PNC TN1102 97-015

公開資料

Research and Development on MOX Fuel for LMFBR

(高速炉用MOX燃料の研究開発)

平成9年12月

動力炉・核燃料開発事業団

公開資料
PNC TN1102 97-015
1997年12月

Research and Development on MOX Fuel for LMFBR

（高速炉用MOX燃料の研究開発）

中 江 延 男*

要 旨

高速炉用MOX燃料の高燃焼度化を目指して、動燃が実施している研究開発の概要を紹介する。

研究開発項目としては、燃料製造技術開発、物性測定、照射試験（安全性試験を含む）、解析コード開発及び炉心材料開発を取り上げた。

また、国際協力についても紹介した。

さらに、高燃焼度化の見通しについて紹介した。

＊：本社　核燃料サイクル技術開発部

Research and Development on MOX Fuel for LMFBR

N. Nakae

Nuclear Fuel Cycle Development Division
Power Reactor and Nuclear Fuel Development Corporation

1-9-13, Akasaka, Minato-ku, Tokyo, 107, Japan

Abstract

This paper deals with the history and current status of the research and development on MOX fuel for LMFBR which has been performed in Power Reactor and Nuclear Fuel Development Corporation (PNC).

The PNC continues to make R&D on MOX fuel in the field of fuel fabrication, property measurement, irradiation test including safety test, code development and core material development for these thirty years. A lot of variable and fruitful results and good achievements are obtained, and they have been reflected in the design and operation of the experimental fast reactor "Joyo" and the prototype fast breeder reactor "Monju".

The results and achievements obtained in the past 30 years and to be obtained in the near future shall indicate that the LMFBR can give us the safety and economical energy and/or electricity and, of course, the fuel used in the LMFBR shall have a very high burnup of more than 150 GWd/t in fuel assembly averaged.

1. Introduction

Though many Light Water Reactors (LWRs) are successfully operated in the world, Fast Breeder Reactors (FBRs) or Fast Reactors (FRs) have been shut down in the leading countries such as United State, United Kingdom and Germany due to the particular reasons. The typical reason is considered to be its poor economy. Therefore, much effort has been done to improve the economy of FBR and its related fuel cycle. The achievement of high fuel burnup is one of the very important key issues, and Mixed-Oxide Fuel (MOX) is the most promising candidate for the Liquid Metal Fast Breeder Reactor (LMFBR) in view points of the technology and the experience accumulated on it, also the availability of existing industrial facilities. Thus major part of R&D on MOX has been focused on a high performance especially in burnup [1,2,3].

The activities and results, which PNC is performing and obtains, are introduced and the possibility realizing the high fuel burnup is also mentioned in this paper.

2. R&D activities and results

The target of the R&D on high burnup fuel is to achieve the assembly averaged fuel burnup of more than 150 GWd/t and it corresponds to the pellet peak burnup of about 250GWd/t and the peak total neutron fluence of about 5x10E23 n/cm2 (250dpa). The fuel cycle cost is strongly depends on the fuel burnup because the cycle cost is related by the following equation;

cycle cost (\$/kwh) = (fabrication + reprocessing) cost (\$/kg)/BU (kwh/kg)

The relation between fuel cycle cost and fuel burnup is shown in Fig.1. The influence of burnup on cycle cost tends to saturate at very high burnup of more than 150 GWd/t and it is explained by the escalation of reprocessing cost of high burnup spent fuel.

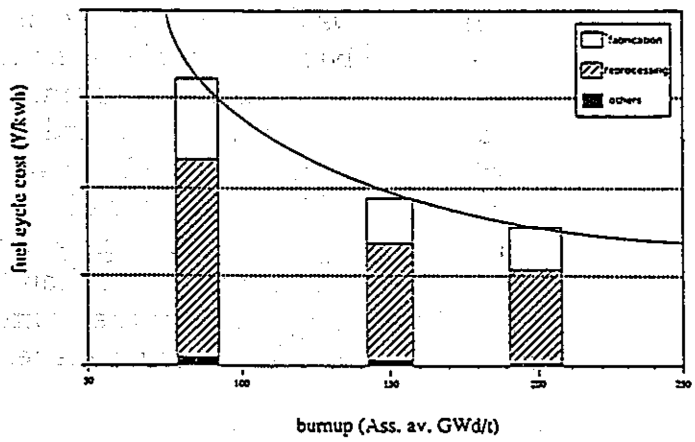


Fig. 1 Relation between burnup and fuel cycle cost

The activities and corresponding results on the R&D such as fuel fabrication, property measurement, irradiation test, core material development and so on are shown in this section.

2.1 Fuel Fabrication Technology Development

The fuel smear density is one of the key parameters in order to obtain high burnup. The smear density is to be determined very carefully by considering the avoidance of fuel cladding mechanical interaction (FCMI) and the improvement of core characteristic. The two phenomena make an inversive influence each another on the smear density. The smear density of 80 - 85 % is selected with the typical MOX fuel for LMFBR in the world and the values are smaller than those of the uranium oxide fuel for LWR. The Monju fuel pellet has a theoretical density of 85 % and the corresponding smear density is 80 %. It seems to be low because of taking much conservative for FCMI failure. It is very difficult to control density of the low density pellet. Much effort is focused on the selection of pore former materials and the optimization of the sintering procedure and so on. The good achievement has been obtained with Monju fuel pellet and the total of about 300 fuel assemblies have been fabricated [4].

Another method obtaining the low smear density is to adopt annular pellet and the annular pellet also makes it possible to increase linear heat rating. The development of the annular pellet is being done for the future adaptation with Monju. The key issues are the developments of the compressing and forming equipments and the inspection device.

2.2 Property Measurement

Many kinds of thermal and physical properties are measured and reviewed in order to use the fuel design. The private handbook of fuel property has been prepared and it is revised when it is needed. The fuel thermal conductivity and melting temperature are now under examination, and the results will be published in the journals [5,6].

The results of melting temperature measurement with the burnup fuels and of thermal conductivity measurement with the compound of Cesium Molybdate (Cs_2MoO_4) are shown here. Fig.2 shows the results of the melting temperature measurement together with those of the burnup simulated samples [5,7]. The trend of the burnup dependence of melting temperature is similar to each another with the samples of in-reactor irradiated and burnup simulated. No remarkable decrease of melting temperature is observed. Fig.3 shows the result of the thermal conductivity measurement with Cs_2MoO_4 which corresponds to the compound formed in the fuel cladding gap of the highly irradiated fuel [8,9]. The thermal conductivity of the compound is much higher than that of helium gas. This means that the gap conductance with high burnup fuel is improved due to the formation of the compound and that no remarkable centerline temperature increase is expected.

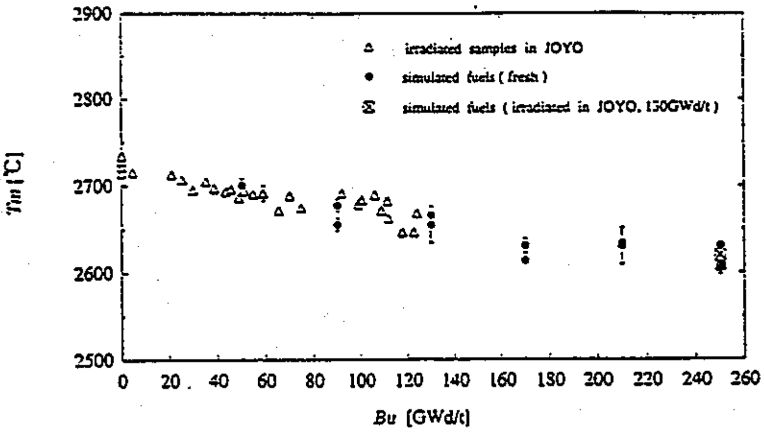


Fig.2 Burnup dependence of the melting temperature together with the result of that of the burnup simulated samples

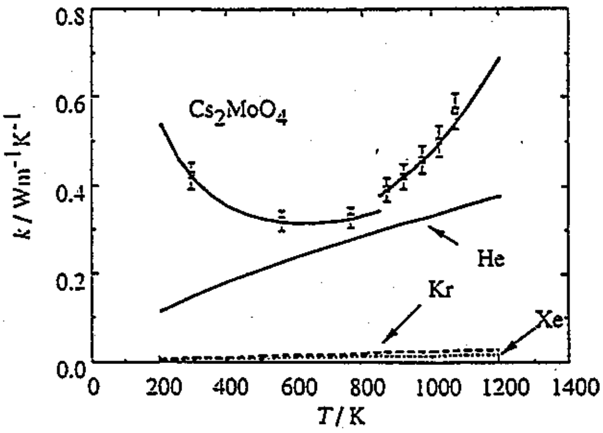


Fig.3 Thermal conductivity of Cs2MoO4

2.3 Irradiation Test

The irradiation test is the most important procedure to examine the fuel integrity during irradiation directly. Therefore, many irradiation tests have been conducted using both foreign and domestic reactors such as DFR in UK, Rapsodie and Phenix in France, EBR-II and FFTF in USA, Joyo in Japan and so on. The irradiation tests are composed with single pin irradiation test, small and large pin bundle tests, power to melt test(PTM), operational transient test (OTT) and run to or beyond cladding breach test (RTCB and RBCB).
Fig.4 shows post irradiation examination (PIE) result of fission gas release [10]. The different behavior of fission gas release is observed with the restructured and unrestructured region of fuel pellet. The rapid and large amount of fission gas release is observed in the restructured region. On the other hand, the slow and small amount of fission gas release is occurred in the outer region of fuel pellet where no restructuring is observed and the amount of fission gas release is increasing with burnup extension.

