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University of California

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545

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# Feasibility of Zircaloy as a Fuel Clad and Structural Material in an LAFR-LWR

Don M. Parkin

# FEASIBILITY OF ZIRCALOY AS A FUEL CLAD AND STRUCTURAL MATERIAL IN AN LAFR-LWR

by

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## ABSTRACT

A preliminary analysis of the radiation environment in a Linear Accelerator Fuel Regenerator (LAFR) has been made. The response of Zircaloy to this environment in combination with that in a Light Water Reactor (LWR) has been projected. It is concluded that the response to irradiation of Zircaloy in a combined LAFR-LWR cycle will lead to a more deleterious change of properties than for an equivalent LWR exposure.

## I. INTRODUCTION

The objectives of this preliminary analysis of the radiation environment that Zircaloy -clad fuel would experience in a Linear Accelerator Fuel Regenerator-Light Water Reactor (LAFR-LWR) cycle are to identify potential radiation damage effects that lead to a question of concept feasibility and to suggest research that would address such questions.

There are critical materials needs associated with many of the subsystems of the LAFR which will have to be identified and addressed in the conceptual development stage. The use of prefabricated LWR or Pressurized Water Reactor (PWR) fuel elements is the fundamental basis for the proposed system. Hence, the projected performance of fuel cladding and structural elements in the LAFR-LWR is one of the most important feasibility questions.

The LAFR-LWR system assumed in the analysis is one that consists of an initial fuel enrichment in an LAFR, followed by an LWR fuel burn and a second LAFR fuel enrichment with a final LWR fuel burn. The fuel will experience four separate irradiations and four "hot" fuel handling stages. This fact is important since it may have an impact on the mechanical properties the fuel assemblies must have at each stage of life.

The radiation environments in the two stages of the fuel cycle are quite different. A great deal of experience with thermal reactor irradiations has shown that Zircaloy performs well as a fuel cladding. Even under these irradiations, it is the fission or fast component of the neutron spectrum that dominates in producing the radiation damage. Because of this fact, the radiation effects data on Zircaloy represent fast-neutron results, which is an important observation for this application.

The LAFR radiation environment consists of two major components, neutrons and protons, which must be considered in the analysis. The leakage neutron spectrum from the target will look like a degraded fission spectrum quite similar to that in a fast reactor but with a high-energy tail extending up to the energy of the proton beam. Most of the neutrons and, as a result, most of the displacement-type damage will come from neutrons with energy below 20 MeV. The importance of the neutron flux component above ≈10 MeV is its potential in producing helium and hydrogen impurities in the structural alloys and new impurity products in the fuel itself. Further, unlike the evaporation or fission-like part of the spectrum, which has a more or less isotropic source distribution, the high-energy component will be more forward peaked with respect to the momentum of the proton beam. Potentially, this inhomogeneity in the spectrum field may have an impact on the radiation damage problem.

The neutron target is also a source of protons originating from similar nuclear processes. The leakage proton flux and energy distribution are strongly influenced by the target geometry. The leakage proton flux spectrum will have a major impact on the radiation damage environment. In this case, as with the neutron spectrum, the low-energy component ($E < 20$ MeV) and the high-energy component ($E > 20$ MeV) present different radiation damage problems. The low-energy protons will stop in a small amount of material (~0.1 cm), whereas the high-energy protons will penetrate long distances (~10-20 cm). The high-energy proton flux will be forward peaked, and coupled with its limited range, it will be a nonuniform source of damage.

This brief and sketchy description of the radiation environment includes the important components that have to be analyzed in order to determine the feasibility of using fabricated fuel in the LAFR-LWR cycle. To describe the estimated response of Zircaloy to the LAFR environment and how it relates to reactor conditions, it is useful to define and calculate a number of radiation damage parameters and to present assumptions used in obtaining flux and spectral estimates.

