"NUCLEAR FUEL CONSIDERATIONS FOR THE 21ST CENTURY"

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NUCLEAR FUEL CONSIDERATIONS FOR THE 21ST CENTURY

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ABSTRACT

There are many external influences that may control the path that nuclear power deployment follows. In the next 50 years several events may unfold. Fear of the consequences of the greenhouse effect may produce a carbon tax that would make nuclear power economically superior very quickly. This, in turn, would increase the rate at which uranium reserves diminish due to the increased rate of nuclear power deployment. However, breakthroughs in the extraction of uranium from the sea or deployment of fast breeder reactors would greatly extend the uranium reserves and, as well, utilize the thorium cycle.

On the other hand, carbon sequestering technology breakthroughs could keep fossil fuels dominant for the remainder of the century. Nuclear power may only then continue, as today, in a lesser role or even diminish. Fusion power or new developments in solar power could completely displace nuclear power as we know it today.

Even more difficult to predict is when the demand for mobile fuel for transportation will develop such that hydrogen and hydrogen rich fuel cells will be in common use. When this happens, nuclear power may be the energy source of choice to produce this fuel from water or methane. In a similar vein, the demand for potable and irrigation water may be another driver for the advent of increased deployment of nuclear power.

With all these possibilities of events that could happen it appears impossible to predict with any certainty which path nuclear power deployment may take. However, it is necessary to define a strategy that is flexible enough to insure that when a technology is needed, it is ready to be deployed.

For the next few decades there will be an evolutionary improvement in the performance of uranium oxide and mixed uranium oxide-plutonium oxide (MOX) LWR fuels. These improvements will be market driven to keep the cost of fuel and the resulting cost of nuclear power electricity as competitive as possible. The development of fuels for accelerator transmutation and for reactor transmutation with inert matrix fuels is in its infancy. A great deal of research has been initiated in a number of countries, which has been summarized in recent conferences. In Europe the work on these fuels is directed at the same problem as their utilization of MOX; namely to reduce the inventory of separated plutonium, minor actinides, and Long Lived Fission Products (LLFP). In the United States there is no reprocessing and thus no inventory of separated civilian plutonium. However, in the United States there is a resistance to a permanent spent fuel repository and thus accelerator transmutation presents a possible alternative. If nuclear power does have a long-term future, then the introduction of the fast reactor is inevitable. Included in the mission of the fast reactors would be the elimination of the

inventory of separated plutonium while generating useful energy. The work that is ongoing now on the development of fuel concepts for assemblies that contain actinides and LLFP would be useful for fast reactor transmutation.

There is still a great deal of work required to bring the fast breeder reactor option to maturity. Fortunately there is perhaps a fifty-year period to accomplish this work before fast breeders are necessary. With regard to fast reactor fuel development, future work should be considered in three stages. First, all the information obtained over the past forty years of fast reactor fuel development should be completely documented in a manner that future generations can readily retrieve and utilize the information. Fast reactor development came to such an abrupt halt world-wide that a great deal of information is in danger of being lost because most of the researchers and facilities are rapidly disappearing. Secondly, for all of the existing fast reactor fuels, and this includes, oxides, carbides, nitrides, and metallic fuels, the evolutionary work was far from being completed. Although mixed oxide fuels were probably the furthest advanced, there were many concepts for improved claddings and advanced fabrication methods that were never fully explored. Finally, with such an extended period before fast reactors are needed there is ample time for truly innovative fuels to be developed that are capable of performing over a wide range of conditions and coolants.

Introduction

Nuclear fuel for advanced fuel cycles is dependent on the possible paths that nuclear power development may take in the future. There are many external influences that may control the path that nuclear power deployment follows. Most authors who analyze deployment alternatives speak in 50-year intervals, which is somewhat arbitrary, but 50 years is in the range of perfecting a new technology from concept to realization.

