Microstructure Analysis of Irradiated NUE and NU Fuel in NPIC Hot Cells

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Abstract

The natural uranium equivalent (NUE) fuel development program is proposed by Third Qinshan Nuclear Power Co., Limited (TQNPC) and CANDU (Canada Deuterium Uranium) Energy, to improve the uranium resource utiliztion rate and reduce the storage pressure of spent fuel. In order to evaluate fuel irradiation performance and pellet cladding interaction of NUE fuel elements, irradiation tests were performed in Qinshan CANDU 6 reactor Unit1 and then post-irradiation examination were carried out. The irradiation swelling along the axial and radial, the crack distribution, and the shape, size and distribution of grain in UO_2 pellet of NUE and natural uranium (NU) fuel were analyzed by optical microscopy in NPIC hot cells. The results show that there is no significant difference on microstructure between NUE and NU fuel.

1. Introduction

The recycling of spent fuel in heavy water reactor （HWR）can efficiently improve the uranium resource utilization rate and reduce the storage pressure of spent fuel[1]. It has great significance for sustainable development for nuclear power and the realization of building the closed fuel cycle system. Thus, Third Qinshan Nuclear Power Co., Limited (TQNPC) and CANDU Energy put forward the natural uranium equivalent (NUE) fuel development program. Fuel that adopted in NUE fuel element was homogeneous mixture of recycled uranium（RU）and depleted uranium(DU). Compared with natural uranium in heavy water reactor, it has consistency in reactor core characteristics and main parameters, no changes in pellet structure, and also has the equivalent physical character. The only difference is material composition of fuel pellet. In order to guarantee the security and functional stability of of reactor operations, irradiation test and post-irradiation examination are required. Thus, in this paper the irradiation swelling along the axial and radial, the crack distribution, and the shape, size and distribution of grain in UO_2 pellet of NUE and natural uranium(NU) fuel were observed and analyzed by optical microscopy(OM) in hot cells. The results will be used to demonstrate the equivalence of the NUE fuel to the reference NU fuel and support the licensing of the NUE full core implementation, which is the main purpose of the NUE fuel development program.

2. Specimen Preparation and Test Method

2.1 Specimen Preparation

The fuel elements were of high radioactivity after irradiation, so the section, sampling, specimen preparation and metallographic examination all should be conducted in hot cell by robotic arm. The specimens were sectioned from 4 NUE fuel rods and 3 NU fuel rods by hot cell cutting milling machine in the hot-cell, Diamond disk was used to cut. A longitudinal specimen containing end cap and a transverse specimen containing the middle spacer pad were cut from each rod. Fuel rod code after irradiation, sample code and sampling location are shown in Table 1.

Table 1 Sampling Location and Quantity of Fuel Elements

Transverse Specimen				Longitudinal Specimen			
Rod Code	Sample Code	Quantity	Sampling Location	Rod Code	Sample Code	Quantity	Sampling Location
NUE-1	NUE-1-H	1	Middle Fuel Element at Spacer Pad	NUE-1	NUE-1-Z	1	Reference End
NUE-2	NUE-2-H	1		NUE-2	NUE-2-Z	1	
NUE-3	NUE-3-H	1		NUE-3	NUE-3-Z	1	
NU-1	NU-1-H	1		NUE-6	NUE-6-Z	1	
NU-2	NU-2-H	1		NU-2	NU-2-Z	1	
NU-3	NU-3-H	1		NU-3	NU-3-Z	1	

2.2 Test Method

Specimens were mounted in mounting mould by epoxy resin. Then specimens were ground in sequence through size 220#, 600# and 1200# diamond disc and polished in sequence through size 9μm, 3μm and 1μm diamond slurry. After ground and polished, the distribution of cracks and pores in pellets were examined; then got pellets etched by etchant consists of H_2SO_4:H_2O_2=1:9, grain shape and size of UO_2 pellets were examined by Leica MEF4A optical microscopy. Preparation and examination process of metallography sample for NUE and NU were shown in Figure 1.