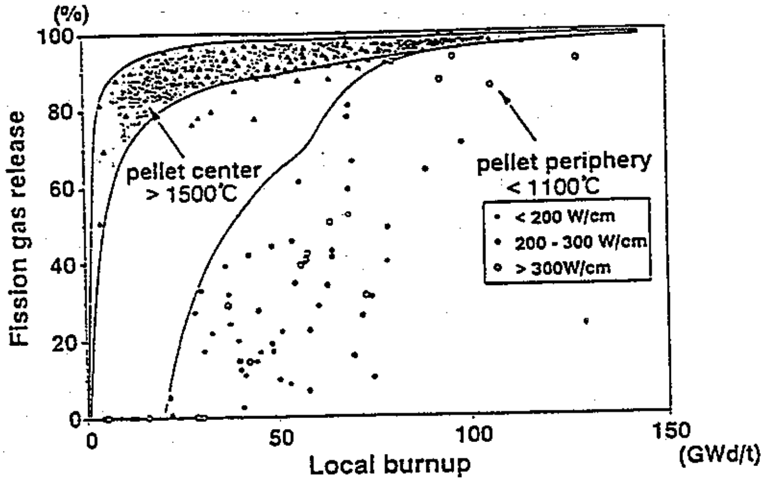


Fig.4 Burnup dependence of fission gas release in both regions of pellet center and pellet periphery

Fig.5 shows the PIE result of fuel cladding chemical interaction (FCCI) [11]. The FCCI is much depended on the fuel burnup and metal to oxygen ratio (O/M). The large FCCI is observed with the fuel pin having high burnup and high O/M and it is reasonably accepted by the fact that the excess oxygen plays an important role on the cladding corrosion.

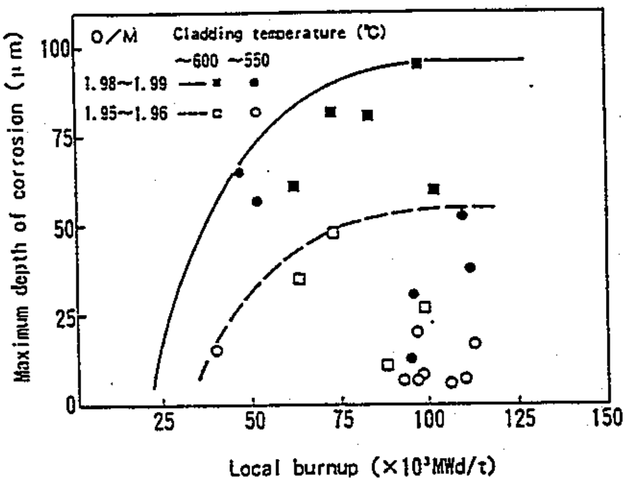


Fig.5 Burnup dependence of FCCI as parameters of O/M ratio and cladding temperature

The operational reliability test (ORT) has been performed using the EBR-II reactor for 15 years from 1981 to 1995 under the collaboration between US-DOE and PNC. The test contains two kinds of the safety irradiation tests such as OTT and RBCB. The safety margin is demonstrated by the result obtained with the ORT [12,13,14,15,16]. Fig.6 shows the result of the OTT and it indicates that the fuel failure during transient over power is observed with the fuel pin having the transient over power of more than 1.5 times of the nominal power. The usual reactor trip level in the case of transient over power is set to be around 120%.

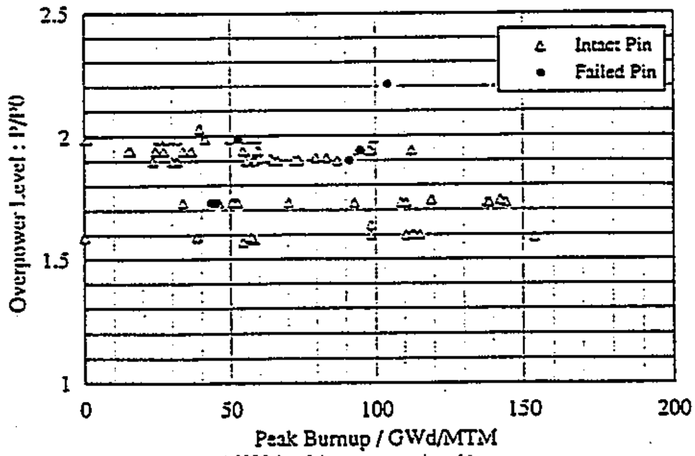


Fig.6 Results of overpower transient tests in ORT program

Fig.7 shows the result of the RBCB. This indicates the additional period of reactor operation after the fuel breach occurrence with the positions of fuel column and plenum, respectively. The very good stability is observed with the breached fuel pin during the run beyond cladding breach and it is due to the fact that steady compound of fuel meat (MOX) and sodium coolant (Na) is formed at the breached position which corresponds the interface of fuel and coolant. No additional formation of the compound is found and the expansion of the breached area is not observed. This means that no rapid reactor shut down and removing (unloading) of breached fuel pin are needed when the fuel failure signal is detected. The effectiveness of cover gas (CG) method and delayed neutron (DN) method, which are methods to detect the fuel failure, are also demonstrated through the RBCB.

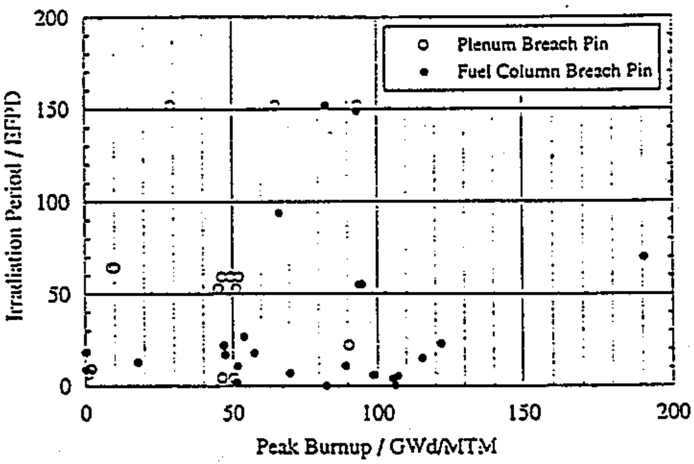


Fig.7 Results of RBCB experiments in ORT program

2.4 Code Development

Many kinds of computer codes have been developed and used in the design calculations. The codes are improved by the verification using the results of in-reactor test and PIE.

Among them the CEDAR code has been developed in order to analyze the fuel pin behavior during steady state and transient operations [17]. The code can predict the thermal and mechanical behavior of fuel pin as a function of burnup or irradiation time. The typical samples of the code calculation are shown in Figs.8 and 9. Fig.8 shows the comparison of the fuel restructuring between the predictions and the PIE. A very good prediction can be obtained by the CEDAR code. Fig.9 shows the comparison between the calculated and measured cladding strain with the steady state and transient tests. The CEDAR code can predict the cladding deformation very well.

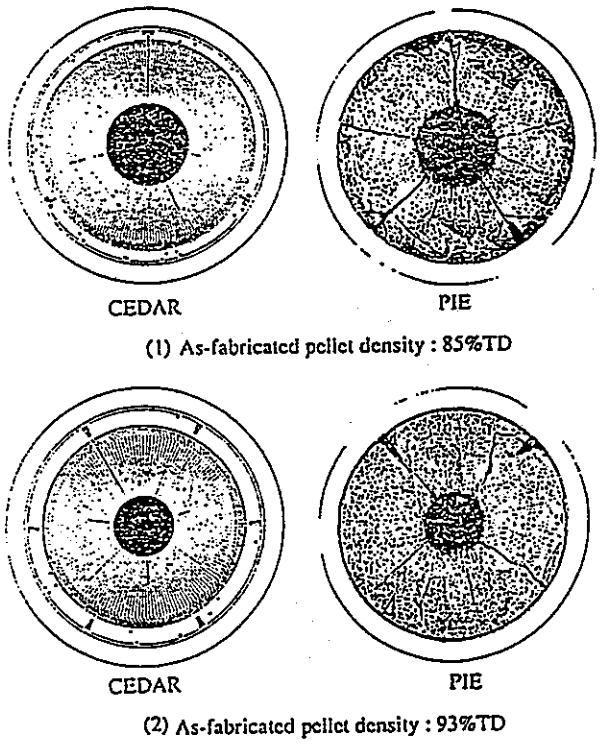


Fig.8 Code prediction of the fuel restructuring by comparison with that of PIE

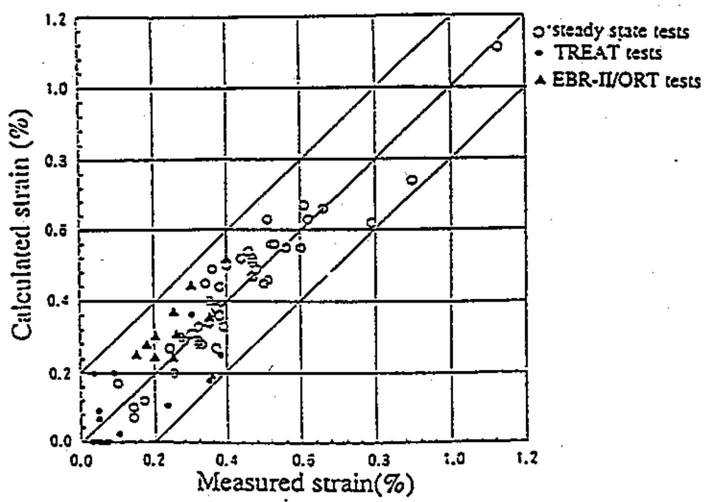


Fig.9 Code prediction of the cladding diametral strain by comparison with that of PIE

The BAMBOO code has been developed in order to analyze the bundle duct interaction (BDI), and the code can predict the clearances between pin and pin, and pin and duct. The code can also predict the bending mode of fuel pin and the stress appeared on the fuel pin. The typical sample of the code prediction is shown in Fig.10. The hard BDI causes the loss of coolant flow in the fuel assembly and induces the cladding temperature increase. This phenomenon may have a possibility to accelerate the fuel pin breach at high burn up. The code is now being developed using the data of out of pile and in pile experiments [18].

