MATERIALS SCIENCE AND TECHNOLOGY

LIQUID-METAL REACTOR FUELS FOR COMMERCIAL ENERGY PRODUCTION

All Speakers Invited

Session Organizer: Douglas C. Crawford (ANL-Idaho)

1. History of Fast Reactor Fuel Development, *J. H. Kittel, B. R. T. Frost (ANL), J. P. Mustelier (COGEMA-France)*

Most of the first generation of fast reactors that were operated at significant power levels employed solid metal fuels. They were constructed in the United States and United Kingdom in the 1950s and included Experimental Breeder Reactor (EBR)-I and -II operated by Argonne National Laboratory, United States, the Enrico Fermi Reactor operated by the Atomic Power Development Associates, United States and DFR operated by the U.K. Atomic Energy Authority (UKAEA).

The early metal fuel element designs experienced difficulties in achieving fuel burnups beyond a few percent without cladding failures. The principal problems were swelling of the fuel at high burnups due to fission gas nucleation into bubbles and anisotropic dimensional changes if preferred grain orientations existed in the fuel.

Initial efforts to improve the dimensional stability of metal fuels focused on investigating the effectiveness of alloy additions such as molybdenum to uranium to maintain the fuel in the isotropic gamma phase at normal operating temperatures. In the case of EBR-II, the residual fission products (termed "fissium") left in the recycled uranium fuel after pyrometallurgical reprocessing fortuitously stabilized the gamma phase.

Progress was slow, and attention in the 1960s therefore largely turned to ceramic fuels and (briefly) to cermets. Although the ceramic fuels of interest have reduced thermal conductivity and high contents of diluent atoms compared to metal fuels, these disadvantages are offset by the higher melting points of ceramic fuels and their reduced tendency to swell at high burnups and high operating temperatures. These characteristics had been discovered through their earlier use in thermal reactors, notably in the U.S. Shippingport pressurized water reactor. General Electric workers in the United States carried out some landmark irradiations of UO_2 and mixed-oxide (MOX) (UO_2-PuO_2) fuel pins in test reactors to very high burnups in the late 1950s, which increased interest in their possible use in fast reactors.

Based on the promising results being obtained with oxide fuels, France and the USSR, respectively, chose MOX to fuel their early fast reactors, RAPSODIE and BR-5.

In their preliminary studies for the prototype fast reactor (PFR) in the early 1960s, the UKAEA selected a cermet fuel—a dispersion of uranium oxide particles in a stainless steel matrix—in the expectation that high burnups would be achievable. While this expectation was ultimately met, the loss in breeding gain was deemed to be unacceptable, and in late 1962, the decision was made to select MOX fuel for the PFR. At about this time, the USSR designed BOR-60 with MOX fuel elements, France chose MOX for Phénix, and Japan planned Joyo to operate with MOX fuel.

Experience with MOX fuel in DFR, EBR-II, RAPSODIE, BR-2, KNK-II, and BOR-60 showed that fuel burnups as high as 150 000 MWd/tonne could be reliably obtained. These findings led to the design of larger MOX-fueled reactors: Superphénix (France), BN-350 and BN-600 (USSR), CFR (United Kingdom), SNR-300 (Federal Republic of Germany), Fast Flux Test Facility and Clinch River Breeder Reactor (United States), and Monju (Japan).

More sophisticated development tools, meanwhile, became available including computer modeling of fuel performance under normal and off-normal conditions and facilities for studying fuel pin behavior under transient and loss-of-coolant conditions.

In the 1960s, attention also turned to other ceramic fuel forms—notably, carbides and nitrides but also sulfides and phosphides. Carbides and nitrides have higher fissile atom densities than oxides and higher thermal conductivities, both of which appeared to be attractive to reactor designers. Extensive test irradiations of these fuel forms were carried out, showing that high burnups could be achieved, although their retention of fission gases because of their lower operating temperatures gave rise to concerns over possible behavior during power transients. However, sufficient confidence was gained for India to choose carbide fuel for its Kalpakam PFR. Recently, the European Community (EC) revived its interest in nitride fuel—in part because of its compatibility with European reprocessing plants. A modest test program continues at the Transuranium Institute and at other EC sites.

