxides dissolved in the matrix are shown in Table 5. Previous investigations of the simulated nuclear fuels reported a barium zirconate-type perovskite phase as representative for the third group of fission products. The oxide precipitates in Figure 4 are expected as the cubic perovskite structure of a $BaZrO_3$-type compound.

Bacause the homogeneity of additives was confirmed in the powder after attrition milling and the microstructure of simulated spent fuel is in agreement with other study, simulated spent fuel pellets can be said to be well fabricated. Then the simulated DUPIC fuel pellets are well fabricated too.

5. Irradiation of Simulated DUPIC pellets at HANARO

The summary of irradiation of simulated DUPUC pellets is as follows.

- start of irradiation : July 1999
- materials : simulated DUPIC pellets (35,000 MWd/THM)
- estimated discharge burnup : 1,500 MWd/THM
- irradiation hole : OR4
- maximum linear power : 488 W/cm
- maximum centerline temperature : 2193℃

## ACKNOWLEDGEMENTS

This work was performed under the long-term nuclear R&D program sponsored by the Ministry of Science and Technology

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Table 1. DUPIC Pellet Irradiation Plan

| Objective | Period | Remarks |
|---|---|---|
| 1. Proof of Concept of Design<br>- capsule design verification<br>- establishment of irradiation conditions at HANARO | 1999. 7<br>~1999. 9 | - Simulated DUPIC fuel<br>- low burnup<br>- power : ~ 420 W/cm<br>- non-instrumented |
| 2. DUPIC Lead-Pellet Irradiation<br>- irradiation behavior of DUPIC pellet<br>- development of remote assembling and handling technology | 2000. 1<br>~2000. 12 | - average burnup<br>- power : 420 W/cm<br>- non-instrumented |
| 3. Thermal Behavior of DUPIC Pellet<br>- analyzing irradiation behavior<br>- produce thermal behavior information | 2001. 1<br>~2001. 12 | - temperature, flux monitoring<br>- instrumented<br>- average burnup |
| 4. Fission Gas Release of DUPIC Pellet<br>- analyzing irradiation behavior<br>- produce F.G.R information | 2002. 1<br>~2002. 12 | - temperature, pressure flux monitoring<br>- instrumented<br>- average burnup |

Table 2. Contents of fission products added in $UO_2$ powder

| Fission Products | Composition(wt%) | Fission Products | Composition(wt%) |
|---|---|---|---|
| $Zr(ZrO_2)$ | 0.422 | $(Nd_2O_3)$* | 0.131 |
| $Mo(MoO_3)$ | 0.392 | $Nd(Nd_2O_3)$ | 0.476 |
| $Ru(RuO_2)$ | 0.269 | $Sm(Nd_2O_3)$ | 0.101 |
| $Pd(PdO)$ | 0.187 | $Sr(SrO)$ | 0.084 |
| $Ba(BaCO_3)$ | 0.218 | $Y(Y_2O_3)$ | 0.052 |
| $La(La_2O_3)$ | 0.143 | $Rh(Rh_2O_3)$ | 0.049 |
| $Ce(CeO_2)$ | 0.278 | $Te(TeO_2)$ | 0.058 |

Table 3. The concentrations of typical elements of additives in the powder after mixing and milling

| Sampling point | Mo | Zr | Nd |
|---|---|---|---|
| Top of the mixed powder | 0.26 | 0.29 | 0.53 |
| Middle of the mixed powder | 0.30 | 0.29 | 0.55 |
| Bottom of the mixed powder | 0.32 | 0.34 | 0.61 |
| 1st milled powder | 0.31 | 0.27 | 0.55 |
| 2nd milled powder | 0.31 | 0.29 | 0.56 |
| 3rd milled powder | 0.32 | 0.30 | 0.57 |

Table 4. EPMA results for metallic precipitates and matrix

| point | U | Zr | Mo | Ru | Pd | Sr | Rh | Ba | La | Ce | Te |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | 38.26 | 1.24 | 34.14 | 19.42 | 1.24 | 2.38 | 2.18 | | | | 1.13 |
| 2 | 98.33 | 0.29 | | | | 0.55 | | | | 0.43 | |
| 3 | 23.97 | 0.88 | 27.92 | 41.56 | 1.59 | 1.30 | 2.83 | | | | |
| 4 | 12.70 | 1.88 | 42.08 | 32.57 | 2.10 | 2.94 | 1.71 | 1.58 | 1.15 | 1.29 | |
| 5 | 26.88 | | 31.98 | 30.13 | 2.28 | 1.74 | 2.05 | 1.36 | 1.47 | 1.08 | 0.47 |