In the next 50 years several events may unfold. Fear of the consequences of the greenhouse effect may produce a carbon tax that would make nuclear power economically superior very quickly. This, in turn, would increase the rate at which uranium reserves diminish due to the increased rate of nuclear power deployment. However, breakthroughs in the extraction of uranium from the sea or deployment of fast breeder reactors would greatly extend the uranium reserves. Development and implementation of a thorium fuel cycle would also greatly extend the nuclear power enterprise.

On the other hand, carbon sequestering technology breakthroughs could keep fossil fuels dominant for the remainder of the century. Nuclear power may only then continue, as today, in a lesser role or even diminish. Fusion power or new developments in solar power could completely displace nuclear power as we know it today.

Hydrogen and hydrogen-rich fuels have been proposed as alternatives to fossil based fuel. However, it is difficult to predict when the demand for mobile fuel for transportation will develop such that hydrogen and hydrogen rich fuel cells might be in common use. When this happens, nuclear power may be the energy source of choice to produce this fuel from water or methane, published (IAEA-TECDOC-1085, 1999). In a similar vein, the demand for potable and irrigation water may be another driver for the advent of increased deployment of nuclear power.

With all these possibilities of events that could happen it appears impossible to predict with any certainty which path nuclear power deployment may take. Adding to the uncertainty are the varying energy options, and political acceptability of some options, in different nations. In spite of these uncertainties, prudence demands that we define an energy strategy that is flexible enough to insure that when a technology is needed, it is ready to be deployed.

Future Nuclear Fuel Development

For the next few decades there will be an evolutionary improvement in the performance of uranium oxide and mixed uranium oxide-plutonium oxide (MOX) LWR fuels. These improvements will be market driven to keep the cost of fuel and the resulting cost of nuclear power electricity as competitive as possible. It is not clear at this time whether the MOX option for LWR utilization will continue. MOX utilization in Europe is a means to reduce the civilian plutonium inventory that results from the reprocessing of LWR fuels. This separated plutonium was originally destined for fast reactors, the construction of which had been anticipated. MOX utilization in the United States is simply a means to dispose of excess weapons plutonium. The economics of MOX utilization has recently been questioned for a number of reasons, published (Charpin *et al.*, 2000). In any event, the evolutionary improvements of LWR fuels will most certainly take place over the next few decades as the commercial enterprise competes for the market in both the developed and developing countries.

The development of fuels for accelerator transmutation and for reactor transmutation with inert matrix fuels is in its infancy. A broad range of smaller research effort has been initiated in a number of countries, which has been summarized in recent conferences, published (Proceedings from Global '99, 1999; Proceedings from Workshop on Advanced Reactor and Innovative Fuels, 1998; Proceedings of ICONE 8, 2000). In Europe the work on these fuels is directed at the same problem as utilization of MOX; namely, reduction of the inventory of separated plutonium, minor actinides, and Long Lived Fission Products (LLFP). The problem in Europe is envisioned to be near-term because the growing inventory of separated plutonium presents a potential proliferation and radio-toxicity problem. In the United States there is no reprocessing and thus no inventory of separated civilian plutonium. However, in the United States there remains some question regarding the licensability of a permanent spent fuel repository and thus accelerator-driven transmutation presents a means to reduce or eliminate quantities of long-lived actinides and fission products..

All of this development on fuels for actinide and waste transmutation would be completely unnecessary with the near term reintroduction of fast reactors. Maturation of these transmutation concepts, such that they could significantly begin to reduce existing inventories of actinides and LLFP, would require at least 30 years. During that period of time the long-term future of nuclear power should become evident. If a long-term future does not develop, and if reprocessing continues for the next fifty years, then a huge inventory of separated plutonium, minor actinides, and LLFP will exist that must be subjected to transmutation or immobilized for a repository. In this situation the availability of proven fuel designs for actinide and waste transmutation is crucial. Moreover, if nuclear power does have a long term future, then the introduction of the fast reactor is inevitable, published (Hori, 2000). Included in the mission of the fast reactors would be the elimination of the inventory of separated plutonium while generating useful energy. The work that is ongoing now on the development of fuel concepts for assemblies that contain actinides and LLFP would be useful for fast reactor transmutation.