In the case of metal fuels, a notable breakthrough in the 1970s and 80s was the realization that achievable burnups could be greatly extended by simply allowing the fuel to swell within its cladding ~25%, at which point the fission gas bubbles become interconnected to the surface of the fuel and little fuel swelling pressure is then exerted on the cladding. Using this design concept, burnups of 20% or more are being reliably achieved in experimental U-Pu-Zr alloy fuel elements for the integral fast reactor being developed by Argonne National Laboratory.

In the 50 yr since Enrico Fermi's historic demonstration of controlled nuclear energy, the whole gamut of fast reactor fuels has been studied. Interestingly, although oxide fuel has been chosen for most of the world's fast reactors, interest in metal and carbide fuels is still very much alive.

2. Status of Liquid-Metal Fast Breeder Reactor Fuel Development in Japan, *Masami Katsuragawa, Hidechiyo Kashihara, Michio Akebi (PNC, Oarai-Japan)*

The development of liquid-metal-cooled fast breeder reactors (LMFBRs) is advancing steadily in Japan. The experimental reactor Joyo has been operated satisfactorily, and construction of the prototype reactor Monju has been completed and is waiting for the start of operations. Thus, a vast amount of knowledge and experience has been accumulated through these development efforts.

There are still hurdles to overcome for commercial deployment of the LMFBR. For fuel development, especially, to keep its superiority in fuel cycle cost compared to that for light water reactors LWRs, LMFBR fuel has to possess very high burnup capability. It is estimated that LMFBR fuel must reach an average burnup of 150 to 200 GWd/t for a commercial reactor core. To attain this goal, further devotion to research and development (R&D) is required, making the best use of our accumulated knowledge and experience. Our current fuel development status and future R&D plans are summarized in this paper.

The Joyo fuels have been developed successfully. Any indication of a breach or even detrimental deformation of driver fuel pins has not been observed since the first criticality was achieved. Now the Joyo driver fuel has the same PNC316 cladding as the Monju fuel in order to demonstrate its excellent performance whole-core data.

Monju is a prototype FBR and will be the last one to start operating in this century. Its fuel consists of low-swelling austenitic steel PNC316 cladding and low-smear-density mixed-oxide (MOX) fuel pellets. The burnup capability of 94 GWd/t has been confirmed by means of several irradiation tests conducted in Joyo and in many foreign fast reactors involved in international collaboration.

Currently, our R&D target is shifting to commercial FBRs through demonstration FBRs. Its target irradiation burnup is approximately twice that of the Monju fuel. In addition, a very high power rate of 480 W/cm will be achieved using mixed-oxide fuel, which has been reliable.

In order to realize this high burnup capability, the Power Reactor and Nuclear Fuel Development Corporation (PNC) has been developing several types of advanced alloys, such as PNC1520, PNC-ODS, PNC-FMS, and high nickel alloy. PNC1520 is austenitic steel similar to PNC316, and has been under development as a principle candidate of the DFBR fuel cladding. Irradiation tests revealed that PNC1520 had sufficient performance for the DFBR's requirements. The PNC1520 will be used in a whole-core demonstration in Joyo and is planned for use in Monju. PNC-ODS is oxide-dispersion-strengthened steel cladding, which has the most promising low-swelling and high-temperature strength properties for CFBR use. PNC-FMS is martensitic/ferritic steel cladding and could be applied to wrapper tubes because of its excellent anti-swelling performance. Improvement of accuracy in thermal analysis calculation is most important to support the use of fuel pins with high linear heat rating. Investigation of annular pellet behavior is also needed. In these respects, accumulation of fuel irradiation experience in high power and high burnup conditions and their detailed analyses are required. Thus, a variety of irradiation test programs at Joyo and Monju, are being planned.

In Joyo, experimental irradiation tests will be carried out in extreme conditions. For example, a series of tests are planned in Joyo under power-to-melt at various burnups, high burnup and extremely high temperature run-to-cladding breach in specially designed irradiation rigs, and instrumental radiation rigs. For material irradiation tests, an irradiation rig (called MARICO) with excellent temperature control is under development. To intensify its irradiation capabilities, thermal output of Joyo is planned to be increased from 100 MW(thermal) for the current MK-II core to 140 MW(thermal) of MK-III core.

Monju will be used as an irradiation bed for large fuel assemblies to demonstrate their irradiation performance and reliability in normal steady-state conditions. Irradiation performance of DFBR and/or CFBR fuel will be confirmed in the Monju core prior to actual application.

