FISSION GAS RELEASE OF URANIUM-PLUTONIUM MIXED NITRIDE AND CARBIDE FUELS

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Abstract

Uranium-plutonium mixed nitride and carbide for advanced fast reactor fuels were irradiated at JRR-2 and JRR-2, and the fission gas release from these fuels were determined. It is confirmed from the irradiation tests that the application of the cold fuel concept to these fuels on the basis of their advantageous thermal properties may realize low fission gas release. Furthermore, the introduction of the thermal stable pellets with dense matrix and relatively large pores can lower the fission gas release to a few percent up to the burnup of 5.5%FIMA. In spite of the retention of fission gas release in the thermal stable pellets, no significant enhancement of the fuel-clad mechanical interaction was observed in the examined range of burnup. It is also suggested that the open porosity would strongly influence the fission gas release from nitride and carbide.

1. Introduction

Uranium-plutonium mixed nitride and carbide fuels have been developed as advanced fuels for fast reactors(FRs) because of the advantage in their thermal stability caused from high thermal conductivity and high melting temperature [1]. Such thermal properties enable us to use these fuels at high heating rates over 800W/cm [2], but it is also an option to apply milder heating rates, according to so-called "Cold Fuel Concept" [3]. In case of the irradiation at heating rates similar to those for MOX or metallic fuels, the central temperature of nitride and carbide fuels could be kept less than 1/2 - 1/3 of the melting temperatures in absolute temperature. At Japan Atomic Energy Research Institute (JAERI), mixed nitride and carbidefuels have been investigated from the viewpoint of the collection of the basic data of these advanced FR fuels and also fuels for transmutation of minor actinides such as Np, Am and Cm.

The research at JAERI covers fabrication technologies [4-6], property measurements [7-10], and irradiation tests [11-12]. Furthermore, studies on the electro-refining of nitride fuel started [13,14] as a pyrometallurgical reprocessing technology. With regard to the irradiation tests, thermal stable pellets with dense fuel matrix and relatively large pores have been developed and supplied besides conventional type pellets. This report describes briefly the results of the irradiation tests of uranium-plutonium mixed nitride and carbide fuels, especially focusing on the fission gas release(FGR).

2. **Program of irradiation tests**

Uranium-plutonium mixed carbide fuel was planned to be irradiated in Japan Research Reactor-2(JRR-2) and Japan Materials Testing Reactor(JMTR) from 1983 to investigate the fundamental behavior of the advanced fuels for FRs. The irradiation tests of mixed nitride fuel were followed by those of carbide fuel and started from 1988. The program of the irradiation tests is shown in Fig. 1, which includes the irradiation at JOYO performed under a collaboration with Power Reactor and Nuclear Fuel Development Cooperation(PNC). Nine fuel pins of carbide fuel and 4 pins of nitride fuel were irradiated at JRR-2 and JMTR and the post irradiation examinations(PIEs) were carried out at the hot cells of Reactor Fuel Examination Facility of JAERI.

3. **Fabrication of fuel pins and capsules**

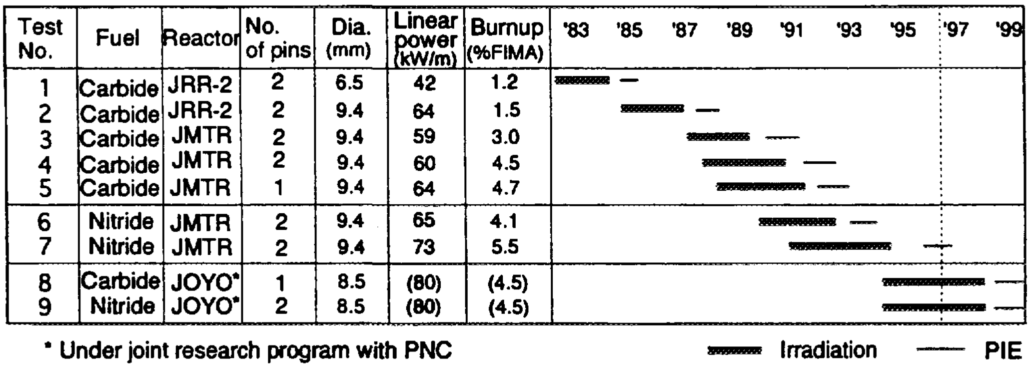
3.1 DESIGN OF FUEL PINS AND CAPSULES

All capsules containing nitride or carbide fuel pins were double-sealed, as the conceptual structure is shown in Fig. 2. An inner thermal medium was NaK and an outer one was made of an aluminum alloy so that the cladding temperature would be around 770K at the linear heating rate of 800 W/cm, the value of which is a design limit requested from the regulation for the research reactors. Several sets of thermocouple were inserted into the thermal media to monitor the burning of fuels. Main design parameters of fuel pins are as follows:

The bulk pellet density of about 85%TD(TD; Theoretical Density)
Plutonium enrichment of 20%
Cladding tubes of 20% cold work SUS 316 of 9.40 mm in diameter
Helium gas bonding