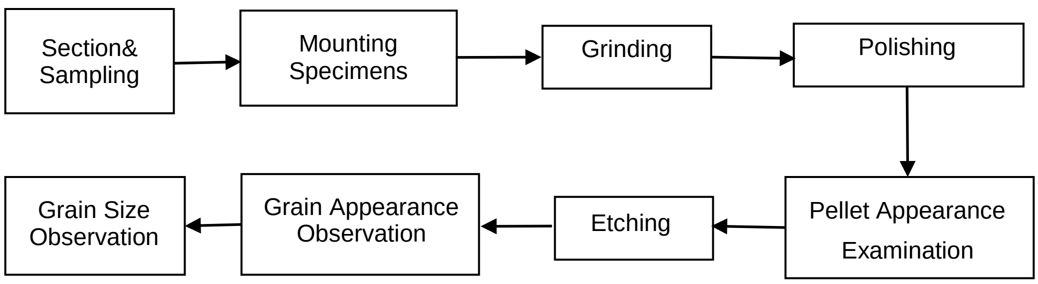


Figure 1 Preparation and examination process of sample

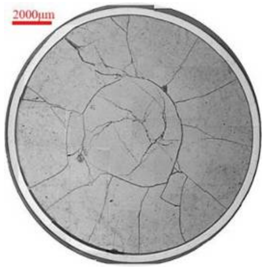
3. Result and Analysis

3. 1 Transverse Specimens

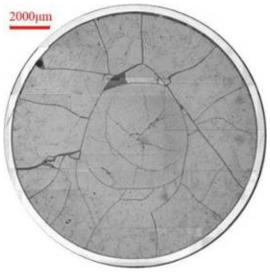
3.1.1 Overall Appearances of Transverse Specimen

Overall appearances of transverse specimens of NUE and NU fuel rods were shown in Figure 2. Pellets of both NUE and NU fuel rods were broken in different degree and there were lots of cracks in the surface in different directions. This was due to the tensile strength of UO_2 pellets is low, thus, at the thermal stress conditions of temperature gradient, cracks were created in radial and circumferential directions.

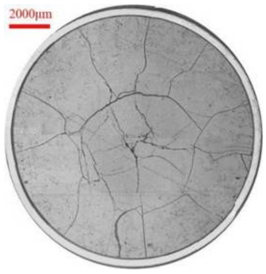
Obvious differences of the cracks between NUE and NU fuel rods were obtained. There were circumferential cracks and no obvious radial cracks inside of the circumferential cracks in NUE fuel rods (figure 2 a~c). But there were big radial cracks in the center of NU fuel rods (figure 2 d~f), which show that the source of cracks were at the center of pellet. These differences were related to fuel manufacturing operation, pellet density, structure and ambient temperature.



a.NUE-1-H

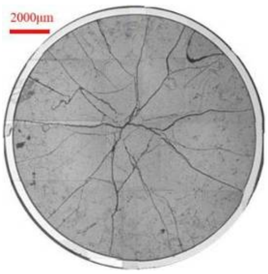


b）NUE-2-H

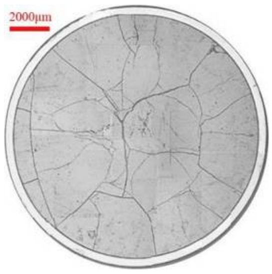


c）NUE-3-H

d）NU-1-H



e）NU-2-H specimen

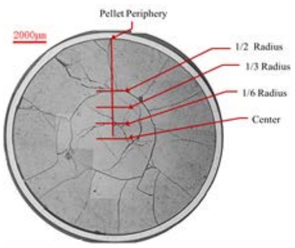


f）NU-3-H

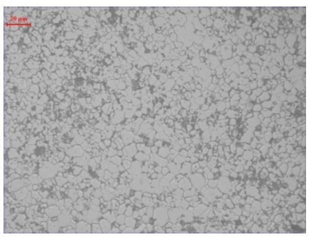
Figure 2 Overall Appearance of Transverse Specimens

3.1.2 Microstructure at Different Locations of Fuel Pellets

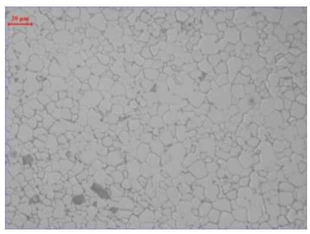
Grain size of UO_2 pellet has effects on fission gas release, thermal stability and breaking strength of fuel, thus, in this paper, the microstructure at the center, 1/6 radius, 1/3 radius, 1/2 radius and the periphery of transverse specimens of NUE and NU fuel rods were observed and analyzed (figure3-figure5). The results show that (1) the grain size distribution in corresponding area of NUE and NU fuel rods was basically the same, which range from 5.8μm to 26.7μm. (2) Grain size changed according to areas. From periphery to center, grain size gradually increased. There was significant difference between periphery and the center. Grain size at the center distributed from 21.4μm to 26.7μm and at the periphery it distributed from 5.8μm to 7.1μm. The grain size at the center of pellets was almost 3 to 5 times larger than that of the periphery.