As a candidate for high-performance fuel for the future, some types of fuels other than MOX fuel are under basic R&D. Nitride fuel is the leading candidate so far.

The PNC has two R&D centers. One is O-arai Engineering Center, where Joyo and postirradiation examination facilities are located. It will continue to play a principal role as an irradiation center for the development of LMFBR fuel in Japan. Postirradiation examination PIE facilities are extending their dimensional handling capability for large fuel subassembly application, including the Monju fuel. At Tokai works, where fuel fabrication facilities are located, R&D of plutonium fuel fabrication technology will be performed. Fuel development efforts for future commercialization will proceed by combining both centers' activities.

From the standpoint of efficient and steady LMFBR development in the world, international collaboration will be more important than ever. We would like to solidify collaborative relationships with foreign countries.

3. Status of LMR Fuel Development in the United States of America, *R. D. Leggett (Westinghouse Hanford), L. C. Walters (ANL-Idaho)*

The United States has pursued the development of multiple liquid-metal-cooled reactor (LMR) fuel systems in a deliberate and highly disciplined fashion with emphasis on fuel reliability and operational safety.[1] Three fuel systems have been shown to be reliable – oxide, metal, and carbide – and a fourth system, nitride, is the fuel of choice for the U.S. Space Power Program[2] and is under irradiation in the Fast Flux Test Facility (FFTF) and Experimental Breeder Reactor II (EBR-II). Oxide and metal are the respective driver fuels for the two U.S. fast flux test reactors, FFTF and EBR-II. Carbide was adopted as the driver fuel for the Indian fast test reactor.[3]

The development of mixed oxide (MOX) (UO_2-PuO_2) was the cornerstone of the U.S. program for more than 20 yr, but with the total success of the FFTF driver fuel, this system is generally recognized as being fully viable and U.S. development emphasis shifted to metal fuel in the mid-1980s. A concept called the integral fast reactor (IFR) concept emerged at Argonne National Laboratory[4] that utilizes metallic fuel.

The IFR concept encompasses the entire reactor system, not just the fuel itself. It includes the fuel cycle, both fuel refining and fabrication, and waste management as well. The goal is to develop a reactor system that answers the questions facing the next generation of nuclear power plants with respect to economics, safety, and waste disposal. The U-Pu-Zr system was selected as the IFR fuel alloy because it offered good breeding potential and superior performance characteristics over other metallic fuels.

Since 1985 (Ref. 5), nearly 14 000 IFR metallic fuel elements covering a range of fuel composition, design variations, and operating conditions have been irradiated in EBR-II and nearly another thousand in the FFTF. These experimental test elements include operation at 2σ operating temperatures (660°C), high fuel-smeared density (85 to 90%), and high fuel-to-plenum volume ratios (0.7), which severely challenge element reliability; however, in every case, the lifetime was greater than expected.

The EBR-II core has been converted to U-10 Zr with limited U-x Pu-10 Zr, where x varies between 8 and 28 wt% plu-