1,3,4,5 : metallic precipitates, 2 : matrix

Table 5. EPMA results for the oxide precipitates and the oxides dissolved in the matrix

| Point No | U | Zr | Nd | Ba | Mo | Remark |
|---|---|---|---|---|---|---|
| 1 | 98.96 | 0.40 | 0.65 | | | matrix |
| 2 | 13.57 | 34.0 | | 52.42 | | oxide precipitate |
| 3 | 28.76 | 28.76 | | 43.93 | | " |
| 4 | 20.53 | 31.50 | | 47.97 | | " |
| 5 | 12.40 | 34.41 | | 53.19 | | " |
| 6 | 86.49 | 5.73 | | 7.78 | | " |
| 7 | 99.26 | 0.47 | 0.27 | | | matrix |
| 8 | 98.09 | 0.72 | 0.87 | | 0.31 | matrix |

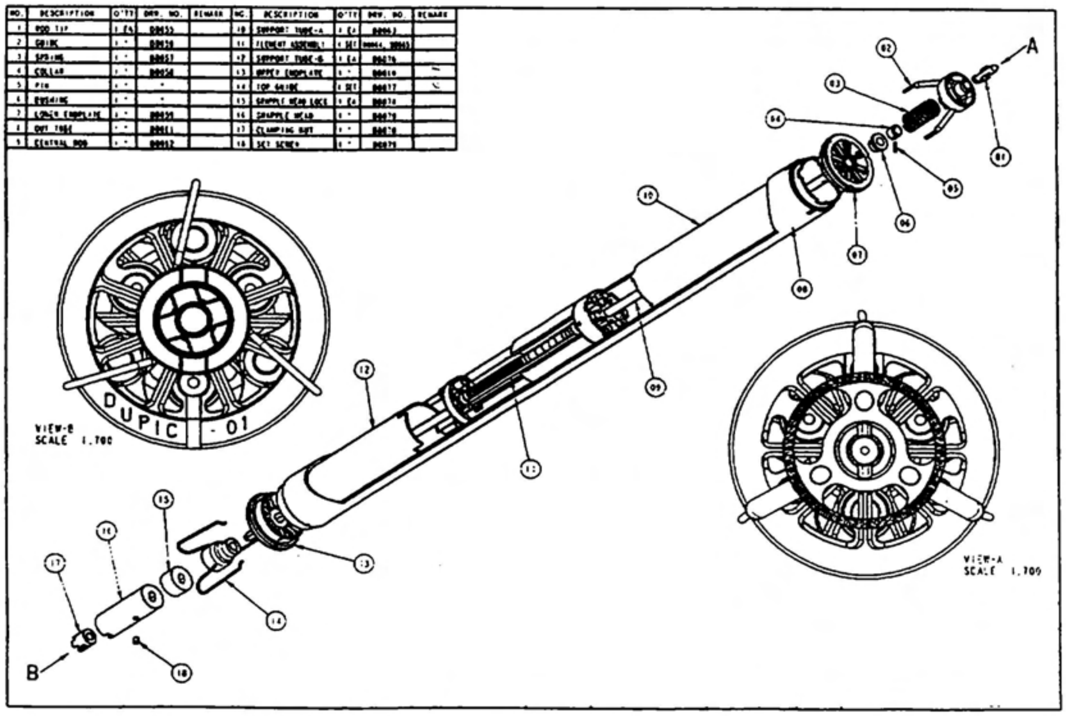


Figure 1. DUPIC capsule for irradiation test of simulated DUPIC pellets