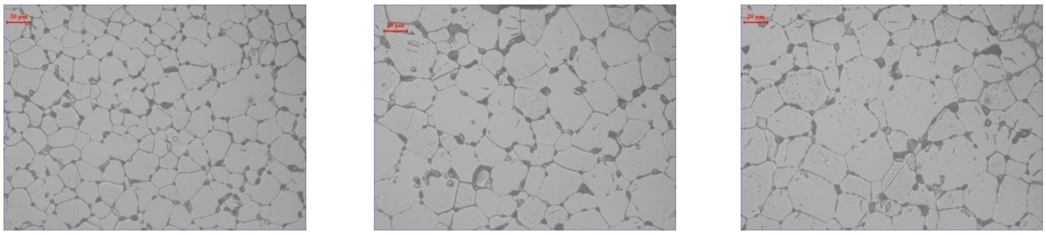
a）Transverse Section at the Five Different Radial Locations



b）Pellet Periphery

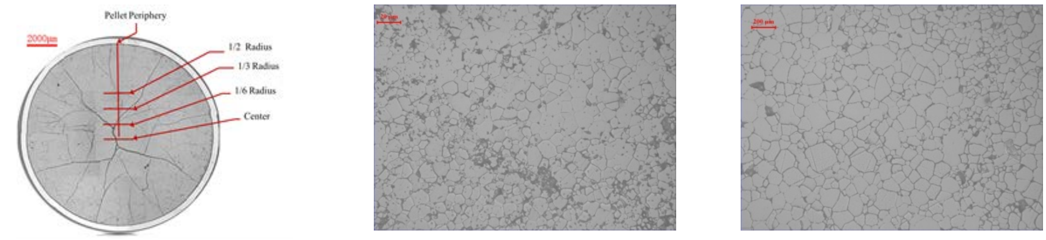


c）1/2 Radius

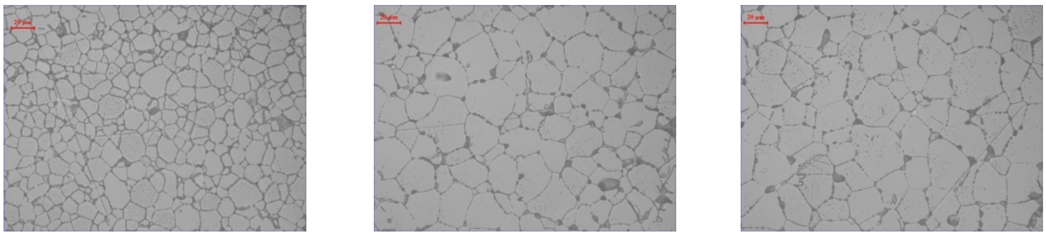


d）1/3 Radius　　e）1/6 Radius　　f）Center

Figure 3 Pellet Microstructures at the Five Different Radial Locations of 1-NUE-1-H Specimen



Transverse Section at the Five Different Radial Locations　　b）Pellet Periphery　　c）1/2 Radius



d）1/3 Radius　　e）1/6 Radius　　f）Center

Figure 4 Pellet Microstructure at the Five Different Radial Locations of 1-NU-1-H Specimen

