

SUMMARY OF THE MEETING

SESSION I - FABRICATION OF MIXED CARBIDE AND NITRIDE FUELS

Chairman: H. Blank

Co-Chairman: A. Majorshin

1. Survey of presentations

Four papers were presented in this session which all covered work performed mainly during the years 1986/87. They present three different levels of development for the fabrication techniques for carbide and nitride LMFBR fuels based on the carbothermic reduction of UO_2 and PuO_2 powders:

- a) The fabrication of mixed carbide and nitride fuel with $Pu/(U+Pu) = 0.7$ as fuel for the FBTR and fabrication studies of mixed carbide and nitride fuel with $Pu/(U+Pu) = 0.2$ for the future prototype fast breeder reactor (PFBR). Thermophysical properties like thermal conductivity and hot hardness data of both U and Pu rich MC and MN. The out-of-pile compatibility of these fuels with 316 SS cladding (work at BARC, India).
- b)
 - i) Studies of the carbothermic reduction and nitriding as the first step for mixed nitride fabrication;
 - ii) the fabrication for thermally stable UC pellets of different density by the use of pore formers as a preliminary exercise for the fabrication of thermally stable mixed carbide fuel pellets (work at JAERI, Japan).
- c) The fabrication of mixed nitride fuel in an oxide facility for irradiation experiments in a prototype fast reactor (Phenix) in order to demonstrate the feasibility of nitride fabrication in existing commercial MO_2 fabrication facilities. This is one of the prerequisites for attaining FBR fuel cycle economy in Europe by substituting nitride fuel for the currently used oxide (work at CEA, Cadarache).

There are obviously no problems in reaching fuel densities for carbides and nitrides in the range of 80 to 85% of the theoretical density with the currently used fabrication procedures in the different laboratories. Yet the degree of residual oxygen and carbon in the nitrides depends mainly on the details of the different fabrication methods employed.

2. Summaries of the papers presented and current status

2.1 Status of (U,Pu) C and (U,Pu)N fuel development in BARC (presented by P.V. Hegde, India)

The carbide fabrication flow sheet for the preparation of Pu-rich and U-rich mixed carbides is well established. Pu-rich mixed carbides have been fabricated on large scale for the first core of FBTR which became critical in 1985. Additional fuel sub-assemblies have been fabricated at the beginning of 1987 to increase the power level. U-rich mixed carbide i.e. (U_{0.8} Pu_{0.2})C with < 1000 ppm of oxygen and ≤ 10 w/o M₂C₃ has been fabricated with a batch size of 500 g. Now, efforts are on the way to increase the capacity of carbide fuel fabrication to 50 kg per day, so that we can meet the requirement of PFBR-500.

Nitrides: Development work in the field of both U-rich and Pu-rich mixed nitrides is in progress. Initial trials on with a batch size of 500 g have shown high amounts of 'O' and 'C' impurities (i.e. ~ 0.5 w/o each). In order to minimize these impurities the efficiency of the carbothermic reduction is being improved by various modifications. Thermophysical properties like thermal conductivity and hot hardness data for both U-rich and Pu-rich carbides and nitrides are being generated. The out-of-pile chemical compatibility of these fuels with SS 316 cladding is also being evaluated.

2.2 Fabrication of U-Pu Mixed Nitride and Thermally Stable Carbide Fuels (presented by M. Handa, JAERI, Japan)

Synthesis of (U,Pu)N by carbothermic reduction in a mixed 92% N₂ - 8% H₂ stream was studied. The initial mixing ratio of

MO_2/C and the reaction time was taken as parameter. The carbon and oxygen contents in the green pellets and also to weight loss and the lattice parameter of (U,Pu)N were measured during the reaction in order to follow the reaction process. In this way (U,Pu)N powder containing 200-300 ppm O_2 and 1000-3000 ppm C has successfully been prepared.

Thermally stable UC pellets having low density were also prepared by using a wax-type pore former and 0.1 wt %Ni sintering aid. An almost linear relationship between the pellet density and the amount of the pore former was obtained. Spherical pores, 10 - 100 μm in diameter were uniformly distributed in a high density fuel matrix. It was shown that the oxygen content increased somewhat and the composition slightly shifted to the hyper-stoichiometric side when the pore former and the Ni sintering aid were used.