3.2 FUEL FABRICATION

Uranium-plutonium mixed carbide was synthesized by carbothermic reduction of the mixture of the dioxides with graphite at 1,723K in a vacuum. For comparison of the effects of carbon contents on the fuel behavior, two kinds of carbide pellets with different compositions were supplied for the irradiation tests; one of them had a nearly-stoichiometric composition; C/(U+Pu)1.00, and almost a single phase of fcc-structured monocarbide, (U,Pu)C. The other one had a hyperstoichiometric composition; C/(U+Pu) 1.10, and contained about 20 % of sesequicarbide, $(U,Pu)_2C_3$, in the matrix of monocarbide. The carbide discs obtained by the carbothermic reduction were milled, compacted and sintered at about 2000K in an argon gas stream. In the last irradiation campaign, the thermal stable carbide pellets were fabricated by use of pore former to obtain dense fuel matrix with relatively large pores without any significant change of pellet density of about 85%TD. Nickel metal



Test No.	Fuel	Reactor	No. of pins	Dia. (mm)	Linear power (kW/m)	Burnup (%FIMA)
1	Carbide	JRR-2	2	6.5	42	1.2
2	Carbide	JRR-2	2	9.4	64	1.5
3	Carbide	JMTR	2	9.4	59	3.0
4	Carbide	JMTR	2	9.4	60	4.5
5	Carbide	JMTR	1	9.4	64	4.7
6	Nitride	JMTR	2	9.4	65	4.1
7	Nitride	JMTR	2	9.4	73	5.5
8	Carbide	JOYO*	1	8.5	(80)	(4.5)
9	Nitride	JOYO*	2	8.5	(80)	(4.5)

* Under joint research program with PNC

Fig. 1 Irradiation program of mixed carbide and nitride fuels

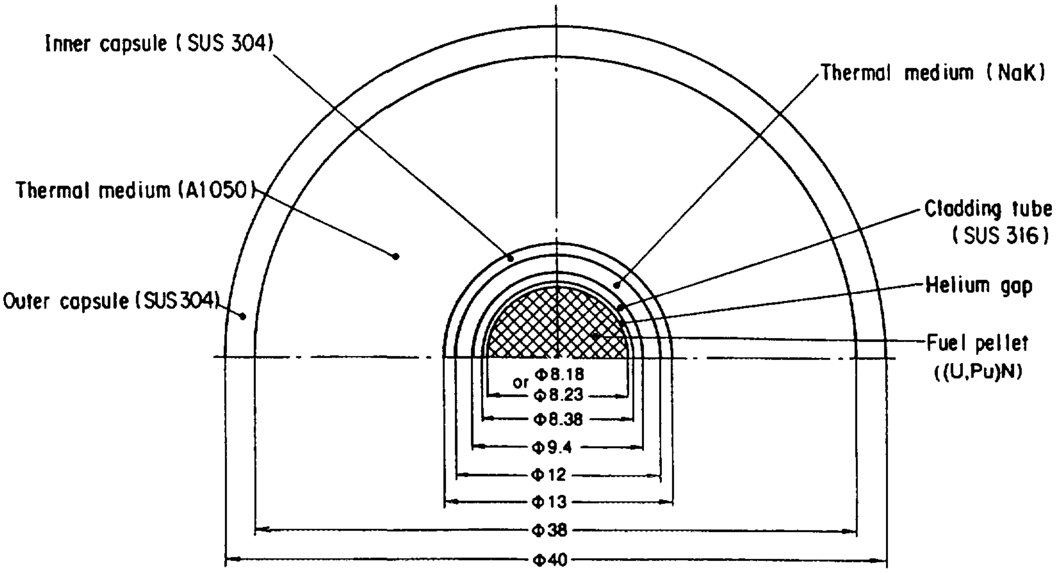


Fig.2 Conceptual design of capsules for irradiation of nitride fuel at JMTR

powder was added to carbide powder as a sintering aid before compacting in order to obtain dense matrix. The details of the fabrication process are described in earlier papers[4]. The microstructure of the conventional type- and thermal stable carbide pellets are shown in Photo. 1 to Photo.3.

Mixed nitride was also prepared by carbothermic reduction of the dioxides with graphite in flowing N_2-8%H_2 mixed gas at 1773K[5,6,11]. The mixing ratio of the dioxides and graphite, C/(U+Pu), was 2.50. The fabrication procedure of nitride pellets was similar to that of carbide fuel except the atmosphere condition for sintering. Pore former was added to nitride powder before compacting to make the thermal stable pellets. Since actinide nitrides originally show poor sintering characteristics, fine nitride powder ground by a tungsten-lining ball-milling was used for obtaining dense fuel matrix No sintering aid was used in the case of the nitride pellet fabrication. The microstructure of the nitride pellet sintered in flowing Ar-8%H_2 gas is shown in Photo. 4.

The fabrication of fuel pellets was carried out in gloveboxes with an atmosphere of argon gas purified to impurity levels of less than 2 ppm and 5 ppm for oxygen and water, respectively, in order to protect the fine powder of carbide and nitride against oxidation.