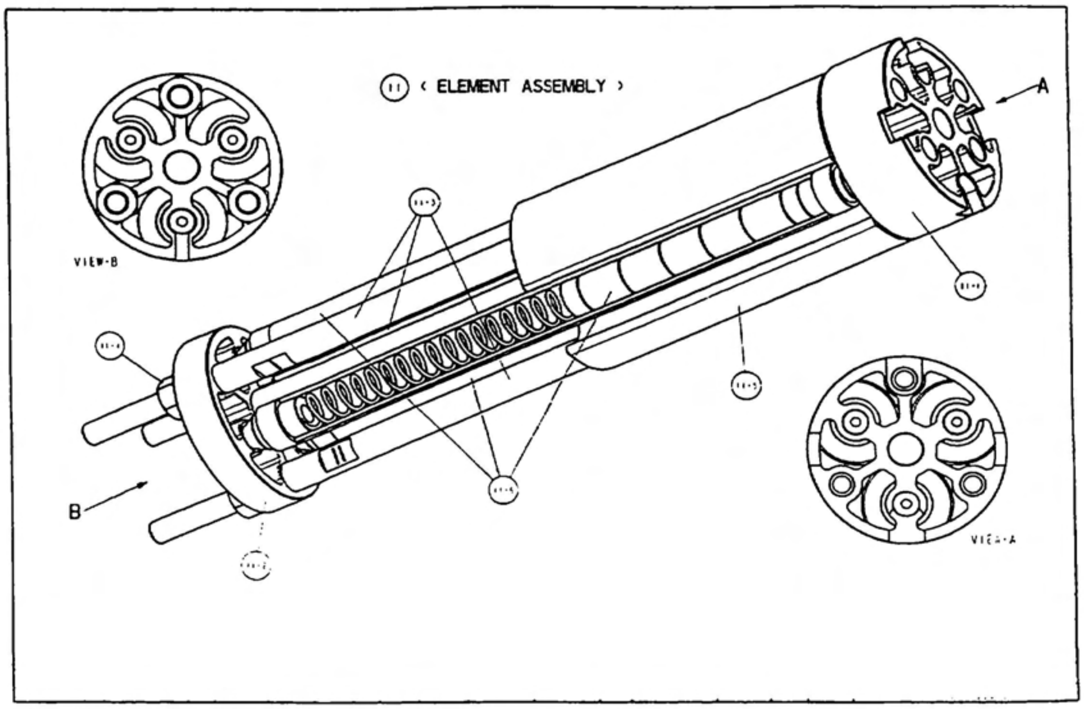


Figure 2. Detail diagram of element assembly in DUPIC capsule

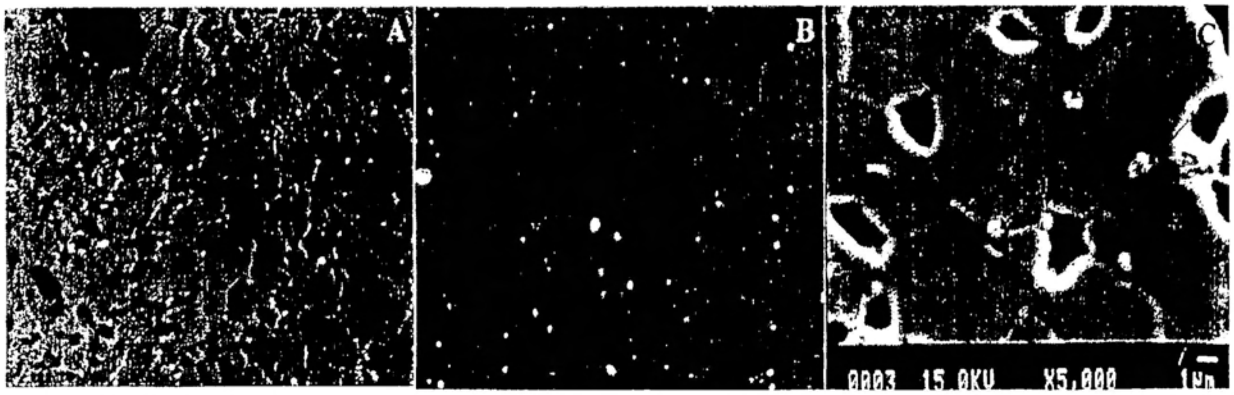


Figure 3. SEM image of a polished and etched surface of pellets: A) simulated spent fuel.($\times$1000), B) simulated DUPIC fuel.($\times$1000) showing quiaxed matrix grains and precipitates, C) simulated DUPIC fuel.($\times$5000) showing metallic precipitates between matrix grains.

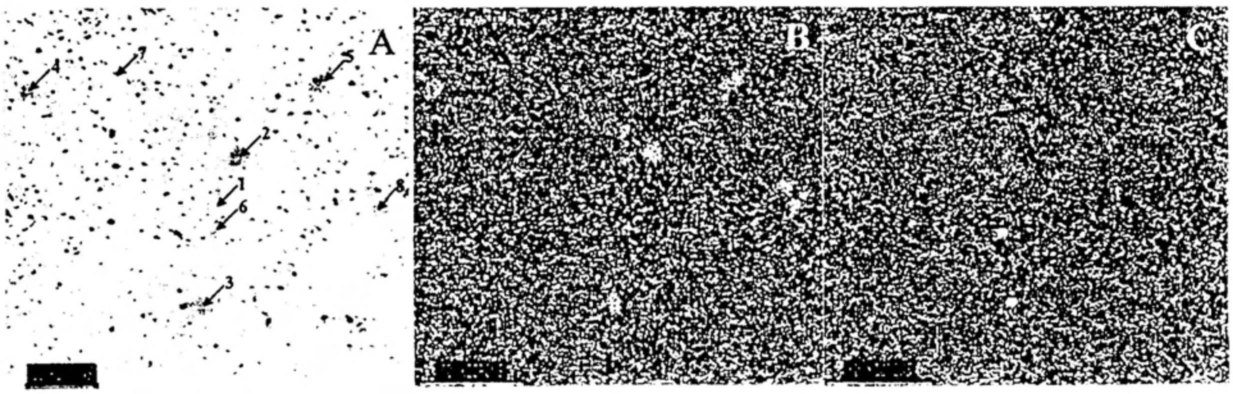


Figure 4. Photomicrograph and X-ray pictures(EPMA) of the simulated spent fuel. The bright and dark regions are metallic precipitates and oxide precipitates, respectively. : (a) microstructure, (b) Mo, (c) Zr