A NEUTRONICS FEASIBILITY STUDY FOR THE LEU CONVERSION OF POLAND'S MARIA RESEARCH REACTOR^{*}

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Presented at the 1998 International Meeting on Reduced Enrichment for Research and Test Reactors

> October 18-23, 1998 Sao Paulo, Brazil

Work supported by the U.S. Department of Energy Office of Nonproliferation and National Security under Contract No. W-31-109-ENG-38

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ABSTRACT

The MARIA reactor is a high-flux multipurpose research reactor which is water-cooled and moderated with both beryllium and water. Standard HEU (80% ²³⁵U) fuel assemblies consist of six concentric fuel tubes of a U-Al alloy clad in aluminum. Although the inventory of HEU (80%) fuel is nearly exhausted, a supply of highly-loaded 36%-enriched fuel assemblies is available at the reactor site. Neutronic equilibrium studies have been made to determine the relative performance of fuels with enrichments of 80%, 36% and 19.7%. These studies indicate that LEU (19.7%) densities of about 2.5 gU/cm³ and 3.8 gU/cm³ are required to match the performance of the MARIA reactor with 80%-enriched and with 36%-enriched fuels, respectively.

INTRODUCTION

Located in Swierk, Poland, the MARIA Research Reactor is a multipurpose high-flux reactor. Standard U-Al alloy HEU (80 wt % 235 U) fuel assemblies (FA) consist of six circular concentric fuel tubes each with a wall thickness (clad plus fuel meat) of 2.0 mm, water channels of thickness 2.5 mm between the fuel tubes, and a fuel height of 100 cm. Fuel assemblies are surrounded by beryllium and are located on a square grid with a 13.0 cm pitch at the core midplane. The reactor power depends on the core configuration, but is typically of the order of 20 MW. About 10% of all source neutrons within the reactor come from the ⁹Be(n,2n) reaction. However, the buildup of ³He and ⁶Li poisons in the beryllium matrix, initiated by the ⁹Be(n, α) reaction, can significantly limit the available excess reactivity. For a more complete description of the MARIA reactor see Ref. 1.

Although MARIA's supply of HEU (80% 235 U) fuel is nearly exhausted, an on-hand inventory of 49 UO₂-Al fresh fuel assemblies with a 235 U enrichment of 36% is available. Each of these highly-loaded fuel assemblies contains about 550 g 235 U compared with 350 g 235 U for the standard HEU (80% 235 U) fuel. The primary purpose of this study was to determine UO₂-Al LEU (19.7% 235 U) fuel requirements needed to match the performance of the 80%-enriched reference fuel and the anticipated performance of the highly-loaded 36%-enriched fuel.

FUEL CHARACTERISTICS AND MULTIGROUP MICROSCOPIC CROSS SECTIONS

Table I summarizes the characteristics of the fuels analyzed in this study. Although clad and meat thicknesses vary with enrichment, fuel tube and water channel thicknesses are fixed at the values of 2.00 and 2.50 mm, respectively. For the proposed LEU fuels, clad and meat thicknesses were taken from Ref. 2. It was assumed that for all enrichments fresh fuel had zero concentrations of 234 U and 236 U and that the length of the fuel column was 100 cm. A dispersant (UO₂) volume fraction of about 40% is probably a practical upper limit for reliable and economical extrusion of fuel tubes.

| Wt % | g ²³⁵ U | Fuel Meat | U Dens. | U Disp. | t _{meat} | t _{clad-in} | t _{clad-out} |
|------------------|--------------------|---------------------|-------------------|-----------------------|-------------------|----------------------|-----------------------|
| ²³⁵ U | per FA | | g/cm ³ | Vol F, % ^a | mm | mm | mm |
| 80.0 | 350 | UAl Alloy | 1.28 | 28.3 | 0.40 | 0.80 | 0.80 |
| 36.0 | 550 | UO ₂ -Al | 2.37 | 25.9 | 0.75 | 0.64 | 0.61 |
| 19.7 | 402 | UO ₂ -Al | 2.53 | 27.6 | 0.94 | 0.53 | 0.53 |
| 19.7 | 524 | UO ₂ -Al | 3.30 | 36.1 | 0.94 | 0.53 | 0.53 |
| 19.7 | 600 | UO ₂ -Al | 3.78 | 41.3 | 0.94 | 0.53 | 0.53 |

Table I. MARIA Reactor Fuel Characteristics

^aThe UO₂ dispersant volume fraction equals the uranium density divided by the product of the UO₂ density and the weight fraction of uranium in the dispersant. A value of 10.38 g/cm³ was used for the UO₂ density which is 95% of the theoretical density.