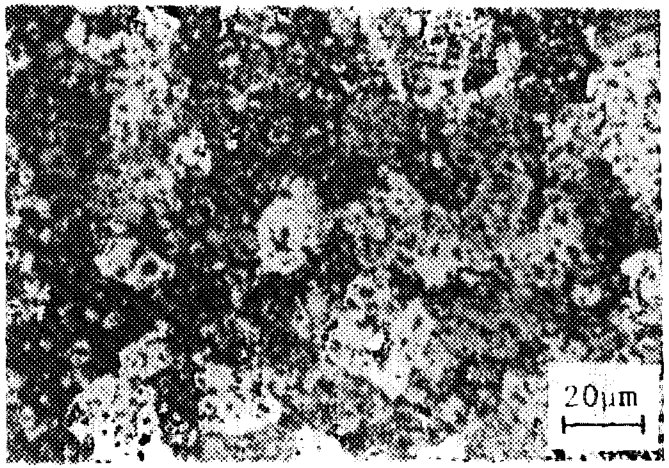


Photo. 1 Microstructure of nearly stoichiometric mixed carbide pellet

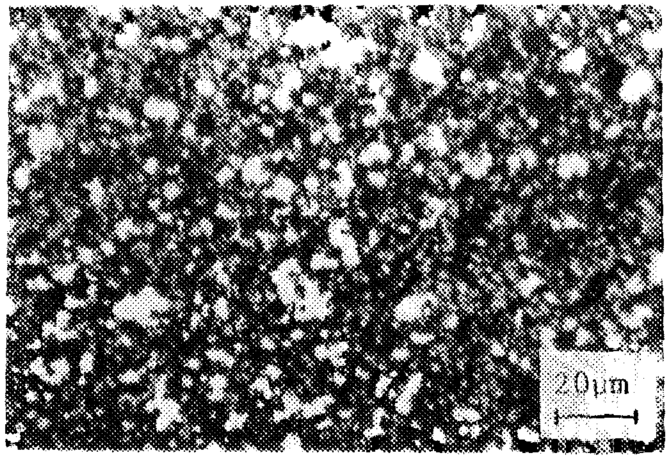


Photo.2 Microstructure of hyperstoichiometric mixed carbide pellet

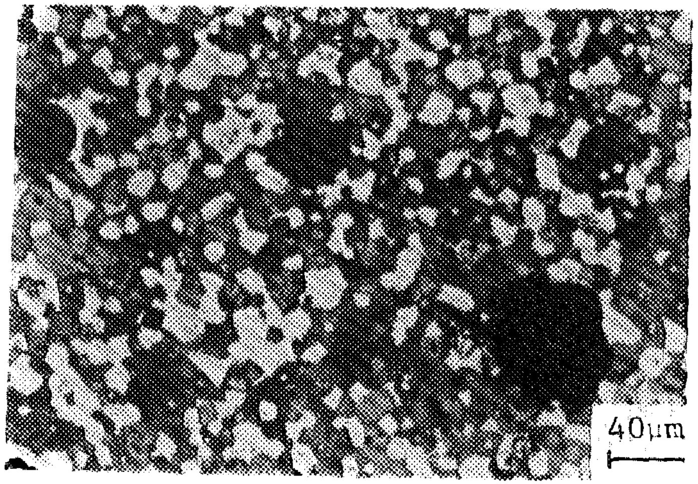


Photo.3 Microstructure of thermal stable carbide pellet

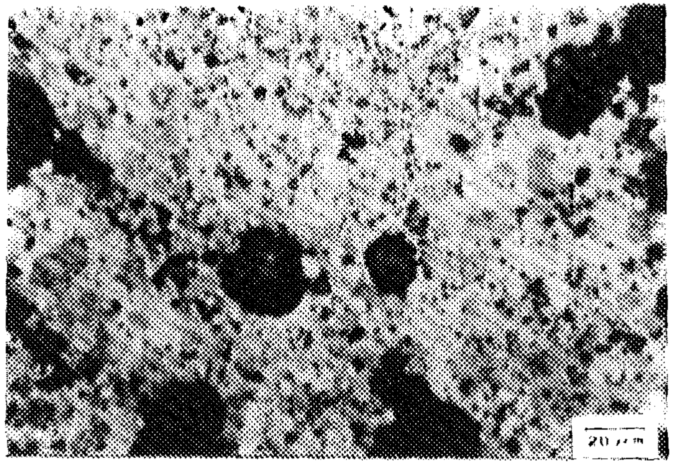


Photo.4 Microstructure of thermal stable nitride pellet

3.3 PIN PREPARATION AND ENCAPSULATING

After centerless-grinding and inspections, the pellets were inserted with thermal insulator pellets of uranium mononitride or uranium monocarbide and a plenum spring into the cladding tube of 20%C.W. SUS316 and an upper endplug was welded by TIG in a helium gas atmosphere. As an except, a cladding tube of 11Cr-2W ferritic stainless steel was used at the final irradiation of nitride fuel to investigate the effects of cladding materials on the fuel-clad mechanical interaction(FCMI). After the fabrication of fuel pins, one or two pins were encapsulated by Mechanical Engineering Division of JAERI.

The characteristics of the carbide and nitride fuel pins are shown in Tables 1 and Table 2.

4. Irradiation

The irradiation and post irradiation examinations(PIEs) were completed as scheduled without any trouble, although the nitride fuel of the last irradiation is still under examinations at present. The irradiation conditions are summarized in Table 3 and Table 4. The heating rate and burnup were estimated from the temperature of NaK and found to be lower than the scheduled one because of conservative capsule design. Therefore, actual heating rates were estimated to be 420 - 730 W/cm. The maximum burnup was estimated to be 4.7% FIMA for carbide and 5.5% FIMA for nitride.