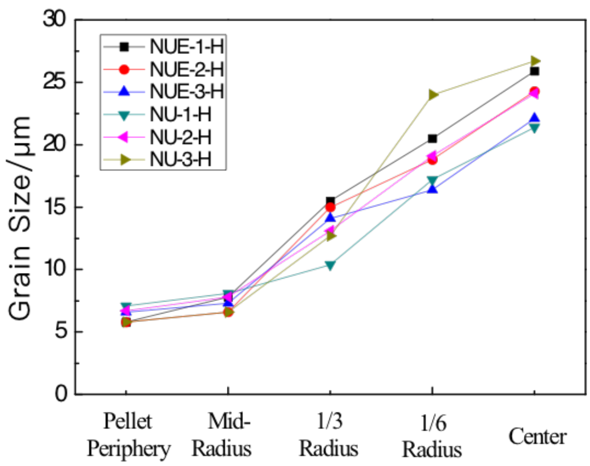
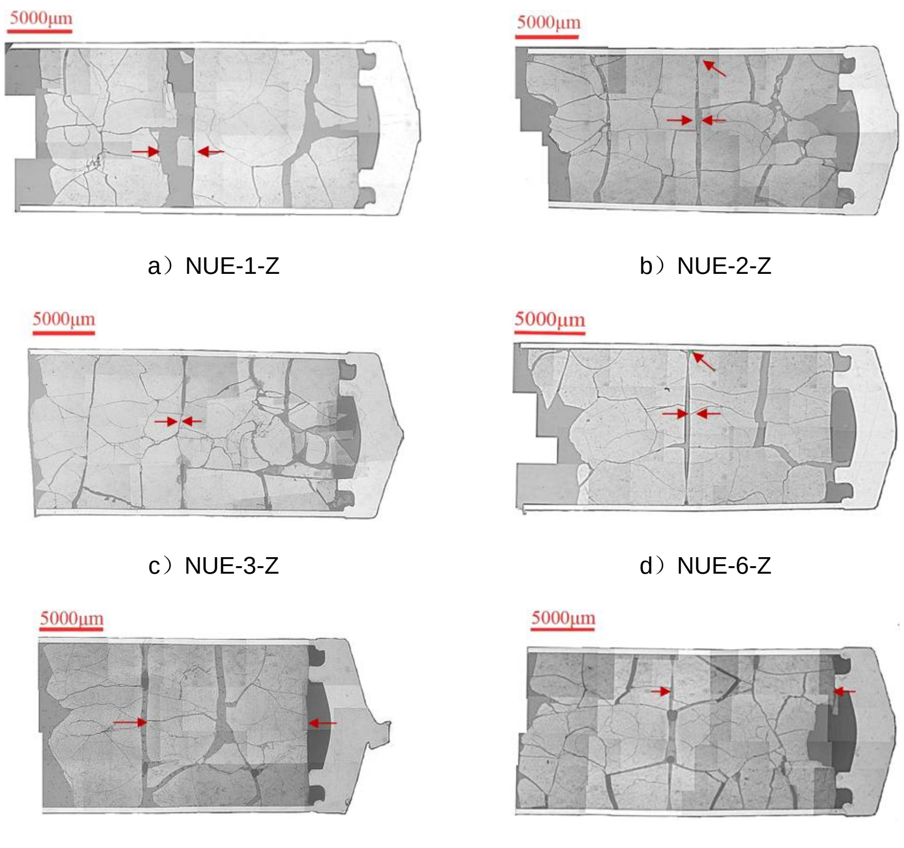


Figure 5 Measurement Result of Grain Size for Pellet's Different Locations in Transverse Specimens

3.2 Longitudinal Specimens

3.2.1 Overall Appearances of Longitudinal specimens

Overall appearances of longitudinal specimens of NUE and NU fuel rods were shown in figure 6. Pellets of both NUE and NU fuel rods were broken seriously, and there were cracks with different sizes and orientations distributing in pellets. The gap between two pellets in specimen NUE-1-Z was big and there were small fragments in the gap (figure6 a). Gaps between two pellets were similar in specimen NUE-2-Z and NUE-6-Z. Butterfly areas were relatively intact(figure6b, 7d). Pellets contacted each other at pellet-to-pellet interface in Specimen NUE-3-Z(figure 6c). There were gaps between pellet and the end cap. There was no obvious difference in the pellet-to-end cap interfaces of six longitudinal specimens.



a）NUE-1-Z　　b）NUE-2-Z

c）NUE-3-Z　　d）NUE-6-Z

e）NU-2-Z　　f）NU-3-Z

Figure 6 Overall Appearances of Longitudinal specimens

3.2.2 Microstructure at Different Locations of Fuel Pellets

The microstructure at the center, 1/6 radius, 1/3 radius, 1/2 radius and periphery in 1/4 and 1/2 pellet length along axial direction of longitudinal specimens of NUE and NU fuel rods were observed and analyzed and all locations for photomicrographs were shown in figure 7. The microstructure at the different radial locations were shown in figure 8 and figure 9.