Complete sets of microscopic cross sections were generated at 300K for each uranium loading using the supercell option in the WIMS-ANL code and a 69-group ENDF/B-VI-based library³. Cross sections were collapsed into 7 broad groups with energy boundaries of 10.0 MeV, 0.821 MeV, 5.530 keV, 4.0 eV, 0.625 eV, 0.250 eV, 0.058 eV, and 1.0E-5 eV. Burnup-dependent cross sections were generated for heavy metal actinides and for fission product nuclei. Cross sections were also created for the non-fueled regions in the reactor including the beryllium matrix (with its poisons), graphite and water reflectors, in-core water holes, and control rods (30 wt % B₄C and 70 wt% Al), Al control rod followers, and control rod channels. Cross section sensitivity studies to unit cell modeling methods are reported in Ref. 1.

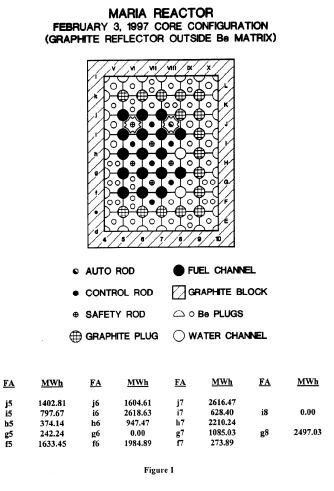
COMPARISONS OF DIFFUSION AND MONTE CARLO CALCULATIONS FOR THE FEBRUARY 3, 1997 MARIA CRITICAL EXPERIMENT

Because of uncertainties in the multigroup cross sections and the limitations of diffusion theory, this experimentally-observed HEU (80%) critical configuration has been analyzed with 3D diffusion and Monte Carlo codes. This procedure determines bias factors present in the diffusion-theory analyses of reactor cores with fuels of different enrichment.

Core Configuration

Figure 1 shows the configuration of the 17-fuel-assembly February 3, 1997 core. The observed MWh's of burnup for each fuel assembly is shown at the bottom of the figure. Using a calculated value of 1.263 g/MWd for this HEU (80%) fuel, the residual ²³⁵U in each fuel assembly was determined. These masses were compared with fuel assembly masses in global depletion calculations in a similar core in order

to determine burnup-dependent atom densities for 5 equal-height axial segments for each fuel assembly. These region-dependent atom densities were used in subsequent diffusion and Monte Carlo calculations. For a xenon-free condition, the reactor was observed to be critical for the safety rods fully withdrawn, the control rods banked at 330 mm above the fully inserted position, and the autorod withdrawn 420 mm. For a fully inserted rod, the bottom of the 975 mm B_4C -Al column was 12.5 mm above the bottom of the active fuel.



Poison Concentrations in the Beryllium Matrix

Because of the previous irradiation and shutdown history of the MARIA reactor, large concentrations of ³He and ⁶Li in the beryllium matrix strongly influence the critical state of the February 3, 1997 core. The buildup and burnout of these poisons begins with the fast neutron threshold reaction ⁹Be(n, α)⁶He, the rapid beta decay of ⁶He to ⁶Li, the strong ⁶Li(n, α)³H thermal neutron reaction, the beta decay of tritium to ³He, and the very strong ³He(n,p)³H thermal neutron reaction. The equations governing these reactions are given in Ref's 4 and 5. Their solutions depend on the neutron fluxes in the beryllium matrix, the nuclear cross sections, and on the detailed irradiation and shutdown history of the reactor. Because the tritium concentration in the beryllium matrix of the MARIA reactor is much larger than that of ³He, the reactor need not be shutdown very long before tritium decay significantly elevates the ³He concentration even though the tritium half-life is 12.3 yr. The methods used to estimate the poison concentrations in an inner beryllium region surrounding the fuel and an outer beryllium reflector region at the time when the critical

experiment was conducted are discussed in Ref. 6. The concentrations (atoms/b-cm) obtained from these calculations and corresponding to the February 1997 critical experiment are:

| | ^{3}H | 3 He | <u>⁶Li</u> |
|---------------|-----------|-----------|-----------------------|
| Inner Be Reg. | 1.108E-05 | 6.033E-07 | 1.424E-06 |
| Outer Be Reg. | 1.357E-06 | 1.389E-07 | 4.225E-07 |