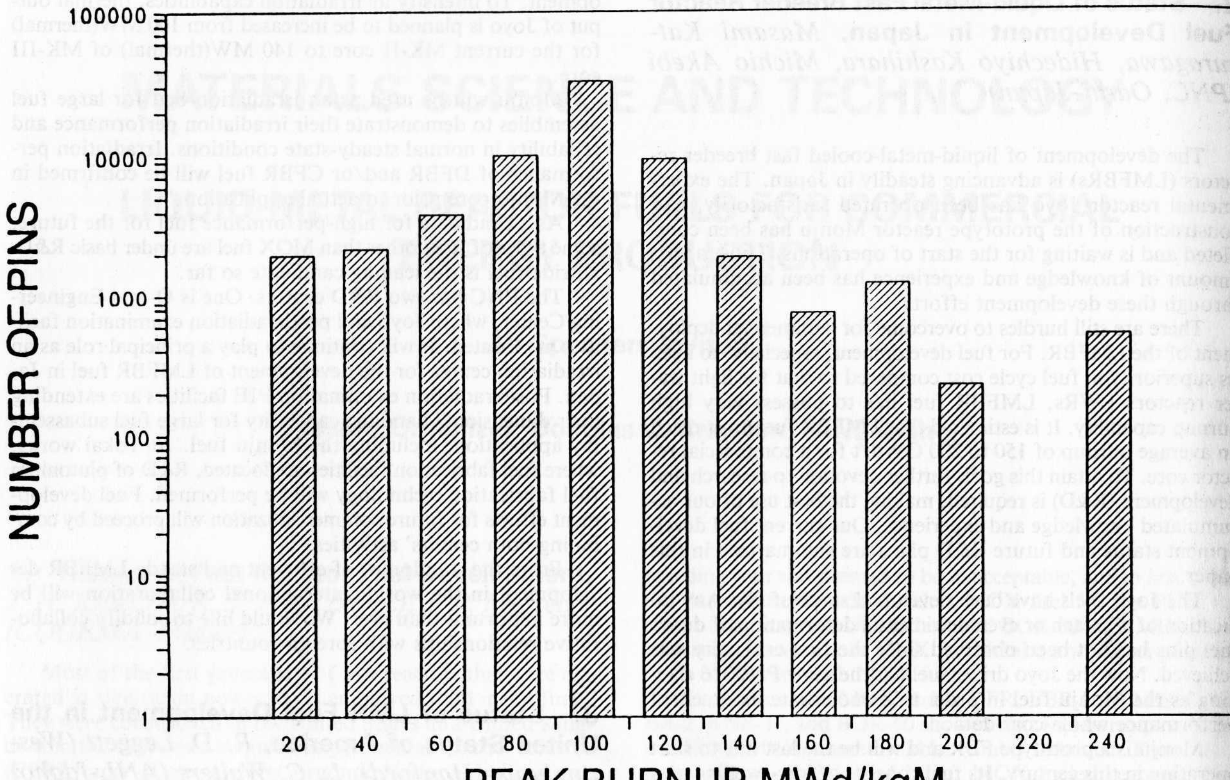

Fig. 1. Mixed-oxide fuel pins irradiated in the FFTF.

tonium to statistically demonstrate reliability and performance. Several experimental lead elements, including those clad with martensitic stainless steel, HT-9, have now reached 19.3 at.% burnup without breach, and no performance limitations have been identified. These elements were aggressively designed with relatively high linear power and low plenum-to-fuel ratios. Thus, much higher burnups are expected with a modest increase in the plenum-to-fuel ratio. The full size assemblies in the FFTF with HT-9 structural components have reached burnups of ~16 at.% with no sign of performance deterioration.

The high burnup capability of MOX fuel has been demonstrated by the irradiation of full-sized assemblies under prototypical conditions in the FFTF. The commercially produced FFTF driver fuel, employing 20% cold-worked Type 316 stainless steel cladding and ducts experienced no breaches below 10 at.% burnup, and selected assemblies reached 12 at.% before duct distortion forced their removal. Allowable exposures to 16 at.% were demonstrated with the lower swelling D-9 austenitic stainless steel cladding and ducts. Exposures to well over 20 at.% have now been demonstrated by utilizing the zero-swelling HT-9 for cladding and ducts. The number of pins compared with burnup irradiated in full-sized assemblies under prototypical conditions in FFTF is shown in Fig. 1.

The high burnup capability of MOX fuel using HT-9 components was demonstrated by an FFTF partial core loading known as the core demonstration experiment.[6] The burnup limits of the system have not yet been reached, but there are reasons to believe that burnups of 30 at.% will be readily achievable with this design with a small increase in plenum volume.

The only recent work on carbide fuel in the United States since that reported on in Tucson in 1986 has involved a joint U.S.-Swiss irradiation test in the FFTF (Ref. 7) that irradiated (UPu)C pellets and microspheres in D-9 cladding. The 91-pin assembly operated successfully at 92 kW/m peak power to a burnup of 85 MWd/kg M. Post-irradiation examination is incomplete, but all information obtained to date shows excellent performance.

The adoption by the Space Power Program of UN fuel and the recent world-wide interest in (U,Pu)N has sparked some limited evaluations in the United States of the potential for application of this fuel system to LMRs (Ref. 8). The earlier EBR-II program with mixed-nitride fuel[9] suggested a burnup potential of 20 at.% at very high (100 kW/m) linear heat ratings. Interestingly, nitride fuel exhibits many of the same desirable characteristics of metal fuel, i.e. high heavy-metal atom density, good thermal conductivity, and excellent compatibility with sodium. It has the added advantage of being compatible with existing fabrication and reprocessing methods established for oxide fuels.