A relationship between damage exposure in one radiation environment to that in another is required so that relative damage levels can be estimated. Two parameters of interest are displacement damage and gas production. Although the effects of displacements and gas production on damage are coupled, they will be described separately. The best way to describe displacement damage is to calculate a spectrum-averaged displacement cross section for the material and irradiating particle of interest. This cross section incorporates the energy spectrum of the irradiating particle field so that displacement levels from different irradiations can be compared by a single exposure index. Specifically, proton and neutron irradiations can be directly compared. For the present analysis we have assumed based on Sabol's review,[1] that for LWR conditions

$$1.5 \times 10^{21}\ \text{n/cm}^2\ (E > 1.0\ \text{MeV}) \approx 12\ 000\ \text{MWD/MT} \approx 1.9\ \text{dpa}, \tag{1}$$

where the damage is expressed in units of displacements per atom (dpa). Since in the reactor, the fuel burnup can be related to the fluence via a fission cross section, we can also relate fuel burnup to dpa.

The displacement cross section for the leakage neutron spectrum has been estimated by calculating this cross section using two spectra obtained for the neutron radiation effects facility at Clinton P. Anderson Meson Physics Facility (LAMPF). The spectra were calculated for positions at 90° and 135° with respect to the center of mass of neutron production. Using ENDF/B nuclear data for Zircaloy-2 the results are

$$\sigma_{dpa}(135°) = 0.94 \times 10^{-21}\ \text{cm}^2$$

and

$$\sigma_{dpa}(90°) = 1.14 \times 10^{-21}\ \text{cm}^2 \tag{2}$$

using $E_d = 25$ eV after Carpenter.[2] For the present calculations we assumed

$$\sigma_{dpa}(\text{LAFR}) = 1 \times 10^{-21}\ \text{cm}^2. \tag{3}$$

It is interesting to note that using Eqs. (1) and (3) we find that

$$\sigma_{dpa}(\mathrm{LAFR}) \approx \sigma_{dpa}(\mathrm{LWR}).$$

In a similar way the gas production cross sections were calculated and for the analysis assumed to be

$$\sigma_{n,p}(\mathrm{LAFR}) = 2 \times 10^{-27}\ \mathrm{cm}^2$$

and

$$\sigma_{n,\alpha}(\mathrm{LAFR}) = 2 \times 10^{-28}\ \mathrm{cm}^2. \qquad (4)$$

For the LAFR radiation field a number of assumptions had to be made. Neutron flux and fluence parameters were obtained from P. Grand,[3] properties of the leakage proton flux spectrum were obtained from Weapons Neutron Research Facility(WNR) calculations by G. Russell[4] and proton gas production cross sections were estimated from calculations by Coulter et al.[5] The following estimates were used.

| | |
|---|---|
| LAFR neutron flux | $10^{14}$ n/cm$^2$s |
| Enrichment Dose | $2.5 \times 10^{21}$ n/cm$^2$ |
| Low-energy ($E < 20$ MeV) proton flux | $10^{13}$ p/cm$^2$s |
| High-energy ($E > 20$ MeV) proton flux | $3 \times 10^{12}$ p/cm$^2$s |
| High-energy proton hydrogen production | $8 \times 10^{-25}$ cm$^2$ |
| High-energy proton helium production | $1 \times 10^{-25}$ cm$^2$ |

## II. ANALYSIS

In analyzing the response of Zircaloy to the radiation conditions expected for the LAFR-LWR cycle,it is important to review three facts derived from the performance of Zircaloy in water reactors. One basis for projecting a long life for Zircaloy cladding is that the radiation-induced changes in strength and ductility tend to saturate at fluences of $1 - 2 \times 10^{21}$ n/cm$^2$ (~12 000 MWD/MT), which is early in the service life of the fuel. Zircaloy picks up hydrogen by corrosion processes and should have a hydrogen concentration less than 200 - 500 ppm.

Finally, and perhaps most importantly, Zircaloy cladding in reactor service does not form voids. This critical observation is most likely related to the fact that large dislocation networks are not observed and that the amount of helium generation is low (~0.3 ppm at $10^{22}$ n/cm$^2$). Since extended life (~100 000 MWD/MT) performance of the fuel is required for the proposed LAFR-LWR cycle, any change in the above conditions of cladding response to irradiation must be considered a question of concept feasibility. The following analysis concentrates on how the LAFR radiation environment may affect these three critical properties of Zircaloy.

## A. Effects Due to the LAFR Neutron Spectrum

The displacement damage level that will be reached by the cladding will be a result of two LAFR enrichment exposures and two fuel burns in a reactor. Each LAFR irradiation will produce an exposure of 2.5 dpa. At a burnup of 30 000 MWD/MT the reactor exposure will be 4.7 dpa. The total exposure disregarding proton damage will be

Total Cladding Exposure = 14.4 dpa ,

which is equivalent to 97 000 MWD/MT fuel burnup. This is roughly a factor of 2 higher exposure than experienced by high-burnup fuels.