A great amount of work remains to bring the fast breeder reactor option to maturity. Fortunately there is perhaps a fifty-year period to accomplish this work before fast breeders are necessary. With regard to fast reactor fuel development, future work should be considered in three stages. First, all the information obtained over the past forty years of fast reactor fuel development should be completely documented in a manner that future generations can readily retrieve and utilize the information. Fast reactor development came to such an abrupt halt world-wide that a great deal of information is in danger of being lost because most of the researchers and facilities are rapidly disappearing, published (33rd IAEA International Working Group Meeting on Fast Reactor, 2000). Secondly, for all of the existing fast reactor fuels, and this includes, oxides, carbides, nitrides, and metallic fuels, the evolutionary work which was far from being completed, should be continued. Although mixed oxide fuels were probably the furthest advanced, there were many concepts for improved claddings and advanced fabrication methods that were never fully explored, published (Nuclear Materials, 1994). Finally, development of innovative fuels should be pursued. With such an extended period before fast reactors are needed, there is ample time for truly innovative fuels to be developed that are capable of performing over a wide range of conditions and coolants. The next sections will discuss the current situation and what might be accomplished in each of the three stages.

Preservation of Fast Reactor Fuel Information

Fast reactor fuel development programs were at their peaks by 1980. Fast test reactors were operating in several countries with commercial size prototypes just under construction or coming on line. From that time onward, fast reactor development in general began to decline. By 1994 in the United States the Clinch River Breeder Reactor (CRBR) had been canceled and the two fast reactor test facilities, the Fast Flux Test Facility (FFTF) and the Experimental Breeder Reactor (EBR-II) had been shutdown; with EBR-II permanently and FFTF in a standby condition. Thus, efforts essentially disappeared for fast reactor fuel development. Similarly, programs in other nations were terminated or substantially reduced.

Not quite so obvious is the disappearance of the aging fast reactor fuel experts. With their continuing retirement from the workforce a great deal of knowledge and experience is being lost. It may be argued that all the fuel performance information gained during this period has been well documented in the open literature, conference proceedings, and company reports. In addition, there have been recent fast reactor fuel reviews. For fuel irradiation performance information this may well be the case. However, a serious effort has not been undertaken, at least in the United States, to collect and review for completeness the existing fuel performance documentation. The degree to which the information has been compiled for posterity is probably far less in the areas of fuel fabrication technology, the design and performance of in-reactor test vehicles, and the operating performance of entire cores of fuel, blanket, and reflector assemblies. Again, no assessment has been made of either the value of this information to future generations nor an assessment of the completeness of the documentation. Even now an effort to correct any deficiencies would be difficult due to the loss of the experts. Soon it will be impossible.

Based upon the author's experience with the Experimental Breeder Reactor II, EBR-II, there are several areas that need a significant effort in order to bring the more than 30 years of operating experience to a state where future generations will receive full benefit. Table 1 lists a number of areas that need to be archived in a much better manner than they are today. To a greater or lesser extent the situation is likely the same world-wide. It is simply a matter of completing the 30-year task with a small fraction of what has already been spent to allow society to realize the benefit of a generation of accomplishment.

Table 1 - Areas to Assess for Archival Completeness

Reactor Design and Construction	<ul style="list-style-type: none">• Rational for key design features.• Original design drawings and justification for major changes.• Specifications for materials and procurement considerations.• Organization structure and evolution.• Documentation of important problems and solutions.• State of archival materials.
Operations	<ul style="list-style-type: none">• Operations organization.• Training and procedures.• Safety analysis reports and system design descriptions.• Fuel handling experience.• Steam system operating experience.• Documentation of important problems and solutions.