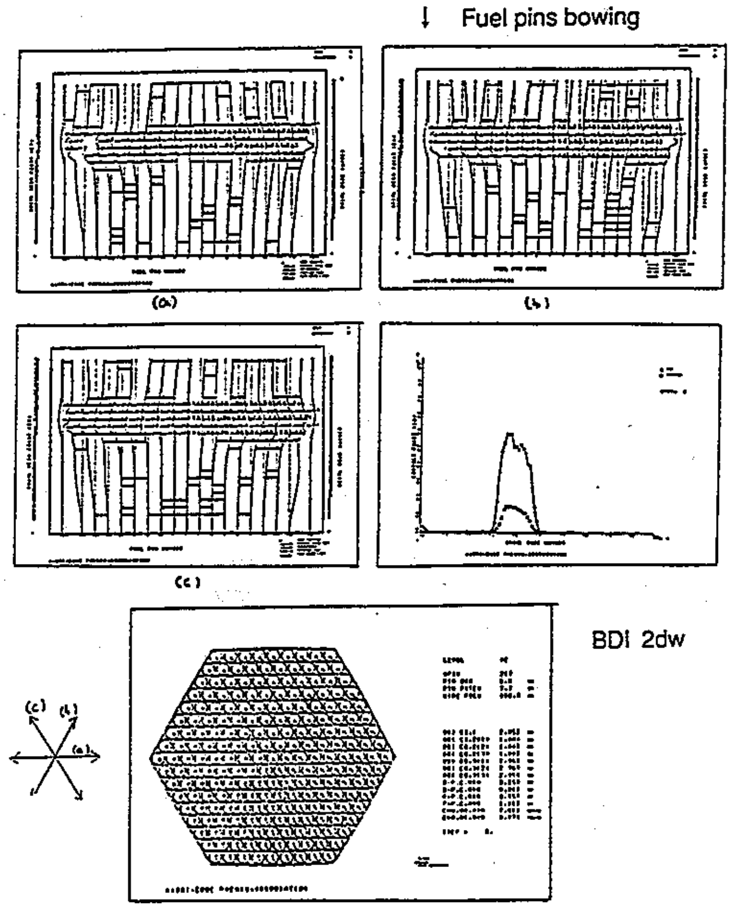


Fig.10 An example of the result of BAMBOO code

The other codes of MINERVA, SAFFRON and CORAL are also being developed. The MINERVA code can predict Cs migration behavior, the SAFFRON code can predict breached fuel pin behavior, and the CORAL code can predict absorber pin behavior, respectively.

2.5 Core Material Development

The core material development is one of the most important key issues in order to achieve fuel burnup as high as possible. Much effort has been done in the countries where the LMFBR has been developed such as USA, UK, France, Germany, Russia and Japan. Major kinds of the materials are focused on the austenitic and ferritic stainless steels. A very good performance of swelling resistance and irradiation creep strength is obtained with the selected materials such as 15-15 Ti (French material), HT-9 (USA material) and PE-16 (UK material).

In Japan, PNC316 of austenitic stainless steel was developed for Monju fuel and advanced austenitic stainless steel such as PNC 1520 and 14Cr-25Ni are now being developed for the future Monju and Japanese demonstration FBR. The special material of oxide dispersion strengthen material (ODS) is also developed for accomplishment of the target neutron damage of more than 250 dpa [19]. Fig.11 summarizes the history of the core material development in Japan. Fig.12 shows the position of the developed materials on the map of creep strength and neutron fluence which corresponds to swelling resistance. Fig.13 shows the austenitic stainless steel development focusing on the swelling resistance improvement. As shown here the amount of the basic components of Cr and Ni are controlled and the effective additions of minor elements are tried in order to improve the swelling characteristic. The current achievement for the swelling resistance is obtained with the PNC316 up to 115 dpa and with PNC1520 up to 120 dpa in the irradiation test using FFTF.

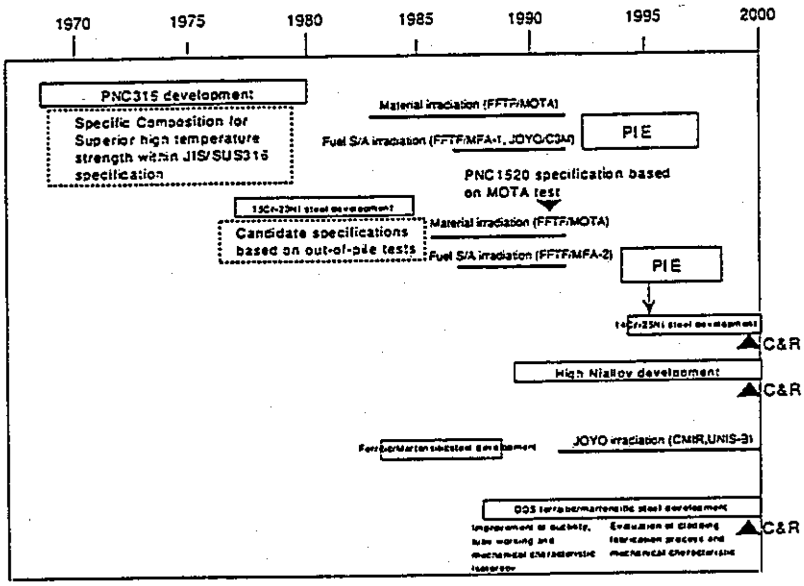


Fig.11 The history of the core material development in Japan

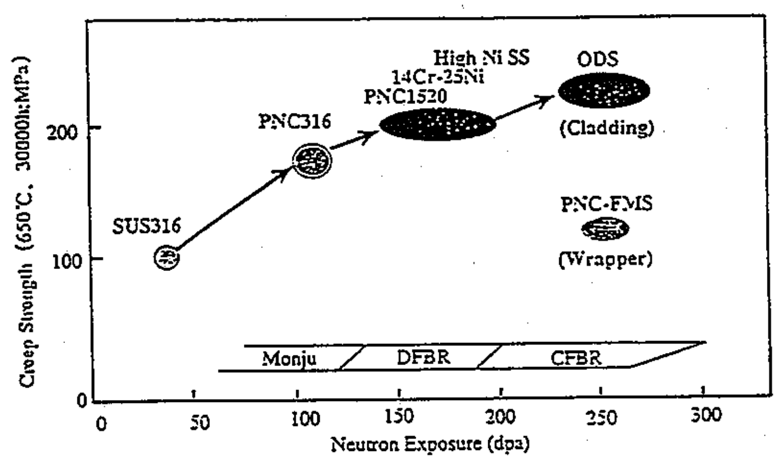


Fig.12 Target of the creep strength and swelling resistance with the developed materials in Japan

3. International Collaboration

The international collaboration in the field of LMFBR development is very important because a vast amount of R&D is necessary and it requires much money and many years. Thus, the international collaboration has been continued for these 30 years and many good results have been obtained especially in the field of nuclear fuel development.

Many fuel irradiation tests have been conducted using GETR in USA, DFR in UK, Rapsodie in France with a small scale of fuel assembly, and using FFTF in USA and phenix in France with a large scale of fuel assembly. The fuel pin analysis code development has also been performed under the good collaboration with US-DOE and the fuel reliability test using EBR-II was done by obtaining good collaboration of US-DOE and ANL. The information exchange on fuels and materials has been continued mainly with European countries.

The contract of the consultion on annular pellet behavior has been continued with BNFL, UKAEA and AEA Technology, and many irradiation data on annular fuel are transferred to PNC. These data are now under analysis and the result will be utilized in the safety licensing of the future Monju upgraded core. The PIE on the fuel, which has been irradiated in the PFR with the burnup of more than 20 % , has been conducted at Cadarache under the collaboration between CEA, BNFL/UKAEA and PNC. The result obtained here will be presented in the ICONE-6 Conference held in USA in 1998 [20].

The vibro and/or sphere packing fuel is expected to be used in the advanced nuclear fuel cycle which will be adopted in the future nuclear energy system. The collaboration on sphere packing fuel has been done with PSI in Switzerland. The collaboration in the field of the vibro packing fuel irradiation data evaluation and irradiation test will be planed with Russia under the ISTC and the international collaboration in the field of annihilation of weapon plutonium.

4. Conclusion

The massive R&D on the MOX fuel for LMFBR have been performed for these 30 years and many fruitful and valuable results are obtained. However, the following problems are still remained to be solved;

(1) Swelling characteristic of cladding materials at very high fluence of more than 3x10E23 n/cm2 (corresponding to 150 dpa)
(2) FCCI behavior at high burnup
(3) Thermal behavior under steady and transient (off normal) conditions
(4) Irradiation behavior of breached fuel under the run beyond cladding breach

However some problems are still remained to be solved, the followings are concluded based on the results obtained in these 30 years:

(1) The R&D activities including irradiation test on MOX fuel for LMFBR have

been successfully performed in order to achieve the fuel burnup and linear heat rating as high as possible.

(2) The good irradiation performance has been obtained on MOX fuel forLMFBR at high burnup including transient over power and run beyond cladding breach conditions.

(3) The performance analytical codes developed here can predict the fuel irradiation behavior well.

References

[1] S. Shikakura, T. Asaga, K. Hirai, S.Nomura, I. Shibahara, S. Iwanaga and M. Katsuragawa, Int. Conf. on Fast Reactor and Related Fuel Cycles, Kyoto, p6.3-1, (1991).