Although direct comparisons between dpa levels in one material to that in another are not valid due to the fact that the damage potential of a dpa is strongly material dependent, it is instructive to note that an exposure of 14.4 dpa is considerably less than that required for stainless steel in a fast reactor. In fast reactors exposure limits are in the range 50-100 dpa. From the calculated exposure level and its relationship to present exposures in water reactors and exposure of stainless steel in fast reactors, there is nothing to suggest on the basis of displacement damage alone that there will be a major change in the response of Zircaloy to the radiation environment in the LAFR-water reactor cycle as compared to that in a water reactor alone.

The presence of the high-energy neutron component produces a major change in the production cross sections for helium and hydrogen gas. It has been shown that helium can play an important role in void formation in Zircaloy.[2] The potential effects of hydrogen in Zircaloy are clear. The amount of hydrogen generated in two LAFR irradiations will be about 10 ppm. This amount of hydrogen is considerably less than what is picked up through corrosion. The helium generation will be about 1 ppm for the two LAFR cycles. During a typical fuel burn in a water reactor about 0.1 ppm is produced. The helium produced in the LAFR does not appear to present a major problem. However, since the specific role that helium may play in void formation in Zircaloy has not been identified, the presence of higher helium concentrations, cycled irradiation, and higher exposures may result in deleterious synergetic effects that cannot be predicted now.

## B. Effects Due to the LAFR Proton Spectrum

In this analysis the displacement effects of protons have been ignored. In a more complete analysis, where more details of the proton flux and spectrum are known, proton displacement effects should be included. It is expected that they will contribute, on the average, at most a 10% addition to the neutron displacement damage.

Because of their short range (~0.1 cm), the low-energy proton flux presents a serious damage problem. To illustrate this fact, we assume that the protons stop uniformly over a distance of 0.1 cm in the Zircaloy pressure tube surrounding the fuel assembly. With the stated assumptions, the low-energy proton flux would produce a concentration of ~400 ppm hydrogen/day of LAFR irradiation in this layer. Even if the low-energy proton flux is 1% of the estimate, the implanted hydrogen is not tolerable. It appears unavoidable to conclude that a low-energy proton filter must be provided to eliminate this unacceptable problem.

Assuming that there is a low-energy proton filter, the high-energy proton flux still represents an important radiation flux. Its penetration range of many centimeters makes it reasonable for the present purposes to treat it in the same way as the neutron flux. The high-energy proton flux will produce a hydrogen concentration of about 60 ppm via (p, xp) reactors during one LAFR irradiation. This hydrogen level is comparable to that picked up from corrosion in one reactor fuel burn. Assuming that the same amount of hydrogen is picked up through corrosion in LAFR irradiation and reactor irradiations, the total hydrogen concentration may reach 360 ppm. This level is in the acceptable range given by Sabol.[1]

Because the range of the high-energy protons is small compared to the dimensions of the LAFR fuel assemblies and the flux will be forward peaked, there will be a nonuniform concentration of hydrogen in the fuel cladding. Rotating the fuel from close to the source to farther away will reduce the average hydrogen concentration and distribute it more evenly. Although the proton-generated hydrogen will only reach moderate levels, the potential synergetic effects it may have on the mechanical properties suggest that further study is required.

As with hydrogen production, the high-energy proton flux produces helium via (p, x$\alpha$) reactions. In one LAFR irradiation about 10 ppm helium is produced. This concentration of helium as shown by Carpenter,[2] when implanted in zirconium and then subsequently irradiated in the electron microscope to a dose of ~3.4 dpa, produced voids. At an exposure of ~8.6 dpa the voids had grown to a diameter of ~200 Å. Based on this experimental result and the calculation of helium concentration and exposure for the first LAFR irradiation, one must conclude that voids will be produced in the Zircaloy cladding. It is expected that even fuel shuffling would not reduce the helium concentration enough to alter this conclusion when all four irradiations are considered.