Results

 $MCNP^7$ Monte Carlo and DIF3D⁸ diffusion calculations were made for this February 1997 MARIA critical experiment using the above poison concentrations, the axially-dependent fission product and actinide atom densities for each fuel assembly, and control rods withdrawn to the elevations stated earlier. For the diffusion calculations the control rods were treated by a set of group-dependent internal boundary conditions (i.e. current-to-flux ratios) applied at the clad surface of the B₄C-Al rod and obtained from a P₁S₁₆ TWODANT⁹ calculation with fresh fuel. Results from these calculations are summarized below.

| <u>Quantity</u> | MCNP-Monte Carlo | DIF3D Diffusion |
|---------------------|------------------|------------------------|
| K-eff | 1.00150±0.00028 | 1.02421 |
| H-VII Rod Worth | -2.32±0.04 %δk/k | -2.56 %δk/k |
| Worth of Be Poisons | -6.96±0.07 %δk/k | -6.55 %δk/k |

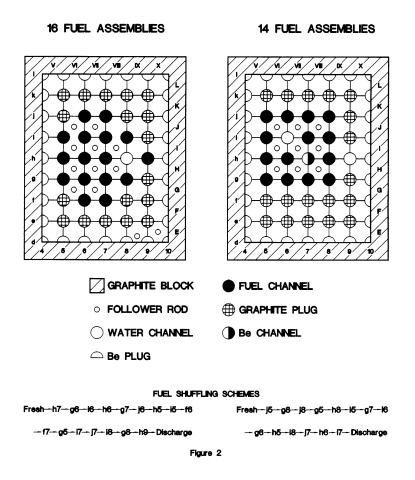
Beam tubes were included in the MCNP model of the assembly, but were omitted in the DIF3D calculations. From the Monte Carlo calculations the worth of the beam tubes was found to be -0.17 ± 0.04 % $\delta k/k$. Control rod worths were measured in this core by the rod drop method. With $\beta_{eff} = 0.00725$, the measured worth of the H-VII rod (see Fig. 1) was -1.48 % $\delta k/k$. Why this measured worth is much smaller than the calculated values is not understood. Relative to MCNP, the DIF3D reactivity bias is about 2.0 % $\delta k/k$.

EQUILIBRIUM CYCLE CALCULATIONS FOR ²³⁵U ENRICHMENTS OF 80.0, 36.0 AND 19.7 Wt %

Burnup calculations have been made for equilibrium cores in the MARIA reactor in order to determine LEU fuel requirements needed to approximately match the performance of the HEU (80%) reference fuel and the on-hand 36%-enriched fuel. The core configurations used for these studies are shown in Figures 2. Each core is radially reflected with graphite and axially reflected with water. The 16-fuel-assembly core is very similar to a MARIA core operated in February 1998. The smaller, 14-fuel-assembly, core was needed to reduce the excess reactivity with the highly-loaded 36%-enriched fuel. For both of these configurations equilibrium cycle calculations use a fuel management scheme in which one fresh fuel assembly is added and one burned fuel assembly is discharged per cycle. These fuel management schemes are shown at the bottom of Figures 2. The REBUS code¹⁰ was used for these equilibrium cycle depletion calculations for which the safety rods, control rods, and the autorod were fully withdrawn.

LEU fuel loadings were chosen so as to approximately match the eigenvalues at the end of the equilibrium cycle (EOEC) for the 80%-enriched reference fuel (16 FA core) and for the on-hand 36%-enriched fuel (14 FA core). For each case the cycle length was chosen so that the average ²³⁵U discharge burnup was 45%, which is the maximum burnup currently allowed for the MARIA reactor¹¹. The peak burnup corresponding to this average value is about 56%. LEU fuel assemblies require a thicker fuel meat and a higher uranium density (see Table 1).

MARIA REACTOR EQUILIBRIUM CORES (GRAPHITE REFLECTOR OUTSIDE 6 x 8 Be MATRIX)



For all these REBUS depletion calculations the initial poison concentrations in the inner and outer beryllium regions are based on the operating history of the MARIA reactor as of February 1998. The February 3, 1997 poison levels were updated to February 1998 values using the reactor operating history described in Ref. 11 and the methods discussed in Ref. 6. The REBUS code allows for changes in beryllium poison concentrations during the burn cycle and during shutdown times between cycles. Table II summarizes results from these equilibrium cycle fuel depletion calculations. The LEU (19.7%) "equivalent" for the HEU (80%) reference fuel has a fuel assembly ²³⁵U mass of 402 g and a uranium density in the fuel meat of 2.53 g/cm³. For 45% burnup the cycle length is 18% longer than that for the HEU (80%) fuel which means that fewer assemblies would be used per year. Similarly, the LEU "equivalent" for the 36%-enriched fuel has a fuel assembly mass of 600 g 235 U, a density of 3.78 gU/cm³ in the fuel meat, and an increased cycle length of about 11%. Note that the LEU fuel assembly with 524 g ²³⁵U has insufficient excess reactivity at the end of the equilibrium cycle (EOEC) to operate because of the DIF3D reactivity bias discussed earlier. To use this fuel a shorter cycle length, a lower discharge burnup, and a higher annual fuel consumption rate would be necessary. However, the performance of this core would still exceed that of the reference core with 80%-enriched fuel.