2.3 Quality Control of U-Pu Mixed Carbide and Nitride Fuels (presented by M. Handa, JAERI, Japan)

The special characterization methods and apparatus developed for characterization of U-Pu mixed carbide and nitride fuel are described. For the determination of oxygen in carbides, a new sample preparation procedure was developed and the true oxygen content was successfully estimated by extrapolating to zero the oxygen values determined on powder samples at various exposure times in an Ar atmosphere. It was shown that the carbon loss via methanegas produced by the reaction of the carbide with trace moisture at room temperature was relatively small, presumably because of the adsorption of methane on the fuel surface. For the X-ray diffraction analysis, a sophisticated method was developed. It consists of mixing the advanced fuel with epoxy resin to prevent the samples from being oxidized during the measurement. The occluded gases in carbide fuels were measured at the temperature range from 1200°C to 1600°C and continuous release of CO gas above 1400°C was observed. These results indicate that these gases should not affect the inner gas pressure in the fuel pin during irradiation.

2.4 Mixed nitride fuel fabrication in conventional oxide line
(presented by H. Bernard, CEA, France)

Mixed UPuN pellets can be obtained in a classical UPuO₂ facility without any significant modification of the set-up. Basic laboratory studies, performed by thermogravimetry, off-gas analysis and dilatometry, allowed us to define an optimized fabrication flow-sheet, based on the carboreduction of the oxides in nitrides. With this process, carbon and oxygen contents can be adjusted at a controlled level.

An industrial validation, conveniently limited to uranium nitride, is now under way at a conventional UO₂ oxide plant where UN powder and pellets are handled without glove boxes.

The NIMPHE 1 capsule, currently irradiated in PHENIX, is the first experiment of the He-bonded dense fuel irradiation programme. Eleven pins are filled with slightly different kinds of UPuN fuels which covered the following parameter ranges:

geometrical pellet density	in the range 81 - 84% TD
smear density	75 - 78% TD
radial gap	170 - 270 μm
carbon content	0.009 - 0.050 wt%
oxygen content	0.034 - 0.225 wt%

Two other irradiation tests are planned in PHENIX for the next year.

SESSION II - PROPERTIES OF ADVANCED FUELS BEFORE, DURING AND AFTER
IRRADIATION AND THEIR CONTRIBUTION TO FABRICATION
TECHNOLOGY

Chairman: C.R. Prunier

Co-Chairman: M. Handa

1. Survey of presentations

The six contributions to this session can be separated into two groups:

Four papers which treat the aspects of fuel R and D, and fabrication processes (by dry and wet routes) for carbide and nitride irradiation tests (papers by Bogatyr, Richter, Blank and Ledergerber).

In the second group there are two papers which refer completely or mainly to mixed oxide fuel. The first one treats the in-pile performance of vibropack mixed oxide fuel in BOR-60. The fuel was prepared by an electrochemical process by which the fuel is granulated in a salt melt system based on alkaline metal chlorides and which is suitable for the remote handling technique (paper presented by Mr. Majorshin). The second paper describes the development of mixed oxide fuels by PNC, Japan, as well as the plans for the evaluation of metal fuel, carbide and nitride (presented by Mr. Endo).

Today it appears to be impossible to make a decision between carbide and nitride (or metallic fuel) for substituting the presently employed oxide fuel. At present, nobody can predict the irradiation performance of carbide and nitride properly, even if we suppose them to be at least as good as the oxide. Up to now mainly irradiation results on carbides are available (French RAPSODIE programme, US carbide programmes in EBR II and the US-Swiss AC3 test in FFTF the (U,Pu)C irradiations in BOR-60 of the USSR, the joint PSB-CEC carbide irradiation in KNK2, Karlsruhe, and the swelling studies of CEC Karlsruhe). One should not forget the reprocessing. In the discussion of the six papers the problems of closing the fuel cycle were mentioned. As regards reprocessing, the direct dissolution of carbide poses certain problems due to the formation of organic compounds, whereas nitride fuel may pose a problem with regard to ^{14}C .