The effects of voids on the physical properties of Zircaloy have not been studied. On the basis of the presence of voids alone,one cannot predict that their presence will seriously limit cladding performance. They do,however, represent a major and significant change in the development of radiation damage microstructure in Zircaloy. As discussed previously, the mechanical property changes in Zircaloy tend to saturate at a dose level of 1 - 2 x $10^{-21}$ n/cm$^2$ (~1 dpa). This observation suggests that the developing damage microstructure tends to stabilize at least as far as it significantly effects the properties measured. Assuming that void growth in Zircaloy would be similar to that in other alloys, their presence represents a dynamically changing and growing damage microstructure. This projection is a dramatic and significant departure from the present picture of saturated physical property changes.

To illustrate the impact of this problem consider the following hypothetical irradiation history of an LAFR-LWR fuel assembly. The fabricated fuel is irradiated in LAFR and a void-influenced damage microstructure is developed. At the start of the first LWR fuel burn we have not a damage-free microstructure but one that has the potential of growing and producing property changes different and greater than that experienced in LWRs. The fuel assembly is then reirradiated in an LAFR with an established but perhaps still growing microstructure. There is some reason to suggest that in the presence of a matured microstructure, a reirradiation in LAFR may develop a new microstructure in the presence of the existing one. This effect has been observed when samples with a developed void structure are reirradiated. In some cases a new void structure develops. For the final LWR fuel burn, the cladding will have a damage microstructure quite different than that for the first one.

The potential for the development of a void microstructure in the LAFR-LWR system must be considered a fundamental concept feasibility question based on a materials requirement of the fuel cladding. For the system to work as conceived, the fabricated fuel assemblies must meet performance requirements in LAFR service, in reactor service,and for hot remote handling. One might expect that the performance codes on the structural components may be stricter in this case due to the increased number of operations.

## III. CONCLUSIONS

The radiation damage produced by the LAFR neutron spectrum is expected to be similar to that produced in an LWR. The increased helium production may alter this picture, but its potential deleterious effects seem to be small when compared to those projected for the proton flux. The total exposure for the Zircaloy structural elements of 14.4 dpa is a moderate level.

Even though the fuel assemblies will not be in the direct proton beam, proton irradiation presents the most serious damage problem. The low-energy ($E <$ 20 MeV) leakage protons must and can be eliminated from the fuel assembly radiation field. The high-energy ($E >$ 20 MeV) protons cannot be eliminated and result in a very serious materials damage problem. The expectation that Zircaloy-clad fuel elements can be used in this system is, in part, based on the assumption that the radiation damage effects saturate at an acceptable level. The proton-induced damage places this assumption in serious question. Since the structural elements in the fuel assemblies will be required to undergo four irradiations without failure, the synergetic effects of the high-energy proton and neutron damage in Zircaloy must be considered a fundamental feasibility question.

### Required Research Activities Relating to Feasibility

1. Simulate the radiation conditions in the LAFR using high-energy protons and neutrons to develop the type of damage microstructure that will exist at the end of the first LAFR irradiation. Grow this microstructure in a fission reactor to simulate the properties of Zircaloy halfway through the proposed cycle. At this point, the response of Zircaloy to the LAFR-LWR conditions should be evident.
2. Proton transport, along with neutron transport in the target and fuel assemblies, should be done along with neutron transport so that better estimates of the proton flux energy spectrum and spatial distributions can be made.
3. Calculations of low-and high-energy proton damage including displacement cross sections, metallic impurity generation cross sections and gas production cross sections should be made so that coupled with the flux and spectral information from transport calculations, better estimates of the proton damage problem can be made.

4. Analysis of the experimental and calculational results should be simultaneously performed so that implications of the results and identified solutions, if any, can be properly factored into the development of the LAFR-LWR concept.

## REFERENCES

1. George P. Sabol, "Materials Limitations to High Fuel Burn-up in the Linear Accelerator Breeder-Light Water Reactor Concept," Westinghouse R&D center report 78-902-BROOK-R1 (1978).

2. G. J. C. Carpenter, "Void Formation in Zirconium Under Irradiation in the High-Voltage Electron Miscroscope," Radiation Effects 19, 189 (1973).

3. P. Grand, Brookhaven National Laboratory, private communication.

4. G. Russell, Los Alamos Scientific Laboratory, private communication.

5. C. Alton Coulter, Don M. Parkin and Walter V. Green, "Calculation of Radiation Damage Effects of 800-MeV Protons in a Thin Copper Target," J. Nucl. Mater. 67, 140 (1977).