| Wt. % | g ²³⁵ U | Fuel | Density | No. of | Reactor | Cycle | K-EFF | K-EFF | Max | FA's |
|------------------|--------------------|---------------------|--------------------|---------|---------|-------------------|---------------------|---------------------|--------|---------------------|
| ²³⁵ U | per FA | Type | gU/cm ³ | FA's in | Power | Length | BOEC | EOEC | MW per | used |
| | | | | core | MW | Days ^a | | | FA | per yr ^b |
| 80.0 | 350 | U-Al | 1.28 | 16 | 17 | 7.50 | 1.0723 | 1.0527 | 1.66 | 21.4 |
| 19.7 | 402 | UO2-Al | 2.53 | 16 | 17 | 8.85 | 1.0766 | 1.0587 | 1.64 | 18.1 |
| 36.0 | 550 | UO ₂ -Al | 2.37 | 16 | 17 | 11.88 | 1.1552 ^c | 1.1367 | 1.62 | 13.5 |
| 36.0 | 550 | UO ₂ -Al | 2.37 | 14 | 16 | 12.69 | 1.0611 | 1.0478 | 1.23 | 12.7 |
| 19.7 | 600 | UO ₂ -Al | 3.78 | 14 | 16 | 14.08 | 1.0594 | 1.0469 | 1.23 | 11.4 |
| 19.7 | 524 | UO ₂ -Al | 3.30 | 14 | 16 | 12.30 | 1.0363 | 1.0234 ^d | 1.23 | 13.1 |

Table II. Summary of Equilibrium Cycle Results for the MARIA Reactor

^aThe cycle length is chosen to give an average ²³⁵U discharge burnup of 45.0%.

^bThese fuel consumption estimates are based on the 1997 value of 3856 hours on power per year¹¹.

^cThis core is too reactive for the control system. A smaller core is needed.

^dThe DIF3D reactivity bias indicates that this core has insufficient reactivity at the end of the equilibrium cycle (EOEC). A shorter cycle length and a lower discharge burnup is required.

Table III shows that the LEU cores have negligible differences in neutron fluxes in the h8 (Fig. 2, 16 FA's) and the i6 (Fig. 2, 14 FA's) water holes relative to the 80%- and 36%-enriched fuels. Note that the thermal neutron fluxes in the 14-assembly cores with 36%-enriched and with 19.7%-enriched fuels are about 30% larger than in the HEU (80%) 16-assembly core even though the power level is 16 MW instead of 17 MW.

| of m-Core water noies | | | | | | | |
|-----------------------|-----------|---|--------------------------|--------------------------|--------------------------|--|--|
| Group | Eupper | BOEC Neutron Fluxes in Units of 10 ¹³ n/cm ² -sec | | | | | |
| | | 16 Fuel Assembly Core | | 14 Fuel Assembly Core | | | |
| | | h8 Water Hole | | i6 Water Hole | | | |
| | | Power = 17 MW | | Power = 16 MW | | | |
| | | 80% Enr. 19.7% Enr. | | 36% Enr. | 19.7% Enr. | | |
| | | 350g ²³⁵ U/FA | 402g ²³⁵ U/FA | 550g ²³⁵ U/FA | 600g ²³⁵ U/FA | | |
| | | | | | | | |
| 1 | 10.0 MeV | 1.58 | 1.58 | 2.22 | 2.23 | | |
| 2 | 0.821 MeV | 2.53 | 2.59 | 3.61 | 3.63 | | |
| 3 | 5.530 keV | 3.10 | 3.14 | 4.36 | 4.39 | | |
| 4 | 4.00 eV | 0.98 | 0.99 | 1.37 | 1.37 | | |
| 5 | 0.625 eV | 0.60 | 0.60 | 0.83 | 0.83 | | |
| 6 | 0.250 eV | 9.25 | 9.02 | 12.15 | 11.99 | | |
| 7 | 0.058 eV | 15.59 | 15.08 | 20.24 | 19.94 | | |
| | 1.0E-5 eV | | | | | | |
| Total | | 33.62 | 32.99 | 44.78 | 44.37 | | |