2. Summaries of the papers presented and current status

2.1 State-of-the art for carbide-nitride fuel in the USSR (presented by S. Bogatyr, USSR)

To provide for the wide use of carbide-nitride fuel in large fast reactors some problems must be solved, particularly, the mechanical fuel-clad interaction must be eliminated or reduced, the fuel-clad compatibility must be improved, as well as, criteria must be established for choosing the optimized parameters of fuels. The paper overviews design-experimental work performed to study carbonitride fuel pin serviceability in BR-5, BR-10, BOR-60 reactors.

The results of investigations confirmed the feasibility of reaching a 10% burnup in carbide-nitride helium bonded fuel rods and made it possible to refine the major fuel parameters that provide for the serviceability of the selected fuel pin, design, up to the burn up of 10% a/o. Besides the further experimental activity, directions have become more clear:

1. to study the irradiation stability of carbide-nitride fuel at higher heat generation rates and the core temperature at higher burnups;
2. to conduct mass tests to verify the optimized design and testing parameters;
3. to study prospects of using Na-K bonded fuel pins

2.2 Fabrication processes and characteristics of LMFBR carbide and nitride fuels and fuel pins (presented by K. Richter, CEC, JRC Karlsruhe)

Mixed uranium-plutonium carbide and nitride fuels can be fabricated economically by carbothermic reduction of oxides, both with the conventional comminution-pressing-sintering technique, as well as, by direct pressing of the reaction product. The industrial equipment for the well established oxide fuel fabrication and reprocessing technology can be used to a large extent.

An optimized fuel pin concept with large pin diameter and He bonding, a smear density of 75-80% fuel operating at a linear power of 45-80 KW/m to a high burn up have been the basis of our investigation. Both carbides and nitrides can be fabricated with a good control of density in the range of 80-88% TD and composition. Carbides with an oxygen content of 1000-3000 ppm are single phased. For oxygen contents below 1000 ppm the material contains 2-15% M_2C_3 , due to the excess carbon used in the master blend. Nitrides with 1000-3000 ppm carbon and oxygen can be fabricated with a stoichiometric ratio of C/MO_2 in the master blend. Utilization of excess carbon, combined with a heat treatment in a nitrogen-hydrogen mixture, allows the fabrication of single phase (U,Pu)N with an oxygen and carbon content below 1000 ppm. The results from chemical, XRD and ceramographic analysis performed on carbide and nitride fuels fabricated for irradiation experiments, are in good agreement and assure the quality control of these fuel types. The new fabrication method, the direct pressing, simplifies the fabrication process, reduces radiation exposure of the personnel and improves the thermal stability of these fuels.

2.3 Performance of He-bonded LMFBR carbide fuels at the beginning of life and consequences for their subsequent behaviour (presented by H. Blank, CEC, JRC Karlsruhe)

The in-pile performance for carbide fuels is more sensitive to their properties resulting from the fabrication method and to the pin design and irradiation parameters than oxide fuel. In the He-bonded pin concept the fuel has its highest operation temperatures at the beginning of the irradiation (BOL). Hence, the probability exists at BOL that thermal restructuring occurs and the as-fabricated fuel structure gets lost. To avoid this, and to have the fabrication porosity available for the accommodation of swelling later in life, fuel properties, pin design and reactor operation conditions have to be chosen in such a way that the outer part of the fuel cross section with radius $R > R_{st}$ retains its fabrication structure.* It is shown that the oxygen content of a mixed carbide fuel plays a critical role for its thermal in-pile stability at BOL. In a given

* The radius R_{st} corresponds to a given thermal in-pile stability limit of the fuel

pin design and reactor operation a carbide containing 270 ppm oxygen was stable although the fabrication gap was closed already after 0.52% burn up whereas a carbide with about 1600 ppm oxygen was not stable. The subsequent irradiation behaviour of the stable carbide is relatively straightforward and can be modelled on the basis of the fuel structure which exists at the situation when the fuel/clad has just been closed.