Clearly, the United States is placing the bulk of its LMR developmental efforts on metallic fuel in support of the IFR program, and the success of these efforts is highly encouraging. The performance of the MOX system is well established, and this system can be adopted for future LMRs should the need arise. Meanwhile, developments elsewhere with carbides and nitrides are being closely monitored.

1. *Proc. Int. Conf. Reliable Fuels for Liquid Metal Reactors*, Tucson, Arizona, 1986, American Nuclear Society.

2. R. B. MATTHEWS, "Overview of SP-100 Fuel Development Activities," *Trans. 7th Symp. Space Nuclear Power Systems*, Albuquerque, New Mexico, 1990, CONF-900109.

3. S. B. BHOJE et al., "Commissioning of Fast Breeder Test Reactor," *Proc. Int. Conf. Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, October 28–November 1, 1991.

4. C. E. TILL, Y. I. CHANG, "Progress and Status of the Integral Fast Reactor (IFR) Fuel Cycle Development," *Proc. Int. Conf. Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, October 28–November 1, 1991.

5. R. G. PAHL, D. L. PORTER, D. G. CRAWFORD, L. C. WALTERS, "Irradiation Behavior of Metallic Fast Reactor Fuel," to be published in *J. Nucl. Mater.*

6. A. E. BRIDGES et al., "Advanced Liquid Metal Reactor Fuel and Blanket Design Using HT9," *Proc. Int. Conf. Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, October 28–November 1, 1990.

7. H. P. ALDER et al., "Status of Advanced LMR Fuel Development in Switzerland," **60**, 310 (1989).

8. W. F. LYON et al., "Performance Analysis of a Mixed Nitride Fuel System for an Advanced Liquid Metal Reactor, *Proc. Int. Conf. Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, October 28–November 1, 1991.

9. A. A. BAUER, P. CYBULSKIS, R. L. PETTY, N. S. DEMUTH, "Helium- and Sodium-Bonded Mixed-Nitride Fuel Performance," *Proc. Int. Conf. Fast Breeder Reactor Fuel Performance*, Monterey, California, 1979.

4. Status of LMR Fuel Development in Europe,

Günter Mühling (KfK–FRG), Christine Brown (UKAEA Dounreay–UK), Alain Languille (CEA/CEN Cadarache–France)

INTRODUCTION

This paper reviews the present status of coordinated European research and development programs where the main emphasis is on the behavior of mixed uranium-plutonium-oxide fuel pins for high burnup application. Only a smaller part is devoted to the ongoing development of mixed uranium-plutonium-nitride fuels.

FUEL BEHAVIOR

The existing status of knowledge and experience is based mainly on the results obtained from the irradiation of a large number of standard and experimental fuel assemblies in the European prototype reactors, the prototype fast reactor (PFR), Phénix, and KNK II.

More than 35 000 pins with mixed-oxide (MOX) fuel have reached burnup values of 130 MWd/kg, and ~2000 pins have exceeded 150 MWd/kg. In addition, some experimental pins irradiated in test clusters in the PFR have attained burnup levels >21%. In most cases, a homogeneous column of fuel pellets has been used, but high burnup values (14 and 18.4%) have also been achieved with axially heterogeneous fuel columns.[1]

The experience gained from these experiments has led to the conclusion that the behavior of the MOX fuel is not a life-limiting factor for the fuel pins even under stringent high burnup requirements.

Based on the excellent results obtained with the MOX fuels, it is currently believed that the targets for future large fast breeder reactors (200 MWd/kg) can be reached with this fuel type. This, together with the existence of large, industrial facilities for fuel fabrication and reprocessing, explains the fact that in the European Fast Reactor (EFR) Program, a fuel pin with MOX pellets has been defined as the reference design.

Activities in the field of mixed-nitride fuel have been devoted mainly to

1. fabrication of various fuel types
2. demonstration that high burnup is achievable with helium bonded fuel elements under convenient operational conditions.[2]

DEVELOPMENT OF ALLOYS FOR CLADDING AND WRAPPER

With regard to the development of cladding and wrapper materials, notable progress has also been made with materials accumulating damage doses approaching those set as targets for a large commercial liquid-metal-cooled reactor.