Table III. MARIA Reactor Neutron Fluxes on Midplane of In-Core Water Holes

SUMMARY AND CONCLUSIONS

The MARIA reactor is an unusual research reactor in that water-cooled fuel assemblies are spaced on a square grid within a beryllium matrix. Thus, beryllium is both part of the core and part of the radial reflector. As a result, the ${}^{9}Be(n,2n)$ reaction contributes about 10% of the total source neutrons within the reactor. On the other hand, the ${}^{9}Be(n,\alpha)$ reaction initiates the development of ${}^{6}Li$ and ${}^{3}He$ poisons within the matrix. Depending on the irradiation and shutdown history of the reactor, these poisons may contribute a substantial negative reactivity. At the time of the February 3, 1997, critical experiment the worth of these poisons was nearly -7% $\delta k/k$. Taking these beryllium effects into account, equilibrium fuel cycle analyses have been done to compare the relative performance for fuels with ${}^{235}U$ enrichments of 80%, 36%, and 19.7%. Based on these calculations, the following six conclusions have been reached.

- With a detailed history of the reactor operation schedule and dividing the beryllium matrix into core and reflector regions, the poison concentrations can be calculated with reasonable accuracy by solving the relevant differential equations. This was successfully illustrated by comparing the calculated and measured state of the February 3, 1997, critical experiment.
- Based on equilibrium cycle calculations, LEU fuel (19.7%-enriched, 402 g 235 U/FA, and $\rho_U = 2.53$ g/cm³) nearly matches the performance of the HEU (80%) reference fuel but with a somewhat longer cycle length for the same 45% average discharge burnup. The LEU core would use 18 FA's/yr instead of the 21 FA's/yr for the 80%-enriched case. The volume fraction of UO₂ in the LEU dispersion fuel is 27.6%. Irradiation tests of many assemblies containing UO₂-Al dispersion fuel with 2.5 gU/cm³ (36%-enriched) were successfully completed¹² by the Russian RERTR program in the 1980's.
- Fuel assemblies from the on-hand inventory of 36%-enriched fuel (550g ²³⁵U/FA) are significantly more reactive than the standard HEU fuel (80%-enriched, 350 g ²³⁵U/FA). For a 16-fuel-assembly equilibrium core, they are too reactive for the control system to accommodate. However, an equilibrium core with 14 fuel assemblies containing 36%-enriched uranium has been identified and meets all operation requirements.
- LEU UO₂-Al dispersion fuel needed to match the performance of the above 36%-enriched fuel requires a loading of 600 g ²³⁵U/FA and a uranium density of 3.78 g/cm³. This corresponds to a UO₂ volume fraction of 41.2%. However, fuel element failures under irradiation for UO₂-Al dispersion fuels with 3.85 gU/cm³ and 19.7% enrichment have been reported¹³.
- Fuel cycle calculations were also made for LEU fuel assemblies of intermediate mass (524 g ²³⁵U/FA and 3.30 gU/cm³). This option can be considered if 3.8 gU/cm³ UO₂-Al fuel cannot be fabricated reliably and economically or if irradiation testing of this fuel is not successful. For the 14-fuel-assembly LEU core, these fuel assemblies with 524 g ²³⁵U will result in a shorter cycle length and lower discharge burnup (<45%) than the existing 36%-enriched fuel. However, core performance will still be better than the HEU (80%) fuel.
- If necessary, one could consider fabricating MARIA LEU fuel assemblies using advanced high-density uranium fuels^{14,15} now being tested^{16,17,18}. For example, the performance of the 36% fuel could be matched with a U-Mo (10 wt %) alloy dispersion fuel with a uranium density of about 4.2 g/cm³ which corresponds to a dispersant volume fraction of 27.6%. The cladding thickness could be increased from 0.53 mm to about 0.60 mm with a dispersed phase volume fraction of about 32% and to 0.68 mm with a corresponding volume fraction of about 40%. Ref. 16 reports that this U-Mo alloy dispersion fuel can be fabricated with good thermal stability, with reprocessing properties similar to aluminide fuel, and with no significant impact on the vitrification process. Positive results from irradiation tests are

given in Ref's. 17 and 18. Effects of parasitic absorption in Mo are discussed in Ref. 14. However, no detailed calculations with these high-density fuels have yet been made for the MARIA reactor.

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