Nuclear Physics and Thermal Hydraulics	<ul style="list-style-type: none"> • Documentation of all computer codes along with utilization experience. • Documentation of special physics and thermal hydraulic experiments. • Documentation and discussion of significant safety-related events.
Fuel Fabrication	<ul style="list-style-type: none"> • Fabrication organization. • Training and procedures. • Fuel specifications. • Equipment design drawings. • Materials specifications and procurement. • Fabrication experience. • Documentation of important problems and solutions.
Fuel Performance	<ul style="list-style-type: none"> • Irradiation test results. • Documentation of computer codes. • Documentation of the analyses of important phenomena. • Description and design drawings of in-reactor test vehicles. • Documentation of important problems and solutions.

Evolutionary Improvements to Fast Reactor Fuels

As is the case for almost all products of industry, evolutionary improvements occur to make the products safer and less expensive. Fast reactor fuel development had by no means reached any sort of asymptote for ultimate performance when work in the United States was terminated. The same situation is true abroad. The problems of excessive irradiation-induced creep and swelling in fast reactor fuel cladding was well on the way to solution with the introduction of martensitic/ferritic steel cladding. However, the low swelling cladding materials had marginal high-temperature strength, so cladding improvements were desired to achieve high outlet temperatures. Although mixed oxide pellet fuel for fast reactors was the most advanced, the data base was not adequate in the United States for licensing a commercial reactor. For metallic, carbide and nitride fast reactor fuels a great deal more performance information was required before licensing could be attempted and, as well, there were many evolutionary concepts that were yet unexplored.

An international assessment should be made of all the evolutionary concepts for fast reactor fuels, and with some selectivity, the evolutionary work should move forward. However, two obstacles must be overcome. The first is a new generation of scientists and engineers must be attracted into this area to provide a continuity of expertise. Secondly, there must be fast reactor facilities to conduct the irradiation and post-irradiation examinations. Currently the United States has no fast reactor irradiation facilities, and there is a struggle to keep the hot cell capability viable. The same is true in Great Britain. France with Phenix, Japan with Joyo and Monju, and Russia with BOR-60 and BN-600 still have irradiation capability. However, the near-term future with all those reactors is uncertain.

It will be a difficult task to convince governments around the world to continue with fast reactor fuel development at some nominal level when there is no near-term foreseeable need for fast reactors while, at the same time, there is pressure from the antinuclear community to cease all fast reactor work. However, the alternative is the complete loss of facilities, expertise, and perhaps information already gained, the replacement of which will be extremely costly a few decades from now.

As pointed out earlier, a major obstacle to the development of a new fuel or the advancement of evolutionary ideas is the loss of irradiation test and examination facilities. A solution, which to some extent is already happening, is to identify the best remaining facilities, on an international basis, and devise a strategy to share in the utilization and cost of operating the facilities. There will not be a large fuel development program conducted over a relatively short period of time such as the world experienced during the 1970's for fast reactor fuel development. During that period several countries had the complete suite of facilities that included critical facilities, transient fast reactors, fast reactors for steady-state irradiations, and extensive examination facilities. Rather than the large, short-term development programs, much lower level programs will continue in each involved country in a search for the system that will replace the LWR's several decades in the future. Within this framework, a sharing of remaining facilities with minimum duplication makes the best solution. For example, fast reactor irradiation facilities such as Joyo in Japan or BOR-60 in Russia might be used in conjunction with transient test facilities such as TREAT in the U.S. to develop new fuels and to demonstrate irradiation performance under a full range of conditions. Irradiation testing programs, then, would become truly international collaborations.

An example, that falls within the experience of the authors of an evolutionary fast reactor fuel program that was not carried to completion, is that for Integral Fast Reactor (IFR) metallic fast fuel, published (Hannum, 1997). When the development program for the U-Pu-Zr metallic fuel was terminated in the United States in 1994, the fuel had been irradiated to 20 at.% burnup without failure, published (Walters, 1998). Fuel with improved cladding materials and lower smeared density was under irradiation that promised substantially greater burnup than 20 at.%. In addition, fuel with high concentrations of minor actinides was being irradiated to learn more about the fabrication and performance of fuel for minor actinide disposal.