5. Results of post irradiation examinations

5.1 CARBIDE FUEL

The items of the PIEs were shown in Fig.3. The destructive examinations of irradiated fuel pins were carried out at the cells with an argon gas atmosphere. The maximum burnup in the program was 4.7% FIMA and no pin failure was observed. The typical macrostructure of carbide pellets irradiated at JMTR are shown in Photo. 5 to Photo.7. The results of PIEs of the carbide fuel are briefly summarized as follows:

The structure of the outer region was kept as fabricated, but in the inner region porosity increased as shown in the photographs.

No significant migration of actinide and FP elements was observed except fission gas such as Kr and Xe, and Cs.

Maximum diametric increase of fuel pins was in the range of 0.3 - 0.5 % at 3 - 4.7% FIMA as shown in Fig. 4.

Slight carburization of cladding material was induced by hyperstoichiometric carbide and a slight reaction of cladding material with fuel material was observed in the case of nearly-stoichiometric carbide.

Table 1 Characterization of mixed carbide fuel irradiated at JRR-2 and JMTR

Capsule Name		ICF-37H		ICF-47H		84F-10A		84F-12A		87F-2A
Pin Number		37-1	37-2	47-1	47-2	10-1	10-2	12-1	12-2	-
Fuel										
Carbon content	(wt%)	4.70	5.21	4.66	5.09	4.67	5.16	4.67	5.19	5.09
Oxygen content	(wt%)	0.27	0.14	0.25	0.16	0.29	0.12	0.30	0.14	0.48
Ceq/(U+Pu)		1.00	1.09	1.00	1.10	1.02	1.10	1.02	1.10	1.14
Diameter (mm)		5.40	5.40	8.23	8.23	8.23	8.23	8.23	8.23	8.23
Density	(%TD)	89.7	93.3	83.1	83.7	84.6	81.5	83.0	82.2	86.7
Pu/(U+Pu)		0.201	0.201	0.194	0.194	0.194	0.197	0.202	0.198	0.196
Stack length	(mm)	96	96	60	60	100	100	100	100	180
Cladding Tube										
Outer diameter	(mm)	6.50	6.50	9.40	9.40	9.40	9.40	9.40	9.40	9.40
Thickness	(mm)	0.47	0.47	0.51	0.51	0.51	0.51	0.51	0.51	0.51
Pin										
Gap size (mm)	0.16	0.16	0.15'	0.15	0.15	0.15	0.15	0.15	0.15	
Smear density	(%TD)	84.6	88.0	80.2	80.7	81.6	78.6	80.1	79.3	83.6
Total length	(mm)	185	185	170	170	250	250	250	250	390

Ceq; Carbon equivalent
%TD; % of Theoretical Density

Table 2 Characterization of mixed nitride fuel irradiated at JMTR

Capsule name		88F-5A		89F-3A	
Pin Number		5-1	5-2	3-1	3-2
Fuel					
Nitrogen content	(wt%)	5.56	5.56	5.42	5.42
Oxygen content	(wt%)	0.12	0.12	0.14	0.14
Carbon content	(wt%)	0.20	0.20	0.20	0.20
Neq/(U+Pu)		1.04	1.04	1.03	1.03
Diameter (mm)		8.18	8.23	8.23	8.25
Density	(%TD)	83.1	83.0	85.8	86.1
Pu/(U+Pu)		0.196	0.196	0.209	0.209
Stack length	(mm)	100	100	100	100
Cladding					
Materials		20%C.W.316SS		←	Ferritic SS
Outer diameter	(mm)	9.40	9.40	9.40	9.40
Thickness	(mm)	0.51	0.51	0.51	0.50
Pin					
Gap size (mm)		0.20	0.15	0.15	0.15
Smear density	(%TD)	79.2	80.1	82.5	83.2
Total length	(mm)	250	250	250	250

Neq; Nitrogen equivalent

Items	Pins	Pellets	Cladding
Visual inspection of fuel pins	○		
Profilometry	○		
Gamma scanning	○		
Puncture & FP gas collection	○		
Density measurement		○	
Metallography		○	○
Pore distribution		○	
Grain size measurement		○	○
Micro-gamma scanning		○	
Auto-radiography		○	
Hardness			○
Electron probe microanalyses		○	○
X-ray diffraction analyses		○	

Fig.3 Items of post-irradiation examinations of carbide and nitride fuels at JAERI

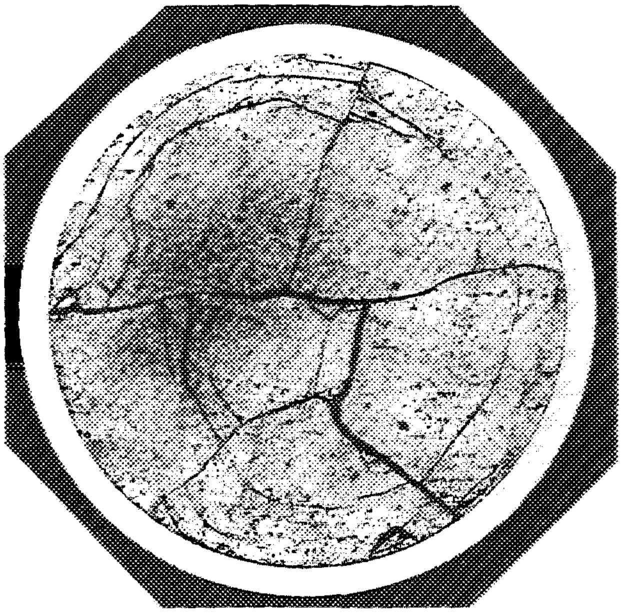


Photo.5 Macrostructure of nearly stoichiometric mixed carbide pellet irradiated up to 4.5%FIMA