[2] M. Katsuragawa, H. Kashihara and M. Akebi, J. Nucl. Mat. 204(1993) p14.

[3] M. Katsuragawa, S. Shikakura, S. Nomura, T. Asaga, S. Ukai and H.Kaneko, Proceedings LMR : A Decade of LMR Progress and Promise, p204, Washington, D. C. November 11-15, 1990.

[4] Y. Aoki, M. Kashimura and T. Yamaguchi, Int. Conf. on Future Nuclear System, Proceedings, vol.1, p500 Yokohama, Japan, October 5-10, 1997.

[5] K. Yamamoto, T. Hirosawa, K. Yoshikawa, K. Morozumi and S. Nomura, J. Nucl. Mat. 204 (1993) p85.

[6] M. Inoue, to be published in J. Nucl. Mat..

[7] K. Konno and T. Hirosawa, to be published in J. of Nucl. Sci. and Tech..

[8] T. Ishii and T. Mizuno, J, Nucl. Mat. 231(1996) p242.

[9] T. Ishii and T. Mizuno, J. Nucl. Mat. 247(1997) p82.

[10] K. Maeda, T. Mitsugi and T. Asaga, Int. Workshop on Interfacial Effects in Quantum Engineering Systems (IEQES-96), Mito, Japan August 21-23, 1996.

[11] H. Kashihara, S. Shikakura, Y. Yokouchi, I. Shibahara, H. Matsushima and K. Yamamoto, Pro. BNES Conf. on Fast Reactor Core and Fuel Structural Behavior, Paper 43 (1990) p234.

[12] J. D. B. Lambert, J. H. Bottcher, K. C. Gross, R. V. Strain, J. I. Sackett, R. P. Colbum, S. Ukai, S. Nomura, T. Odo, S. Shikakura and M. Katsuragawa, Proceedings LMR : A Decade of LMR Progress and Promise, p233, Washington, D. C. November 11-15, 1990.

[13] A. Boltax, H. D. Garkisch, K. Hirai and T. Mitsugi, J. Nucl. Mat. 204(1993) p148.

[14] R. V. Strain, J. H. Bottcher, S. Ukai and Y. Arii, J. Nucl. Mat. 204(1993) p252.

[15] J. D. B. Lambert, R. Mikaili, K. C. Gross, R. V. Strain, T. Aoyama, S. Ukai, S. Nomura and N. Nakae, Proc. BNES Conf. on Fuel Management and Handling, paper 64 (1995).

[16] J. H. Bottcher, M. Inoue and S. Nomura, Int. Conf. on Future Nuclear System, Proceeding, vol.2, p1086 Yokohama, Japan, October 5-10, 1997.

[17] T. Mizuno, S. Nagai, T. Itaki, N, Nakae, K. Tanaka and J. Komatsu, Int. Conf. on Reliable Fuels for Liquid Metal Reactors, Tucson, p5-28(1988).

[18] T. Uwaba and T. Mizuno, to be published in Nucl. Tech..

[19] S. Nomura, S. Shikakura, S. Ukai, I. Seshimo, M. Harada, I. Shibahara and M. Katsuragawa, Int. Conf. on Fast Reactor and Related Fuel Cycles, Kyoto, p7.4-1 (1991).

[20] M. Naganuma, K. Maeda, N. Nakae, J. Rouault, J. Noirot, G. Crittenden and C. Brown, to be published in the ICONE-6 San Diego, CA. USA (1998).

Japan - Russia Seminar

Research and Development on MOX Fuel for LMFBR

8-10, December, 1997

N. NAKAE

Nuclear Fuel Cycle Development Division

Power Reactor and Nuclear Fuel Development Corporation

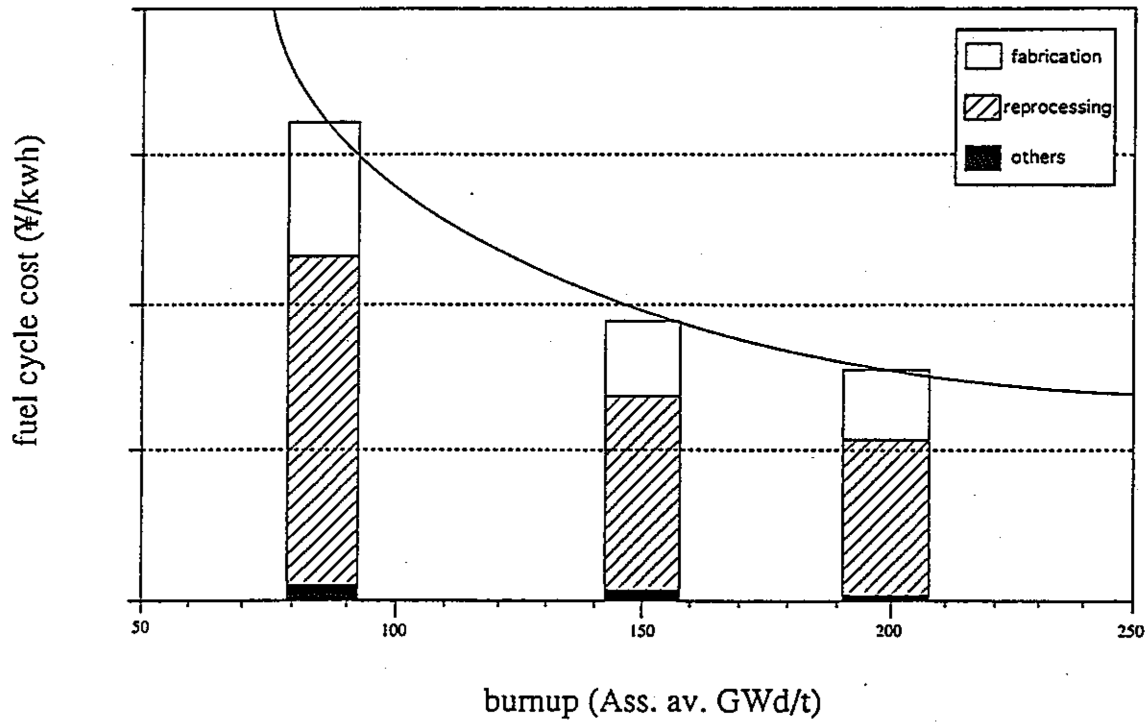
Target of the R&D on High Burnup Fuel

- To achieve the assembly averaged fuel burnup of more than 150 GWd/t

  corresponding pellet peak burnup : 250 GWd/t

  corresponding peak neutron fluence : 5×10^{23} n/cm^2

  (250 dpa)



Relation between burnup and fuel cycle cost

R&D Items

1. Fabrication Technology Development
2. Physical Property Measurement
3. Irradiation Test (including PIE)
4. Analytical Code Development
5. Core Material Development

Fabrication Technology Development

(Low Density Pellet development)

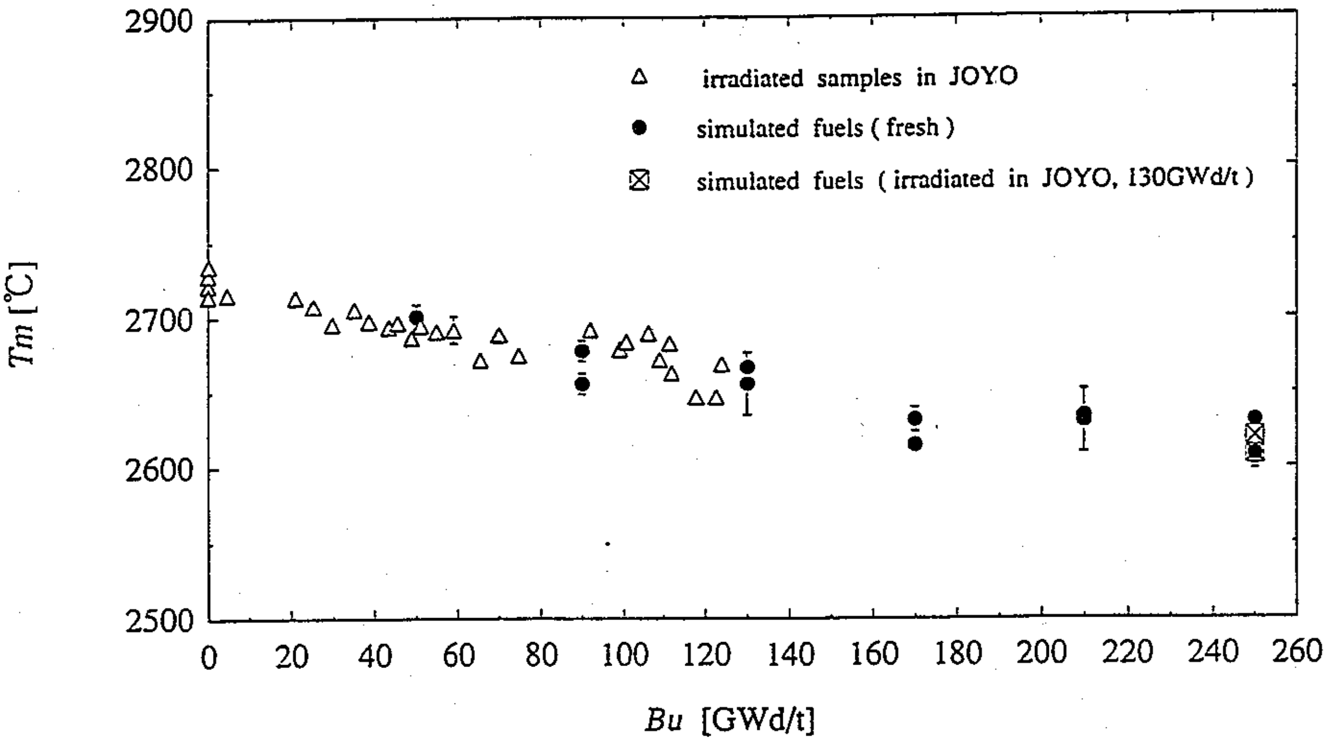
- Selection of appropriate pore former materials
- Optimization of the characteristic of granular powder
- Optimization of the pellet sintering procedure
- Obtaining the homogenions pore former distribution