2.4 Advanced fuel for fast breeder reactors produced by gelation method (presented by G. Ledergerber, Switzerland)

The Internal Gelation Process has been qualified for the preparation for carbide spheres in two sizes needed to get the required smear density of a pin design for a burnup of 10 a/o of fissile material. Therefore a "direct conversion - fuel fabrication" process with a significant reduced number of processing steps and with no fabrication step which requires high purity atmosphere is available.

The direct packing of the spheres into a pin is achieved by vibratory filling, which results in a very homogeneous fuel column with a comparable behaviour to a pellet-fuel column as far as restructuring, fission gas release, swelling and fuel clad interaction is concerned.

Preliminary experimental results on the fabrication of nitride look promising in preparing nitride spheres. This means all the process advantages of sphere-pac fuel will be valid for this candidate advanced fuel.

EIR will carry on development of process improvements, fabricating experimental amounts of fuel, studying the loss of sphere from defective pins and develop a general sphere pac code.

2.5. Analysis of performance of BOR-60 vibropack uranium-plutonium oxide fuel pins (presented by A. Majorshin, USSR)

Presented are results from in-pile tests and material investigations on the BOR-60 vibropack U-Pu oxide fuel pins. The vibropack fuel is

also being investigated as an alternative to the traditional pelleted fuel for fast reactors which may appear to be more exact from the point of view of the remote automatized technology.

Over a period from 1981 to 1986 about 120 fuel sub-assemblies have been operating up to 15 a/o burnups. A mean burnup for the fuel pins being now removed from the core is 10-12%. The fuel pins operate at a maximum linear heat rate of 500 W/cm and cladding temperature of 710°C. The depth of the cladding corrosion in the fuel pins operated to 13-15 a/o burnups has reached up to 60-120 µm. In the fuel pins with the substoichiometric (o/m = 1.97-1.98) $UPuO_2$ the depth of corrosion was 20 µm.

By toughening technological requirements to the fuel fabrication process it became possible to decrease probability for fuel pin failure down to 0.1% at burnups at 12 a/o. The successful operation of the BOR-60 with the vibropack $UPuO_2$ fuel has confirmed the validity at the fuel pin design concepts and technological approach and offered the new lines for their further improvement.

2.6 Present status of FBR mixed-oxide development and plan for R & D advanced fuel development in PNC (presented by H. Endo, Japan)

Power Reactor and Nuclear Fuel Development Corporation (PNC) will perform development of the fabrication technology for mass production, evaluation of design, safety and economy and technological evaluation of the availability of advanced fuels.

Although PNC has little experience of advanced fuels, PNC has a lot of experience in MOX fuel cycle technology. It is very important to further develop MOX fuel cycle technology more than before, besides it is planned to carry out the R & D of advanced fuels, and to establish a base technology for them.

First of all, it is necessary to carry out the evaluation of fuel fabrication and design technology by the irradiation. And we will confirm the fuel design itself by the safety evaluation. Last of all, we plan to carry out the evaluation of practical application by

the evaluation of economics of fuel cycle. These plans must be based on our MOX experience under collaboration with JAERI.

MOX experience is shown as follows:

We have fabricated more than 80 t of MOX fuels since we started operation of the first fuel fabrication facility in 1966. There are two major fuel fabrication facilities, the Plutonium Fuel Development Facility (PFDF: several Kg/year ~ ton/year) being operated since 1966 and Plutonium Fuel Fabrication Facility (PFFF: several tons/year ~ 10 tons/year) being operated since 1972. Further we have continued the construction of the third facility, the Plutonium Production Facility (PPF: several tons/year) to supply MOX fuel to be loaded to "MONJU" since July 1992. The construction was completed in October, 1987.

PFFF will start operation in 1988 and has adopted the remote and automatic operation technologies which are essential for a full-scale MOX fuel fabrication facility.

PNC will carry out the R & D of the advanced fuel by means of MOX experience and PFDF will mainly be used to fabricate the advanced fuel.