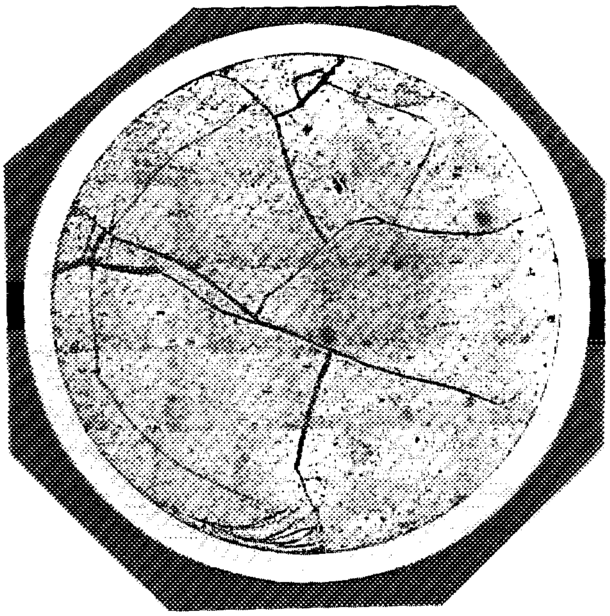


Photo.6 Macrostructure of hyperstoichiometric mixed carbide pellet irradiated up to 3.7%FIMA

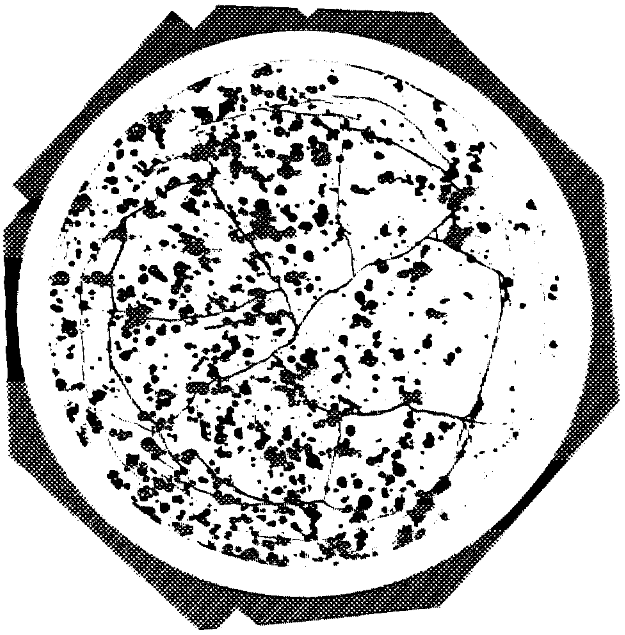


Photo.7 Macrostructure of thermal stable carbide pellet irradiated up to 4.7%FIMA

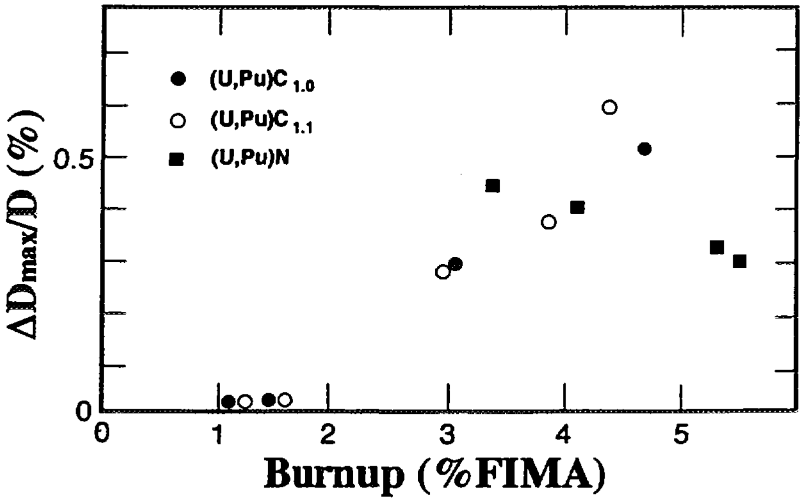


Fig.4 Maximum diameter increase of carbide and nitride fuels irradiated at JRR-2 and JMTR

Segregation of plutonium was measured in the fuel containing sesquecarbide.
Precipitation of Pd and Mo was observed.

The FP gas released from the pellets to plenum was collected by puncturing and its amount and composition were determined. The results are summarized in Table 5, where the first irradiation at JRR-2 was preliminary, so the pellets of high density of 90%TD and cladding tubes of 6.5 mm in diameter were used. It can be seen from the table that the carbide fuel shows the FGR rates of 2 - 3%/% FIMA except the irradiation of the first preliminary campaign and the last one. The latter irradiation test was carried out by using the thermal stable pellets and showed very low FGR; less than 1% at 4.7% FIMA.

The results on the FGR obtained by the irradiations in USA and European countries lie in the range of 10 - 30 % at 5% FIMA at the heat rates of 700 - 1200W/cm and smear densities of 75 - 80% TD[15]. Of course, FGR strongly depends on the fuel design and irradiation conditions, especially the fuel density and heating rate. The tests in USA and European countries were basically performed around the heating rates of 800W/cm or more. Considering the difference in heating rates, the present results obtained by us are considered to be in fairly good agreement with those reported previously except the case of the thermal stable pellets.