Fabrication Technology Development

(Annular Pellet Development)

- Development of the equipment for compressing and forming pellet shape

  (core rod development)
- Optimization of the characteristic of granular powder
- Clarification of sintering mechanism
- Development of the inspection equipment



The burnup dependence of the melting temperature

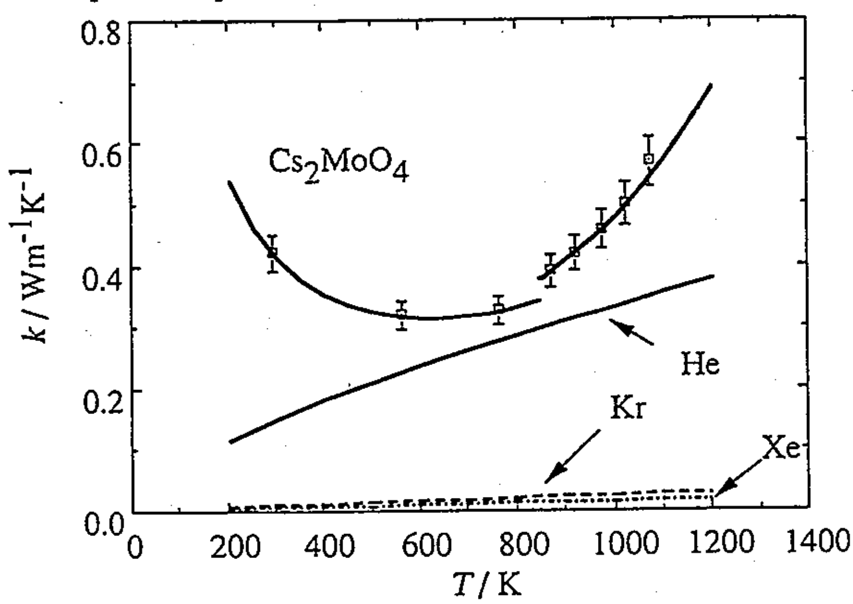
Investigation of thermal conductivity of Cs_2MoO_4

[Measurement]

○ Sample (disk)
Dense : 94.3%TD
Dimension : 9.9mm diameter by 1.6mm thick
Surface : coated with graphite

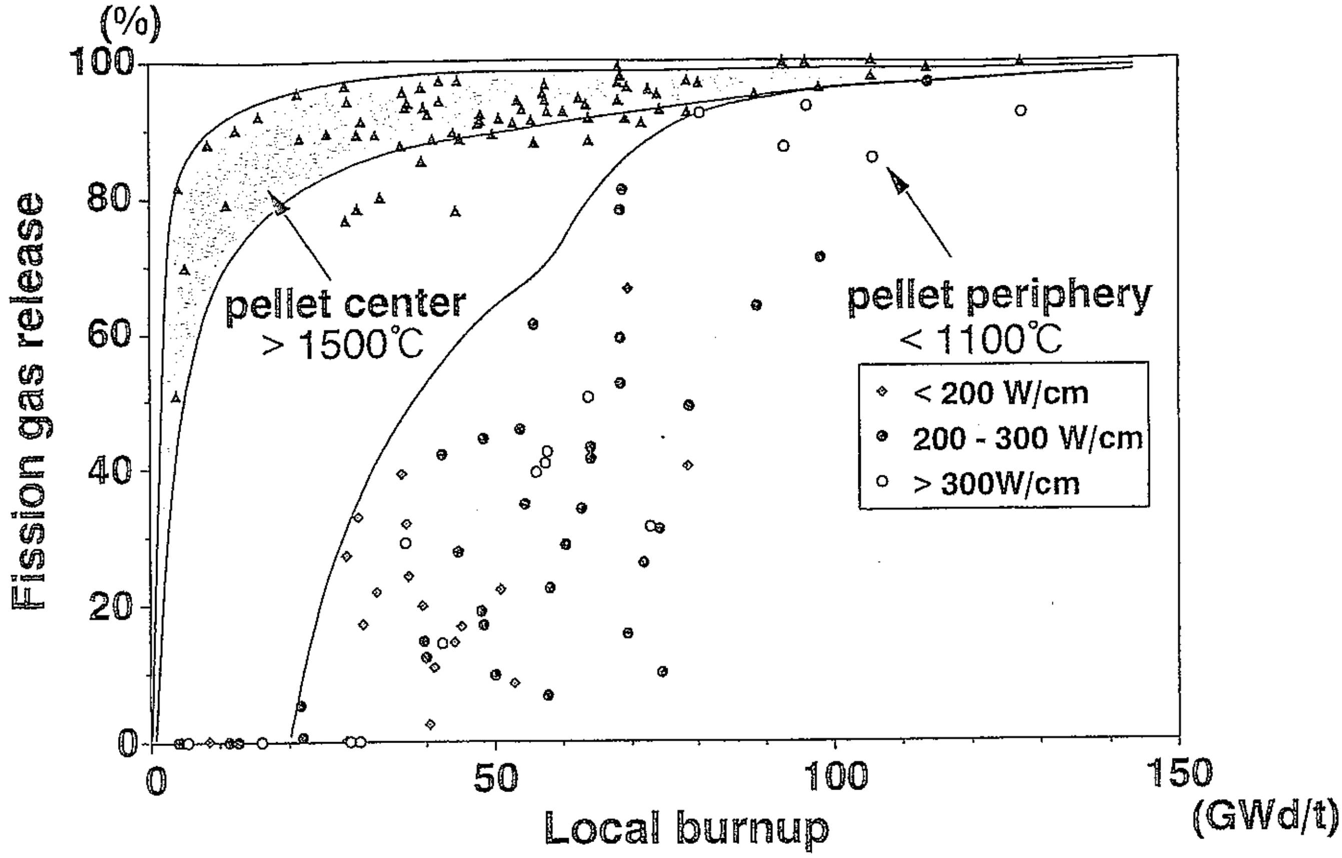
○ Procedure of evaluation
1. Measurement of thermal diffusivity
 - using laser flash
 - in a vacuum (~10-2Pa)
 - from RT to 1073K
2. Calculation of thermal conductivity
 - thermal diffusivity = measured value
 - specific heat capacity = reference
 - uncertainty in thermal conductivity = about 7%

[Results]

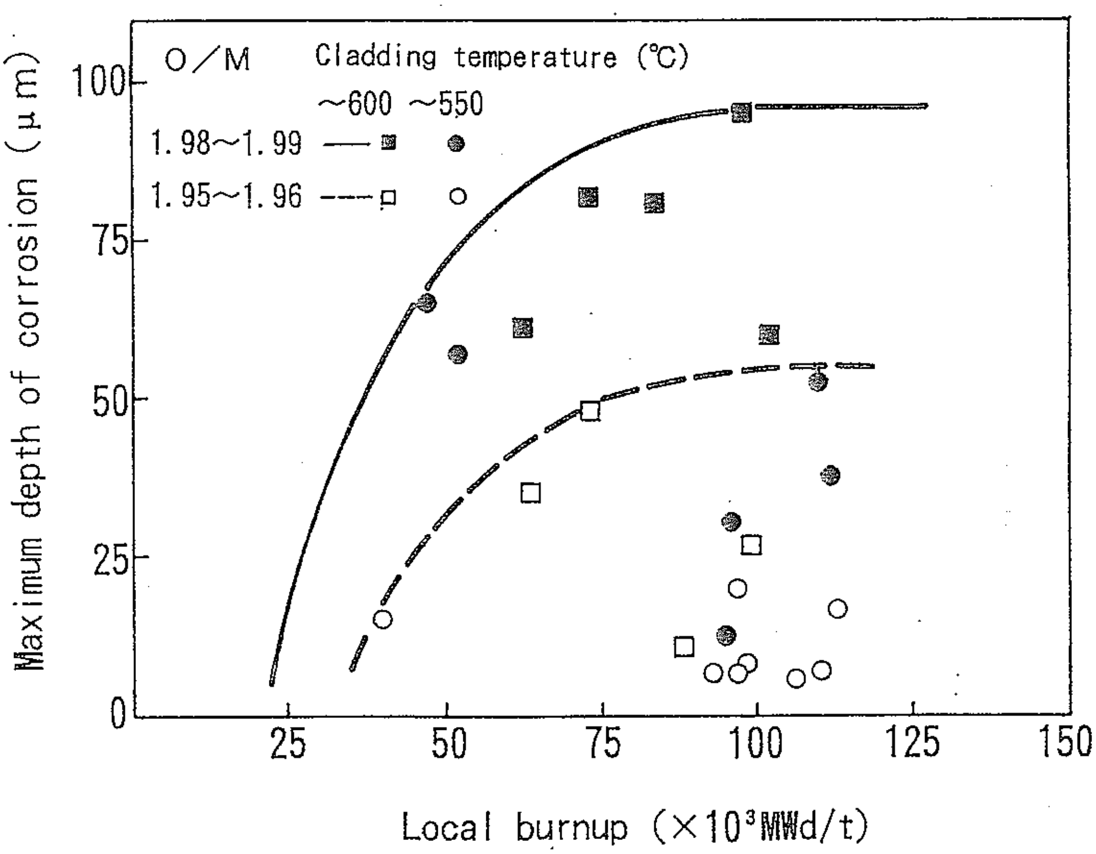


thermal conductivity of Gases
- evaluated using the Bird model
- evaluated values of Kr and Xe = $\sim 10^{-2}$ [$Wm^{-1}K^{-1}$]