The IFR concept was included in the United States Energy Policy Act of 1992, where a schedule was prescribed for design and construction of a prototype plant (PRISM-type). Title II design was to be initiated for the prototype ALMR in 1998. In response to the Policy Act, GE Nuclear Energy produced (early in 1993) the ALMR Fuel and Safety R&D Requirements document which outlined the issues to be addressed in order to document ALMR fuel performance sufficiently so that an ALMR could be licenced. This document was reviewed by the Nuclear Regulatory Commission against the IFR program documentation and the NRC produced NUREG-1368, published (NUREG-1368, 1994).

The issues that were raised were addressed in the Fuel Test Plan (authored by General Electric, Westinghouse, and Argonne National Laboratory) in 1994 with either technical justification or plans for additional R&D, published (Porter, 1994). Interestingly, these beginnings of the licensing process were underway just as the IFR program funding was terminated in 1994.

The Fuel Test Plan showed that U-Pu-Zr metallic driver fuel and U-Zr blanket fuel performance was known, documented, and modeled sufficiently in many areas. The fundamental areas included swelling, constituent (elemental) migration, fuel pin length changes, and fission gas release, in regards to steady-state performance. Fuel/cladding chemical compatibility and 'eutectic limits' were included in transient performance issues as well as post-failure performance (run beyond cladding breach). Proof of statistical reliability of the fuel, cladding and duct performance, and the accuracy of fuel performance modeling were also examined and the status addressed. The performance differences caused by minor actinide (Am, Np) also needed additional work based upon experiments which had been completed. The issues that remained are outlined in Table 2 below.

Table 2 demonstrates that the fuel performance was nearly complete; schedules shown with this information in the Test Plan indicated that 2-3 years were required to complete the requirements for cold-fabricated fuel, and 4-5 years for the Am, Np-bearing fuel. Note, however, that several open issues required the study of as-irradiated fuel from experiments; this fuel is no longer available. Therefore, if started now, it would require much longer times to get to the point of being ready for licensing.

Table 2

Fuel Performance Issue	Remaining Questions/Actions
Fuel Swelling	Complete exams for porosity distributions/Final Model Validation.
Constituent Migration	U-Zr complete/Complete model validation for U-Pu-Zr.
Effects of Minor Actinides	'Eutectic' temperature effects.
Fuel/Cladding Compatibility	Complete tests of high Pu (26-28 wt%) alloys with HT9 cladding at high fuel burnup/complete ex-reactor tests to be able model effects of lanthanide fission products.
Fission Gas Release	Complete
Fuel Pin Length	Complete
Transient Performance	One test at >40% overpower; steady-state irradiation after overpower tests.
Run Beyond Cladding Breach	Testing complete; complete modeling of gas release signatures, etc., for various breach locations and fuel types.
LIFE-METAL and F-PIN code development for element reliability	Complete model validation for fuels containing Am and Np; Use validated codes to run reliability analyses for prototype ALMR.
Fuel Specification	In-reactor performance-based specification complete/fuel recycle demonstration to test acceptability in remote manufacturing process.
Cladding and Duct Performance	Complete

Innovative Fast Reactor Fuels

As discussed earlier it is impossible to predict future demands on nuclear power when all the external forces are considered. However, up to this time, nuclear power has been used primarily for electricity generation. There are some desalination applications for nuclear power, such as the former operation of the BN-350 fast reactor in Kazakhstan. Breeding potential and the use of the higher temperature liquid metal coolants created additional challenges for the nuclear fuel developer. In the future, the need to produce H_2 by the high temperature cracking of water, compact-high energy density cores for space travel, and perhaps the wide use of nuclear power for desalinated water will further challenge the fuel designer. Concepts have been put forward for reactors that could serve these multiple uses, published (Wade, 2000). Perhaps by the end of the century nuclear parks will exist that produce electricity, breed Pu^{239} and U^{235} for further power production, H_2 by electrolysis or some other process, and heat for other uses such as water purification.