5.2 NITRIDE FUEL

Four pins of nitride fuel were irradiated at JMTR only by using the thermal stable pellets. The typical macrostructure after irradiation is shown in Photo.8, but it seems to be quite strange. It is presumed that such structure might be caused from an incomplete treatment of pore former at fuel fabrication although the structure before irradiation, which is shown in Photo. 4, was not so strange.

Table 3 Irradiation conditions of mixed carbide fuel

Capsule Name		ICF-37H	ICF-47H	84F-10A	84F-12A		87F-2A
Pin Number					12-1	12-2	
Reactor		JRR-2	JRR-2	JMTR	JMTR		JMTR
No. of cycle		6	10	12	17		17
Irradiation time	(hr)	1,543	2,655	6,143	8,748		8,924
Linear heat rate	(W/cm)	420	640	590	600	490	640
Central Temperature	(K)	1,270	1,420	1,490	1,530	1,370	1,510
Burnup	(GWd/t)	10	13	26	39	32	40
	(%FIMA)	1.2	1.5	3.0	4.5	3.7	4.7

Table 4 Irradiation conditions of mixed nitride fuel

Capsule name		88F-5A		89F-3A	
pin number		5-1	5-2	3-1	3-2
Reactor		JMTR		JMTR	
No. of Cycle		15		19	
Time	(hr)	7,626		9,972	
Linear heat rate	(W/cm)	650	535	700	730
Central temperature	(K)	1,770	1,470	1,710	1,740
Burnup	(GWd/t)	35	29	45	47
	(%FIMA)	4.1	3.4	5.3	5.5

Table 5 FGR data of mixed carbide fuel irradiated at JRR-2 and JMTR

Capsule name		ICF-37H		ICF-47H		84F-10A		84F-12A		87F-2A
Pin Number		37-1	37-2	47-1	47-2	10-1	10-2	12-1	12-2	
FGR	(%)	0.4	0.1	9.4	9.4	6.1	13.7	7.1	15.5	0.6
Porosity										
Before irradiation	(%)									
After irradiation	(%)									
Open porosity		1.3	0.5	11.5	11.0	9.7	12.3	9.7	11.8	2.2
Closed porosity		11.3	8.6	7.8	6.7	9.0	8.4	11.4	8.5	15.1
Grain size										
Before irradiation	(μm)	-	-	6.3	2.8	4.0	2.7	-	-	-
After irradiation	(μm)									
Outer region		10.6	7.9	2.9	2.8	3.7	3.0	2.5	2.8	10.6
Intermediate Region		10.5	7.5	3.8	3.9	4.7	3.1	2.6	3.2	10.6
Inner Region		14.3	7.3	6.3	4.9	5.3	3.2	7.0	3.3	10.9

At present, the results of PIEs of nitride fuel are under analysis, but the features of fuel behavior observed in the present irradiation tests are briefly summarized as follows:

Precipitation of Pd was observed.
Actinide and FP elements did not migrate significantly.
No significant reaction of fuel material with the cladding tubes was observed.
Maximum diameter increase of fuel pins was smaller than those of carbide fuel (see Fig.2).

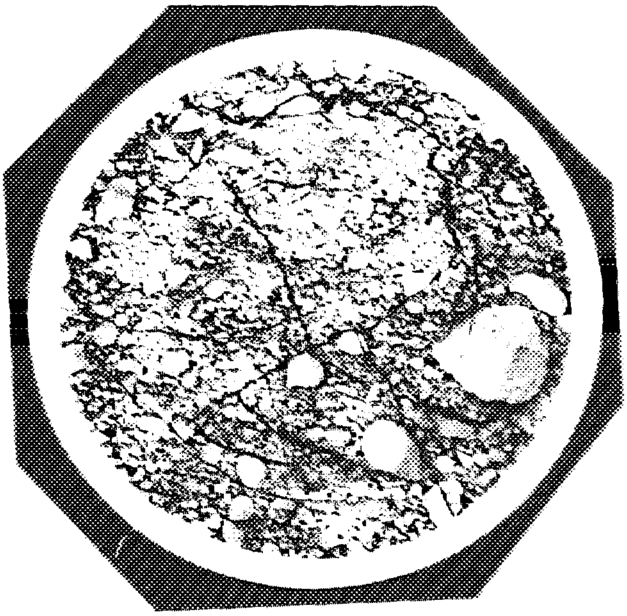


Photo.8 Macrostructure of thermal stable nitride pellet irradiated up to 5.5%FIMA

The results of the FGR from the mixed nitride fuel irradiated at JMTR are shown in Table 6 and Fig 5 summarizes the results of the FGR from nitride and carbide fuels obtained from the present irradiation program. The factors controlling the FGR from nitride fuel could not be determined in this campaign, since only the thermal stable pellets were irradiated and their structure seemed to be strange. In spite of this negative condition, the nitride fuel was confirmed to show very low FGR of a few percent at about 5%FIMA. Further depression of the FGR of nitride fuel might be expected by proper handling of pore former, as the thermal stable carbide pellets showed the FGR of less than 1%.