[References] T.Ishii & T.Mizuno, J.Nucl.Mat vol.231 (1996)
R.B.Bird et al., " Transport Phenomena " (1962)



Fission gas release rate as a function of burnup



Maximum FCCI vs. local burnup

Operational Reliability Testing in EBR-II

<u>Operational Transient Tests (OTT)</u>

to determine the performance capability of reference and advanced MOX fuel pins during steady-state irradiation combined with a range of operational and overpower transients

<u>Run Beyond Cladding Breach (RBCB)</u>

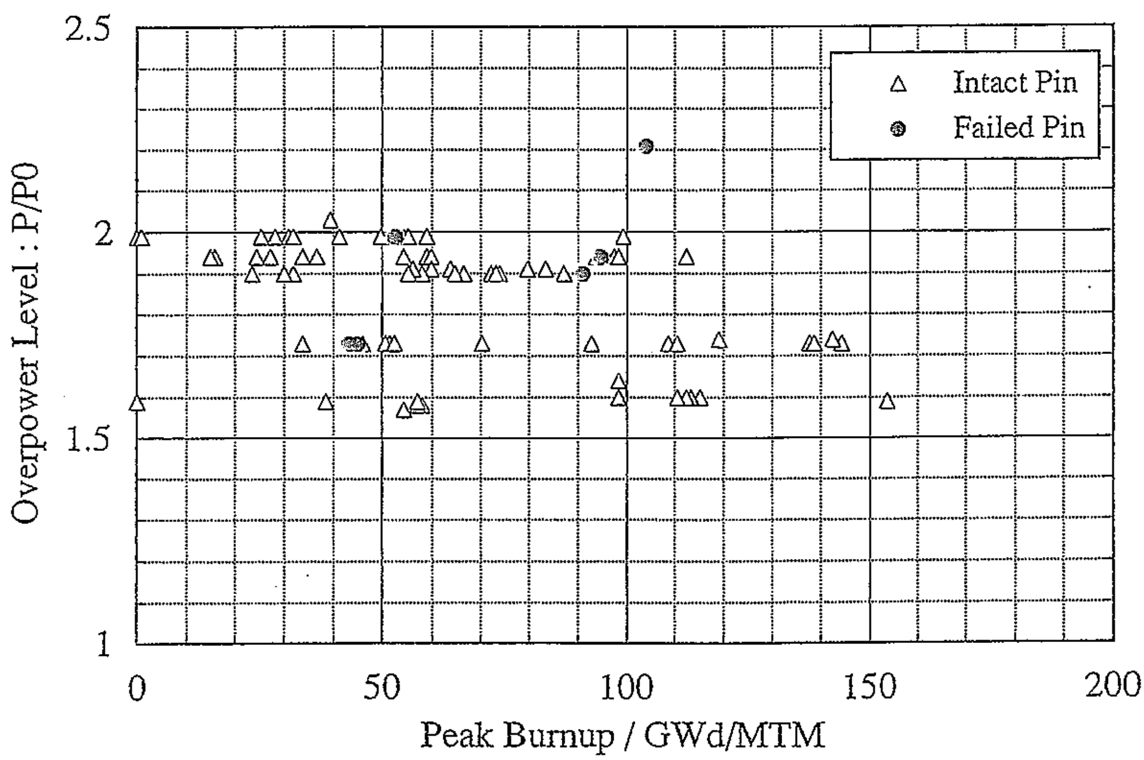
to investigate the behavior of breached MOX fuel pins during continued irradiation

<u>Phase-I</u>

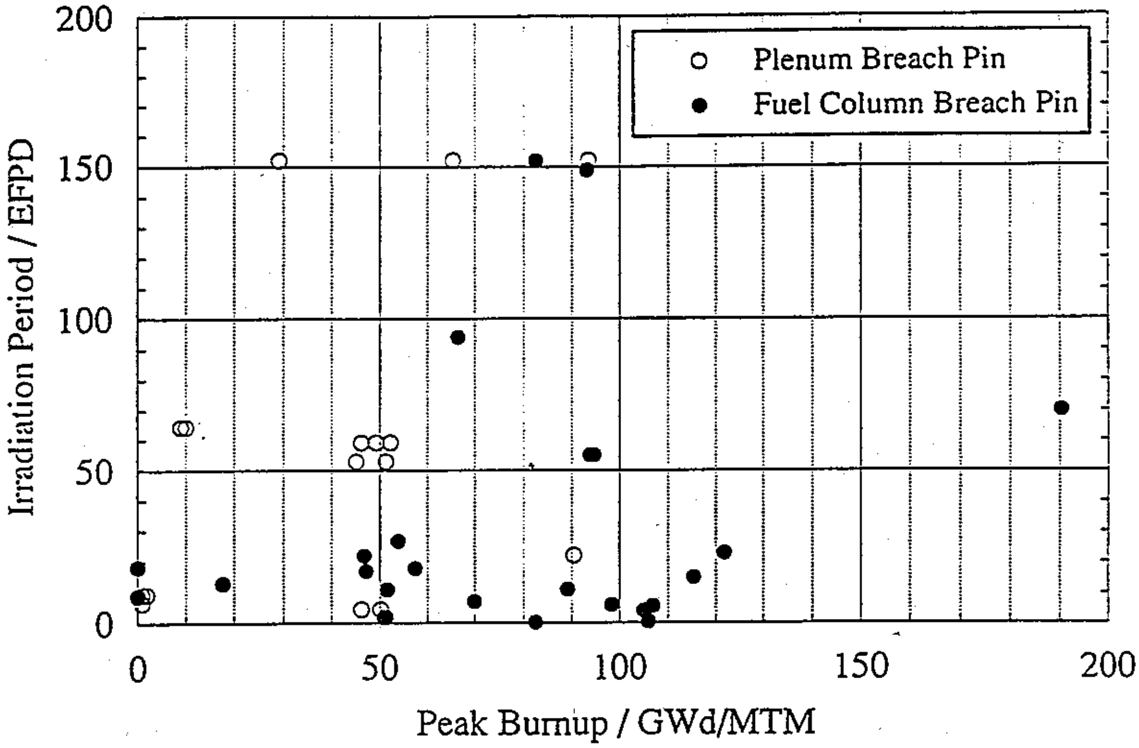
for MONJU licensing
PNC316

<u>Phase-II</u>

for DFBR and/or Large Scale FBR
Advanced Austenitic Steel, Ferritic Steel, ODS
Annular Pellet, Axial Heterogeneous Fuel Pin



Results of Overpower Transient Tests in ORT Program



Results of RBCB (Run-Beyond-Cladding-Breach) Experiments in ORT Program

ANALYTICAL CODE DEVELOPMENT

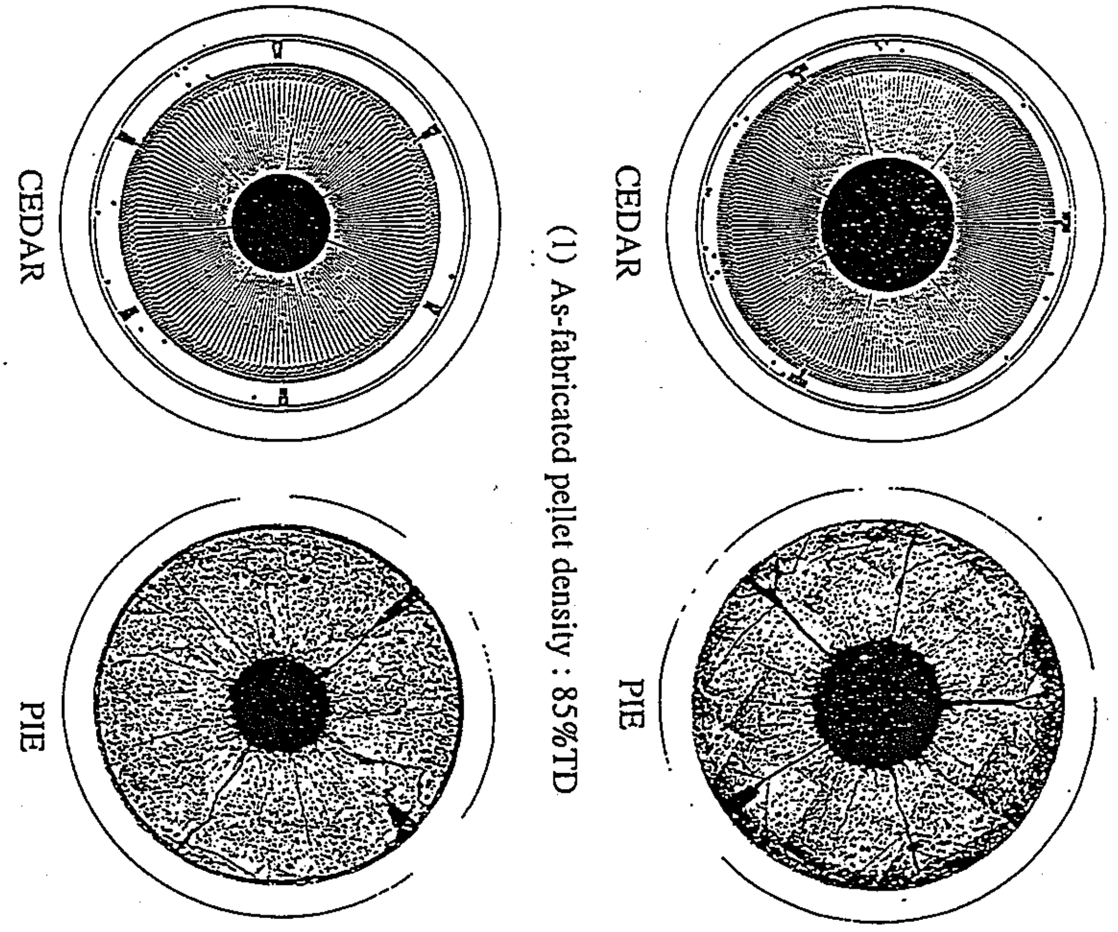
CEDAR : Fuel pin behavior, steady and transient condition