Because of the long period, 30-50 years, to bring a truly innovative concept to fruition, it is the time to begin this advanced thinking process. It would also keep a new generation of nuclear scientists challenged.

However, as pointed out earlier, the larger challenge is to convince short-sighted governments that such work is needed.

Many of the fuel systems that the authors regard as "innovative" are, in fact, fuel types for which there is little or no data regarding performance, fabricability or properties. Other fuels regarded as innovative are actually evolutions of previously investigated fuel types now under consideration for new and innovative applications. Some of the requirements now emerging for new fuels are common to many applications, such as high-temperature or extended-burnup operation, while other requirements are particular to an application, such as fast transmutation rates. Some of these requirements are addressed in the following paragraphs, which also describe the fuel types being considered or proposed.

Nearly all reactor or transmutation concepts being proposed anticipate high-burnup use of fuel. For some concepts, such as those with long-life cores designed for only occasional fuel handling, extended burnup capability is essential. For other concepts, extended burnup is highly desirable to increase proliferation resistance of spent fuel with relatively high contents of higher actinides. And for many instances, extended burnup is regarded, as it has been historically, as desirable to improve economics of reactor operation by reducing the number of fuel assemblies required to produce a obtain amount of nuclear energy, with associated costs reductions for fuel handling, fabrication, recycling, and/or post-irradiation storage. Some of the design features being proposed for achieving high burnup include dispersion and vi-pac fuel forms as well as new cladding materials that are corrosion resistant or that have improved creep strength.

Some of the innovative reactor concepts being considered, or simply discussed, would employ relatively high coolant outlet temperatures to increase energy conversion efficiency or to provide heat for high-temperature processes (e.g., certain hydrogen production techniques). Accommodation of higher coolant outlet temperatures would require fuel and cladding to operate at higher temperature. For fast reactor fuel, this is a challenge to cladding performance as plenum pressures increase due to fission gas release, inducing irradiation creep deformation. Fuel materials that retain configuration under irradiation at high temperature and cladding materials with improved creep resistance will be sought. Nitride fuels are being re-considered, particularly for high-temperature applications for which metal fuels seem ill suited.

Transmutation of plutonium and higher actinides is accomplished at faster rates in systems with little or no fertile material (i.e., with no U-238). This has generated interest in non-fertile fuels, published (United States Department of Energy, 1999). There is little experience with non-fertile fuels for fast reactor application, although some efforts are underway in Europe, published (Piccard, et al., 2000), Japan, and the U.S. Effort in the U.S. is specifically directed at fuel forms for the Accelerator Transmutation of Waste concept, which will consider Zr-matrix dispersion and zirconium-actinide nitride forms. Requirements for non-fertile fuel forms are typically the same as other fast reactor fuels, but with less expectation for high-burnup or high-temperature operation.

Conclusions

There is a great deal of uncertainty regarding the future utilization of nuclear power. A popular and expanding school of thought is that the share of the world's total energy from nuclear power will increase during this century and not only for the production of electricity. If this is the case, then a strategy must be defined that includes deployment of fast reactors by mid-century.

Beginning soon there should be a concerted world-wide effort to preserve the wealth of fast reactor information while the last nucleus of researchers is still professionally active. At the same time, on a selective basis that is commensurate with available funding, a cooperative international effort should be undertaken to complete evolutionary work on existing and promising fast reactor fuel systems. This evolutionary work will require the sharing of remaining test and examination facilities. Finally, there is time to explore truly

innovative fuel systems that could bring advances in safety, economics, minimization of waste, and proliferation resistance. Emerging requirements for fuel systems other than for electricity generation may lead to the innovative development of high temperature fuel and coolant systems. However, it must be appreciated that the time period from concept to a licensed fuel system is on the order of 30 years.

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