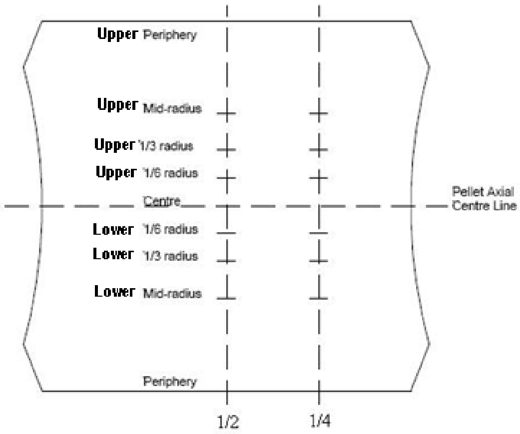
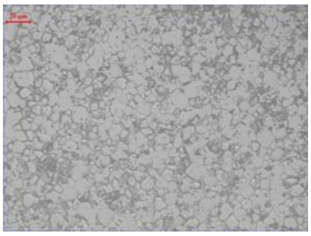
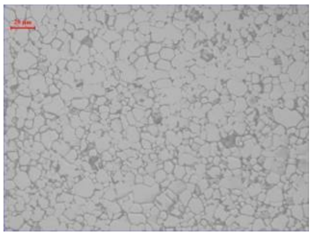


Figure 7 Longitudinal Specimen Locations for Photomicrographs

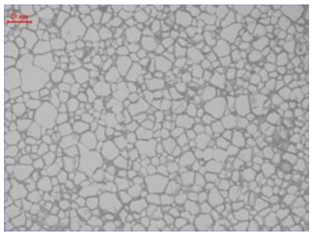
The results show that (1) The variation in grain size distribution in specimens of NUE and NU was basically the same, but there were differences in grain size. Different grind depth was regarded as the main reason. (2) Grain size changed according to areas. Grain size near the cladding pellet periphery area was small and it distribute from 4.8μm to 7.5μm. While, grain size at the center area distribute from 6.9μm to 15.5μm. Grain size in specimens of NUE and NU show the same trend: grain size increased from the periphery to the center area. It is most prominent in NUE-3-Z.