BDI : Fuel pin bundle and wrapper tube behavior

MINERVA : Cs migration behavior

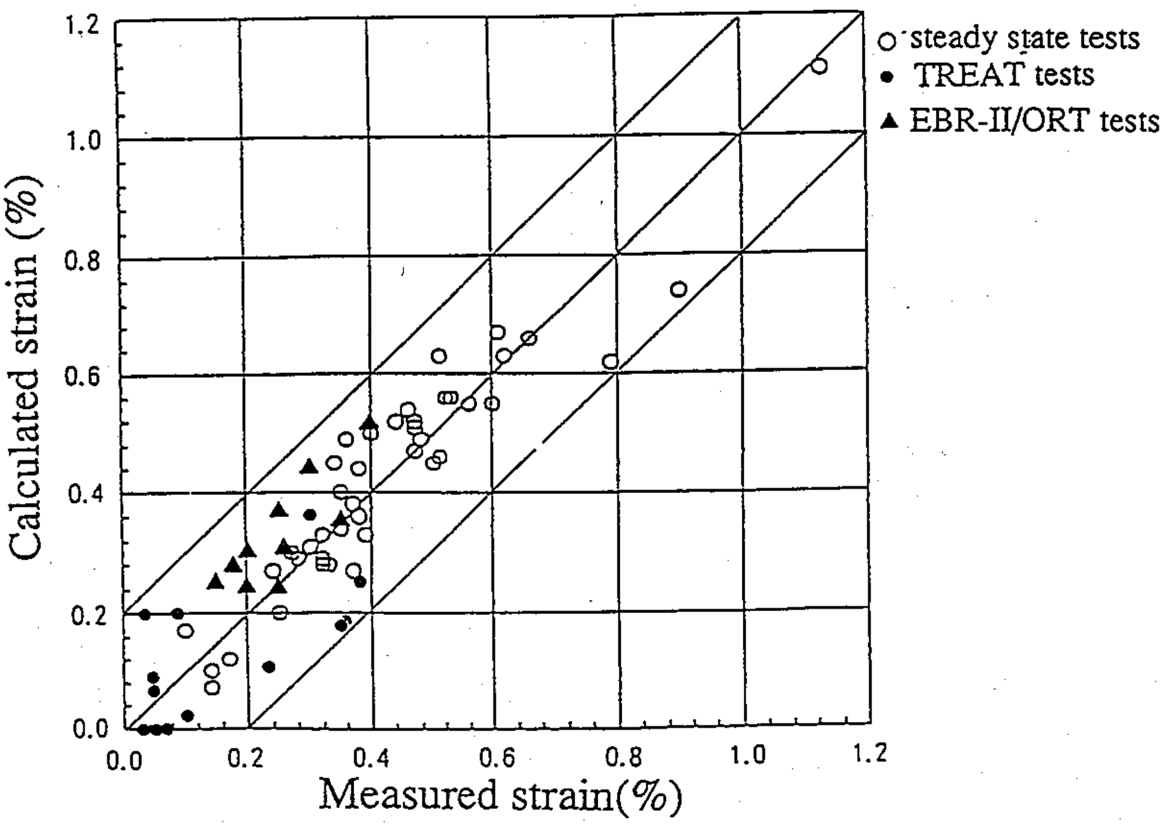
SAFFRON : Breached fuel behavior

CORAL : Absorber pin behavior

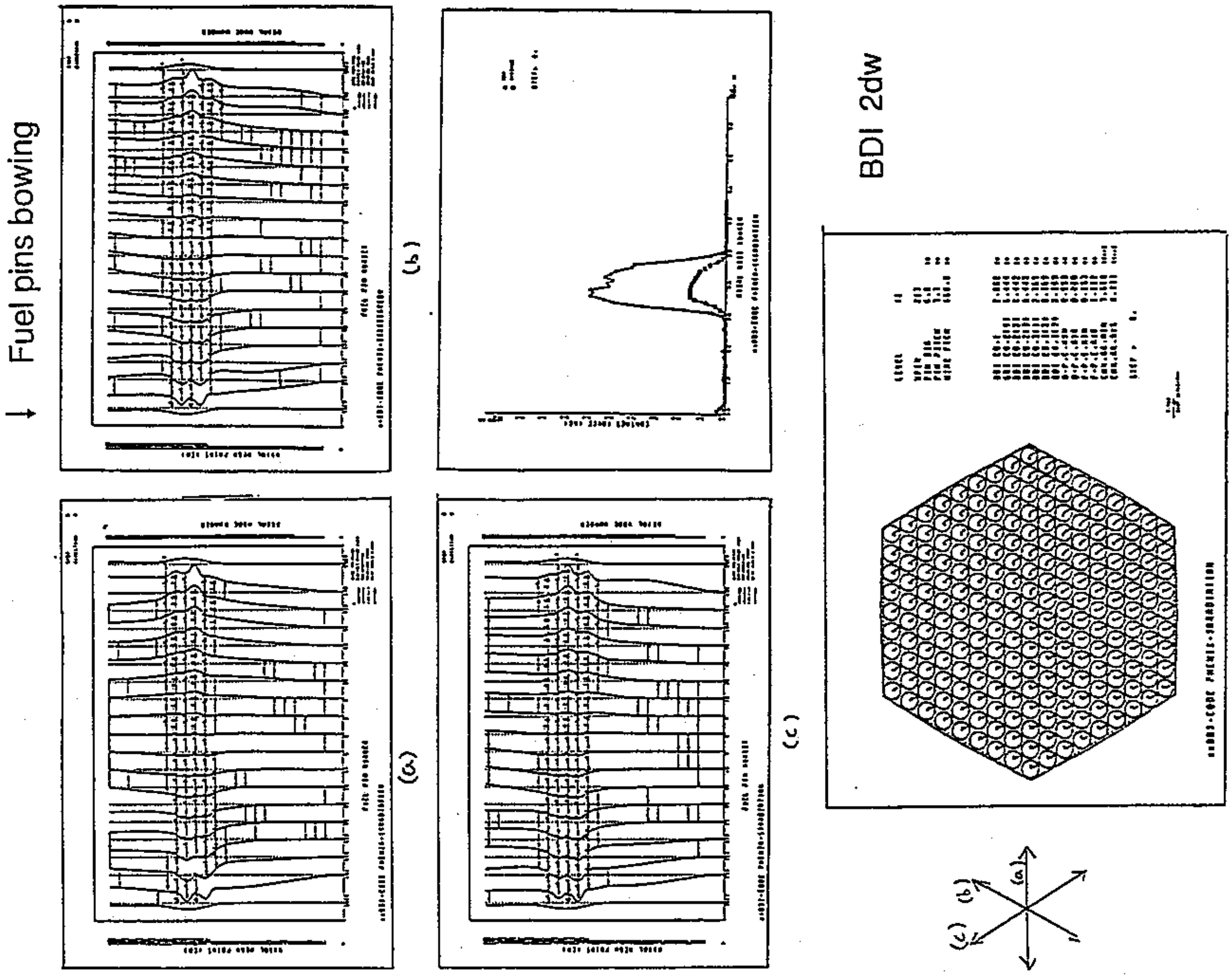


Fuel Restructuring of High and Low Density Fuel

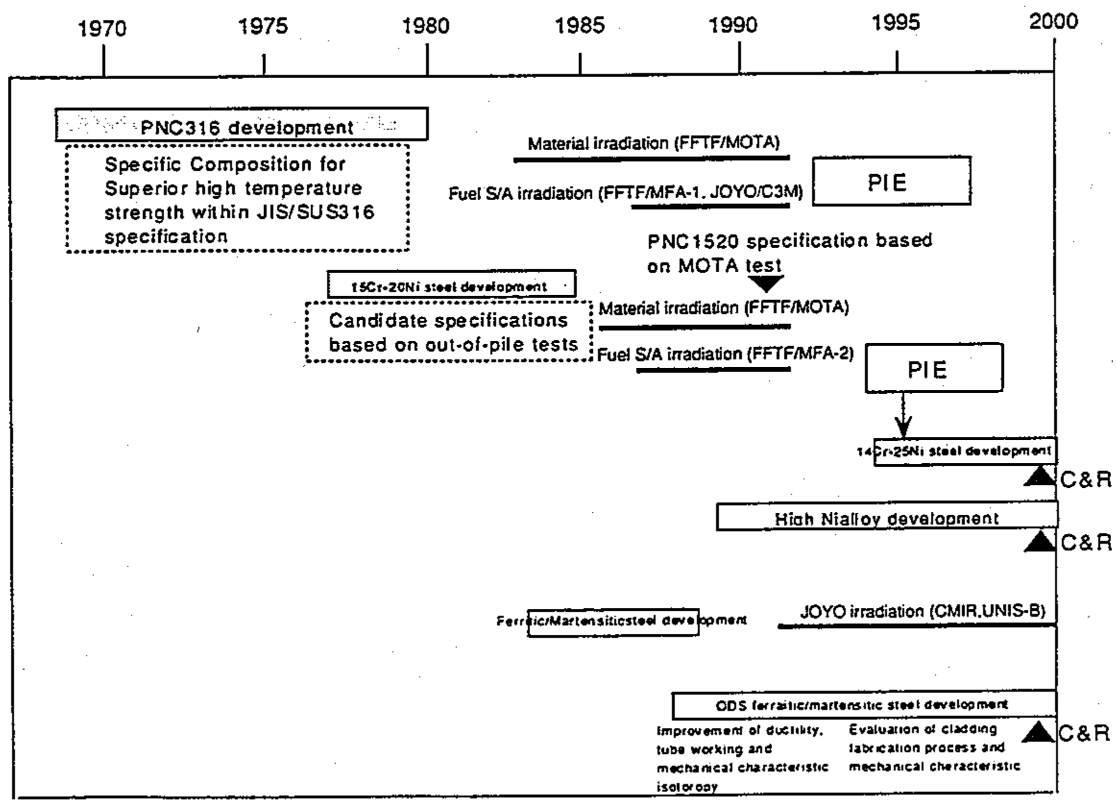
(Burnup : ~110,000MWD/MTM)