This low FGR of nitride fuel observed in the present studies can be explained by both of the low fuel temperature and the stability of the structure. Although the structure shown in Photo.8 seemed strange, the open porosity was much smaller than those of carbide pellets fabricated without pore former. Therefore, it is suggested that the nitride pellets still kept the features of the thermal stable pellets. As pointed out by Blank[3], fuel temperature of carbide and nitride pellets decreases with irradiation up to the burnups of 2 - 3 % FIMA because of the increase of the gap conductance and keeps a constant value after the closing of the gap. Figure 6 shows the change of the central temperature of nitride fuel during the irradiation at JMTR, where the temperature was measured by a set of thermocouple inserted into the central holes of fuel pellets. It is understood from the figure that the gap seemed to close around 2 - 3 % FIMA and fuel temperature continued to decrease till the burnup.

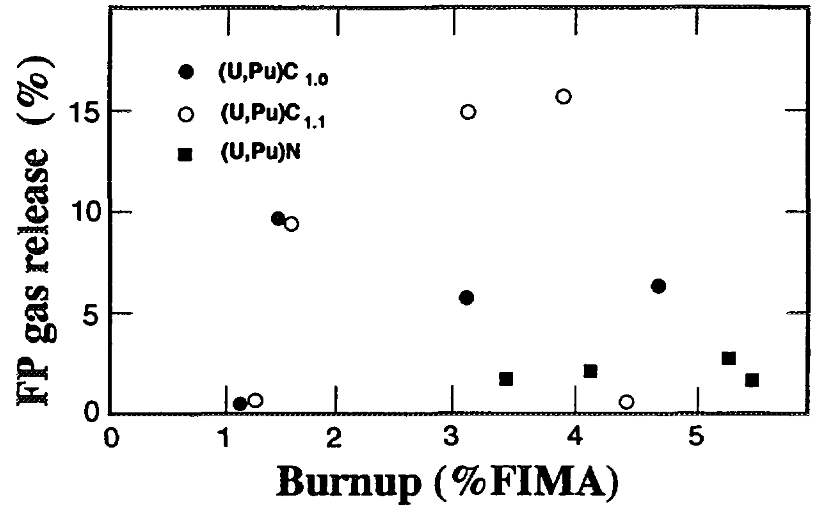


Fig.5 Fission gas release from carbide and nitride fuels irradiated at JRR-2 and JMTR

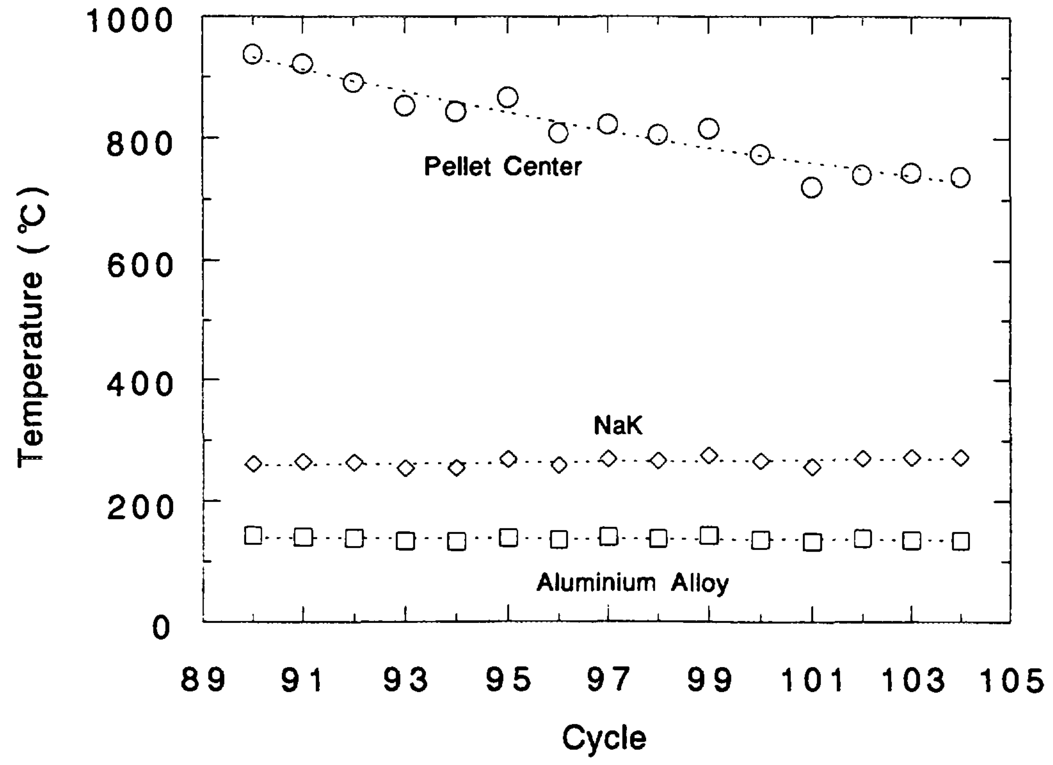


Fig.6 Temperature change at fuel center and thermal media under irradiation of nitride fuel at JMTR

6. Discussion

6.1 THERMAL STABLE PELLETS

The thermal stable carbide pellets irradiated at JMTR showed very low FGR of less than 1% at 4.7%FIMA as expected. The reason why the pellets showed very low FGR might be explained by the stability of the fuel matrix. It is also found that the FGR from nitride pellets was very low, although the fuel pellets showed a strange structure perhaps because of the improper addition of pore former. Further improvements of fabrication technology of the thermal stable nitride pellets might make the FGR negligibly small. The values of the FGR from the thermal stable pellets of carbide and nitride are much lower than those of carbide pellets fabricated by a conventional route without pore former and also the results of irradiation tests in USA and European countries. It is considered that the lowering of the FGR has been realized by the introduction of the thermal stable pellets as well as the adoption of the cold fuel concept.