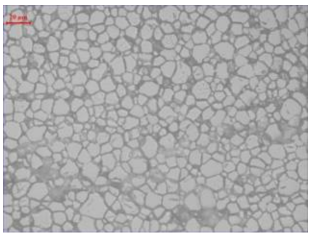
a） Upper Pellet Periphery



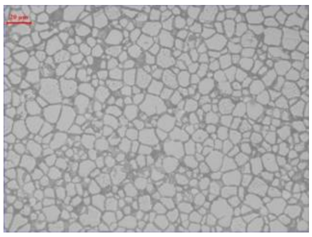
b） Upper 1/2 Radius



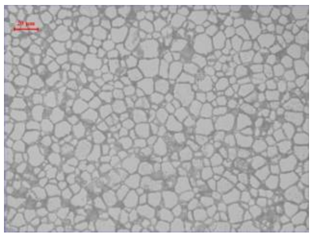
c） Upper 1/3 Radius



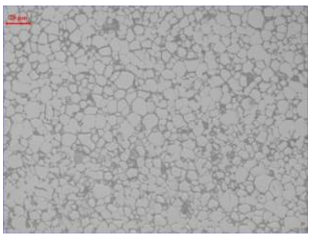
d） Upper 1/6 Radius



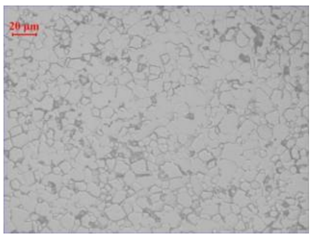
e） Center



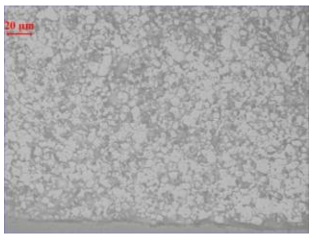
f） Lower 1/6 Radius



g） Lower 1/3 Radius

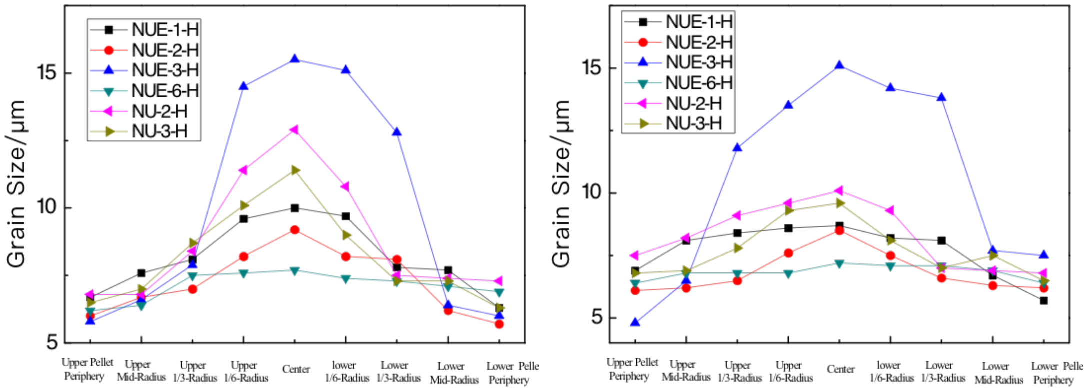


h） Lower 1/2 Radius



i） Lower Pellet Periphery

Figure 8 the Pellet Microstructure at the Different Radial Locations at 1/2 Pellet Length of NUE-1-Z Specimen



a）1/4 Pellet Length of Specimens b）1/4 Pellet Length of Specimens

Figure 9 Distribution of Grain Size for Pellet’s Different Locations in Longitudinal Specimens

All results mentioned above show that obvious growth of grain occurred in the NUE and NU pellets during irradiation. Thermal conductivity of UO_2 is very low (only 1/10 of uranium), So there have large temperature gradient and thermal stress in the center and the periphery during irradiation. As a result, grain size increases gradually from periphery area to the center of the pellets. According to the microstructure of pellets during process of operation, it can be divided into three areas: higher than 1700ºC is the columnar grain growing area; 1400ºC to 1700ºC is the equiaxed grain growing area, and lower than 1400ºC is the unchanged area. UO_2 pellet's growing under irradiation is a complex process, so in this paper, we only discuss the influence of temperature on grain growing in respect of relationship between temperature and grain size. Grain size statistical result shows that there was no obvious changes in UO_2 pellet's periphery area after irradiation, which shows that temperature in pellet's surface was not high enough for grain growing. More close to the center of the pellet, the temperature got higher and the grain size became larger. In addition, growing grain distribute in UO_2 pellet was still equiaxed grain and there was no existence of columnar grain area and center area hole because fuel pellet's temperature was not high enough for its structure to transform to high burn-up columnar grain. Similar grain sizes in corresponding area of NUE and NU pellets shows they had similar temperature and thermal gradient during irradiation. As for CANDU-PHW fuel, appropriate grain size can reduce fission fuel gas release, improve pellet's thermal stability and its breaking strength. Studies showed that grain size within 5 to 35μm was acceptable. In conclusion, compared with NU fuel element, NUE fuel element basically shows the same performance in HWR and irradiation swelling was within the allowable scope; there was no significant difference with NU pellet in pellet's broken degree and growing process. NUE can be used in heavy water reactor.

4.Conclusion

(1) Lots of cracks were found in NUE pellet and NU pellet. Analysis shows that during in-pile irradiation, temperature in the center of the pellet was high, and thermal gradient between the center and the periphery was the main reason for the serious damage.

(2) The distribution rule and appearance of grain size in corresponding area of NUE and NU fuel pellet were basically the same. Grain size changed according to areas. From periphery to center, grain size gradually increase.

(3) The irradiation swelling of NUE pellet was within the allowable scope，there was no significant difference with NU pellet in pellet's broken degree and growing process. NUE can be used in heavy water reactor.

References

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[2] Li Guan-xin, Ma Wen-jun, Zhang jie. “Manufacturing process of CANDU-6 fuel bundle for HWR nuclear power station”, The Chinese Journal of Nonferrous Metals, 14, 301-305(2004).

[3] Lewis B J, Iglesias F C, Dickson R S, et al. “Overview of high-temperature fuel behaviour with relevance to CANDU fuel”, Journal of Nuclear Materials, 394, 67-86(2009).