In the case of wrappers, attention is currently focused on fully martensitic ferritic steels, and two candidates are under consideration, a plain 9 Cr–1 Mo steel (EM 10) and a 10 to 12 Cr MoVNb (FV448/1.4914). The highest doses achieved so far are 132 displacements per atom (dpa) NRT for a FV448 wrapper in the PFR, 115 dpa NRT for 1.4914, and 146 dpa NRT for EM10 wrappers in Phénix.

The selection of these steels for wrapper applications was based on their excellent dimensional stability at high dose and their satisfactory mechanical properties after irradiation.

In the case of cladding materials, the European program concentrates on two types, a high nickel alloy STA PE16 and the improved austenitic material AIM1, which is closely related to 15.15 Ti/1.4970.

Nimonic PE16 was originally selected in the United Kingdom because of its high-temperature strength, and subsequent irradiations in both DFR and PFR showed an inherent resistance to void swelling. There have been no indications of any rapid rise in swelling rate at high dose, and diametral strains of ~2 to 3% are predicted at the EFR target dose of 200 dpa NRT. Postirradiation mechanical properties tests show that considerable strength is retained at high doses, but the ductility of the material decreases with increasing irradiation temperatures.[3]

The austenitic alloy was originally under development in both France and Germany. Recent developmental work has been carried out within the framework of the European collaboration, and major improvements have been achieved through optimization of composition and fabrication. At present, 15 Cr–15 Ni–TiMoB is the reference cladding material for the cores of SPX1, Phénix and KNK II. The large number of pins irradiated have emphasized the beneficial effect of an increase in cold working and the addition of some minor elements on swelling and irradiation creep behavior. Also, the mechanical properties of the material are influenced by irradiation and are dependent on temperature, dose, and stress; the results obtained, however, are encouraging and allow the assumption that the ambitious EFR targets can be reached.

BEHAVIOR UNDER OFF-NORMAL CONDITIONS

An experimental program was launched some years ago to determine the performance capability during steady-state irradiation combined with a range of operational and overpower transients that challenge the breaching thresholds of the pins. The main results have already been reported[4]; it was demonstrated that the pins survived a wide range of ramping and overpower events.

A disadvantage of all these experiments is that in the past, only low-burnup fuel pins could be tested; therefore, the program now concentrates on using high-burnup pins for the following experiments:

1. overpower tests in the High-Flux Reactor (HFR) with two PE16 and two 1.4970 pins, preirradiated in the PFR to 190 MWd/kg and to 90 MWd/kg in Phénix
2. RELIEF experiments, also in the HFR, to provide information on the effective differential axial expansion and reactivity changes under nearly loss-of-flow conditions
3. POTOM experiments in the HFR to study the power-to-melt behavior of homogeneous and heterogeneous fuel pins
4. thermal calibration tests in CABRI, where high-burnup pins of various designs are ramped.

FAILED PIN BEHAVIOR

The behavior of subassemblies containing failed pins is of great interest with respect to reactor operation, element handling, and intermediate storage. Hence, a substantial program of work in the field has been carried out, partly in the thermal reactor SILOE, where the specific instrumentation allowed a high flexibility in selecting the experimental parameters, and in the running reactors PFR, Phénix, and KNK II, where mainly processes of failure detection and localization were studied.

At present, the experimental program concentrates on two items:

1. behavior of failed breeder pins (the new experiments are carried out at SILOE)
2. behavior of failed fuel pins under low-power conditions (simulation of the intermediate in-pile storage).

1. J. C. MELIS et al., "Fuel Modeling at High Burnup: Recent Development of the GERMINAL Code," *Trans. Am. Nucl. Soc.*, **66**, 223 (1992).

2. C. PRUNIER et al., "Flow Sheet, Fabrication, and Properties Comparison of Nitride Fuel Obtained by Powder Metallurgy and Gelation Processes," *Trans. Am. Nucl. Soc.*, **66**, 187 (1992).

3. C. BROWN et al., "Cladding and Wrapper Development for Fast Breeder Reactor High Performance," *Proc. Int. Conf. Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, October 28–November 1, 1990.

4. H. PLITZ et al., "New Experiments to Assess Fuel Pin Transient Operation," *Proc. Int. Conf. Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, October 28–November 1, 1990.

Panel Discussion