Validation on Diametral Strain

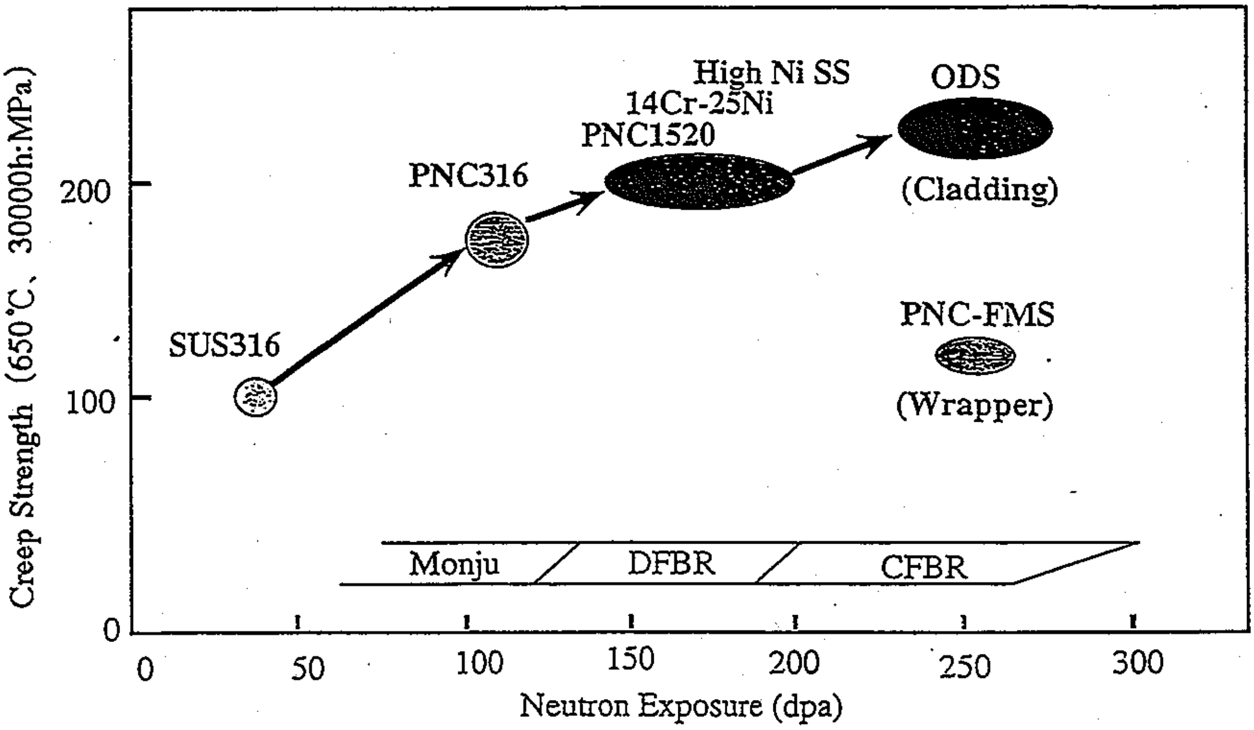


An Example of the Result of BAMBOO Code
(217 pins bundle, 2dw by irradiation)

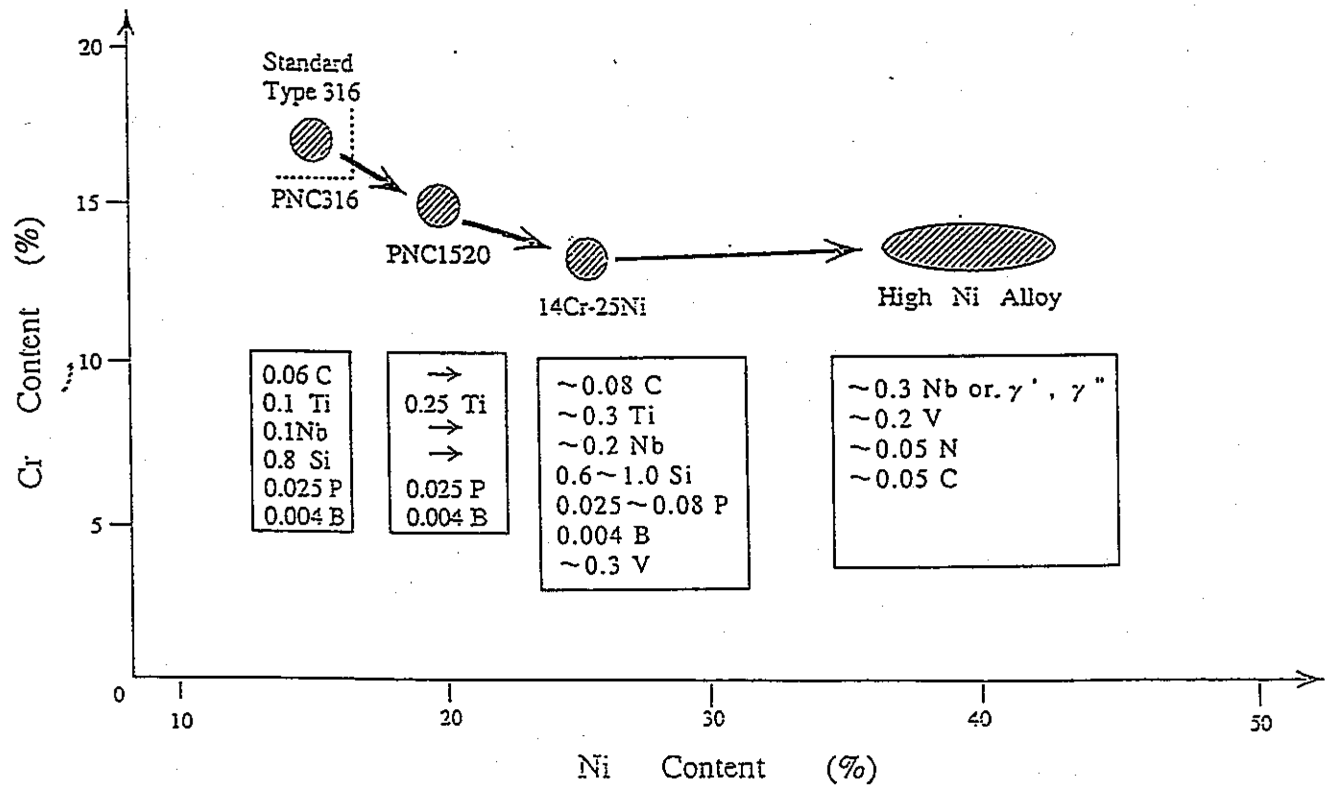


History of Fast Reactor Core Material Development

Overview of Core Materials Development in PNC



Austenitic Steel Development in PNC
Focusing on Swelling Resistance Improvement



International collaboration

[Past]

1. Fuel Behavior Analysis Code Development (NUS/ERDA, ANL/ERDA, WH/DOE)
2. Irradiation Tests in GETR (USA), DFR (UK), Rapsodie (FR), Phenix (FR) (GE/ERDA, UKAEA, CEA)
3. Irradiation Test in FFTF (USA) (WH/DOE)
4. Fuel Reliablity Test in EBR-II (USA) (ANL/DOE)
5. Information Exchange on Fuel and Materials
   (Europe UKAEA, KFK, CEA)

International Collaboration (continued)

[Current]

1. Consultation on Annular Pellet Behavior (UKAEA/BNFL)
2. Irradiation Test in Phenix (FR) (CEA)
3. Post Irradiation Examination on PFR High Burnup Pin (UKAEA/BNFL, CEA)
4. Post Irradiation Examination on Core Materials (ANL/DOE)
5. Collaborative Study on Sphere Packing fuel (PSI)
6. Information Exchange on Fuel and Materials (mainly with CEA)

[Plan]

1. Irradiation data on High Burnup Vibro Packing Fuel (ISTC)
2. Irradiation Test of Vibiro Packing Fuel in BN-600 (Russia)

Problems to be solved

1. Swelling characteristic of cladding materials at high fluence of more than 3×10^{23} n/cm^2 (150dpa)

2. FCCI behavior at high burnup

3. Thermal behavior under steady and transient (off normal) conditions

4. Irradiation behavior of breached fuel under the Run Beyond Cladding Breach (RBCB)

Conclusion

- The R&D activities including irradiation test on MOX fuel for FBR have been successfully performed in order to achieve the fuel burnup and linear heat rating as high as possible.

- The good irradiation performance has been obtained on MOX fuel for FBR with high burnup including transient over power and run beyond cladding breach conditions.

- The performance analytical codes developed here can predictt the fuel irradiation behavior well.