6.2 EFFECTS OF SECOND PHASE AND POROSITY

Comparing the FGR of the carbide fuel irradiated in the present program, the pellets containing sesequicarbide as a second phase had a tendency to show larger FGR. The presence of sesquicarbide might disturb the grain growth of the monocarbide during sintering at the fabrication process. It would be common that the small grain sizes have a tendency to enlarge the FGR because of shorter distance for diffusion of fission gases to the boundaries. The reason why hyperstoichiometric carbide fuel showed large FGR might be explained by the presence of sesquicarbide. However, the difference in the grain size between both carbide fuel pellets was not so large. On the other hand, the thermal stable pellets of carbide showed very low FGR as shown in Table 5. These pellets had a hyperstoichiometric composition but large grains because of use of a sintering aid. Therefore, it is too easy to explain the difference in FGR only on the basis of the presence of the second phase.

The porosity after irradiation were determined in the present study as shown in Table 5 and Table 6. It is easily understood that the thermal stable pellets have very low open porosity. The pores introduced by the pore former seem to stay close for the pellet surface and the fission gas was considered to be retained in the fuel matrix. In the case of carbide pellets fabricated without pore former, hypersotichimetric carbide pellets had larger open porosity. It is suggested from the comparison of the porosity that the FGR depends on the open porosity rather than the presence of a second phase or grain size. The results of the effect of open porosity on the FGR are in fairly a good agreement with those reported on the nitride fuel, which shows rapid increase of FGR when the open porosity exceeds 10%[16]. Of course, there is some possibility that the formation of open porosity might be caused from the pressure of sesquicarbide. The controlling of the open porosity should be a key technology to lower the FGR.

Table 6 FGR data of mixed nitride fuel irradiated at JMTR

Capsule name		88F-5A		89F-3A	
Pin Number		5-1	5-2	3-1	3-2
FGR	(%)	2.9	1.8	2.8	1.7
Porosity					
Before irradiation	(%)				
After irradiation	(%)				
Open porosity		8.8	8.7	5.1	4.3
Closed porosity		11.2	10.2	13.3	12.7
Grain size					
Before irradiation	(μm)				
After irradiation	(μm)				
Outer region		4.0	3.7	+	+
Intermediate Region		3.9	3.6	+	+
Inner Region		3.6	4.2	+	+

+: under analysis

6.3 FUEL CLAD MECHANICAL INTERACTION

In the design of MOX fuel, almost all of FP gas generated would be released in the gas plenum, especially at high burnups, although the outer region still keep some fission gas in fuel matrix. In addition the creep rate of MOX fuel is relatively large so that the life of the fuel might be controlled by either the increase of inner pressure and/or the swelling of cladding materials. On the other hand, the FGR from the carbide and nitride fuels are confirmed to be lowered by adoption of the cold fuel concept. Especially FGR of the thermal stable pellets was found to be negligibly small at least up to 5.5%FIMA. It is well known that FP gas retained in the fuel matrix has a tendency to enlarge the fuel swelling. Since nitride and carbide have relatively low creep rates, the FCMI would become important for fuel design.

In the present study, the maximum increase rates of fuel diameter were 0.2 - 0.3%/% FIMA or less after the closing of fuel-cladding gap, as shown in Fig. 4. The values are equivalent to or lower than those reported from the irradiation of carbide and nitride, D/D of 0.3-0.8% at 5% FIMA[17] and 0.48%/% FIMA[18], respectively. Therefore, at least up to the burnup of 5.5% FIMA, the FCMI would be not essential in spite of the depression of FGR. At higher burnups, especially over 10%FIMA, there is some possibility that the FCMI caused from the swelling due to retained gases and solid FPs would become significant. It cannot be concluded that the large pores in the thermal stable pellets can take a role for accommodation of gas swelling. Furthermore, the temperature increase at transition conditions might enlarge fuel swelling. Therefore, it is necessary that the data of irradiation should be collected from the viewpoint of FCMI.

7. Conclusions

The irradiation tests of nitride and carbide fuels at JRR-2 and JMTR have been performed at JAERI to investigate the fuel behavior of these advanced fuels. From the tests we can conclude on the FGR as follows:

(1) The carbide pellets fabricated by a conventional route showed the FGR rates of 2 -3%/% FIMA, which is considered to be equivalent to those in literature.

(2)The thermal stable pellets of carbide and nitride fuels, which were fabricated by use of pore former, showed very low fission gas release of less than 3 % at 4 - 5.5% FIMA.

(3) It is suggested that the fission gas release of carbide and nitride fuels might be controlled by open porosity.

(4) The acceleration of fuel-cladding mechanical interaction by the depression of fission gas release was not essential at least up to 5.5% FIMA.

In addition to the irradiation tests in JRR-2 and JMTR, the thermal stable pellets of carbide and nitride fuels are being irradiated to goal burnups of about 4.5%FIMA at the fast experimental reactor JOYO under collaboration with PNC. The analyses of the FGR and the other behavior of these fuels are programmed by combining the results from the irradiation at JOYO with those at JRR-2 and JMTR.

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