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Monolithic Fuel and High-Flux Reactor Conversion

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Abstract. Monolithic fuels are the most promising candidate for a next generation of high-density research reactor fuels. If successfully developed, the remaining HEU-fueled reactors in the world could presumably be converted to low-enriched fuel — and the use of highly enriched uranium in the civilian nuclear fuel cycle eventually terminated.

The most challenging type of reactors to convert are single element reactors because their core geometry is generally the least flexible. This specific reactor type is therefore the primary focus of this article. Based on new computational tools and optimization methods, neutronics calculations are presented to assess the potential of monolithic fuels for conversion of high-flux reactors in general and of single element reactors in particular.

Introduction

First reported in 2002, initial tests of so-called monolithic fuel revealed excellent irradiation behavior of this material up to very high burnup levels [Hofman and Meyer, 2002]. Additional irradiation experiments with monolithic fuel, which is characterized by effective uranium densities of 16–17 g/cc, are currently underway, while a variety of adequate fabrication techniques are being investigated in parallel [Clark et al., 2004]. Unfortunately, during the same period, serious problems with UMo-dispersion fuels have surfaced. As has been previously reported [Hofman et al., 2004], [Lemoine et al., 2004], porosity formation in the fuel leads to excessive swelling for elevated fission rates and densities, which may ultimately complicate the qualification of this fuel.

For two reasons, these developments are particularly relevant for the conversion of high-flux reactors: First, effective uranium densities achievable with UMo-dispersion fuels may in some cases be insufficient to match the original performance of a previously HEU-fueled facility. Here, monolithic fuel would be the only alternative to attain or approach the LEU limit. Figure 1 compares effective U-235 densities achievable with different fuel-types.

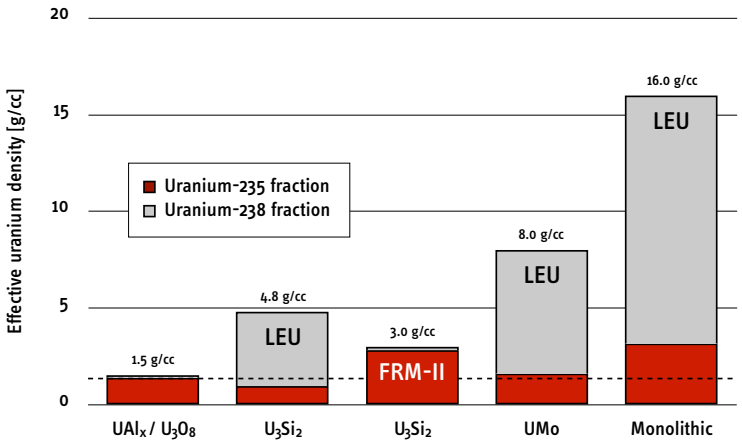

Figure 1: Effective uranium densities in research reactor fuels.

Second, the use of UMo-dispersion may be precluded by the operational conditions encountered in this reactor type anyway: high-flux reactors in general, and single element reactors in particular, are characterized by extremely high life-averaged fission rates *and* very high maximum fission densities at end-of-life. While UMo-dispersion fuels may display stable irradiation behavior in medium-flux reactors and be qualified for these conditions, it is highly unlikely that these fuels will be usable in high-flux and single element reactors.

Neutronics calculations are therefore important to explore the potential of monolithic fuel for single element reactor conversion. Below, a specially developed computational system (M^3O) and optimization procedure is used to re-optimize the core geometry of a single element reactor for use with monolithic fuel and reduced enrichment. To demonstrate the technique, the case of FRM-II is discussed as an example.

Computational System

Figure 2 illustrates the functional relationship of the individual codes that build the computational system M^3O (*Mathematica*-MCODE-MCNP-ORIGEN), which has been developed and used to produce all results discussed below.

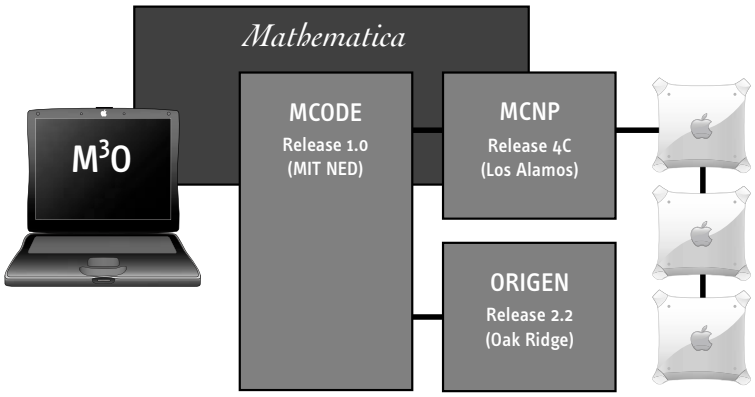

Figure 2: Computational system M^3O for research reactor analysis.

At the basic level of the system, the Monte-Carlo neutron-transport code MCNP [Briesmeister (ed.), 2000] and the point-depletion code ORIGEN2 [Croff, 1980/2002] perform the actual neutronics calculations. The communication between both programs is coordinated by the linkage program MCODE [Xu et al., 2002]. Initial MCNP input decks of complete three-dimensional models of arbitrary single element reactors are prepared by numerous modules programmed in *Mathematica*. The latter is also used to evaluate and visualize the results returned by MCNP and MCODE.

As a special feature and instead of having a regular and strictly rectangular structure with burnup zones of equal size, a characteristic *adaptive cell structure* (ACS) is used for all burnup calculations [Glaser et al., 2003]. The idea of such an adaptive cell structure is to join smaller areas within the plate with expected similar burnup behavior in one single burnup zone. Typically, between 10 and 25 burnup zones are defined using about 100 MCNP cells to describe the structure of each fuel plate. The basic MCNP model is modified correspondingly and used for subsequent burnup calculations executed with ORIGEN2 for all segments of the plate using spectrum-averaged and burnup-dependent data determined with MCNP.

Case Study FRM-II

The Forschungsreaktor München II (FRM-II) is a 20 MW single element reactor based on a very compact core using involute-shaped fuel plates.[1] FRM-II is chosen as the reference system for analysis because it is the only research reactor worldwide that uses highly enriched fuel with an effective uranium density of 3.0 g/cc (uranium-silicide fuel). The reactor can therefore be considered the most difficult reactor to convert to low-enriched fuel. For this reason, FRM-II also is a prime candidate to explore the potential of monolithic fuel and to develop optimization methods for identification of optimum options with reduced enrichment based on this fuel.

LEU in current core geometry

The most straightforward approach to use monolithic fuel in FRM-II would be to simply replace the fully-enriched U_3Si_2 dispersion-type fuel with monolithic LEU fuel.[2] Figure 3 shows the results of corresponding M^3O burnup calculations.

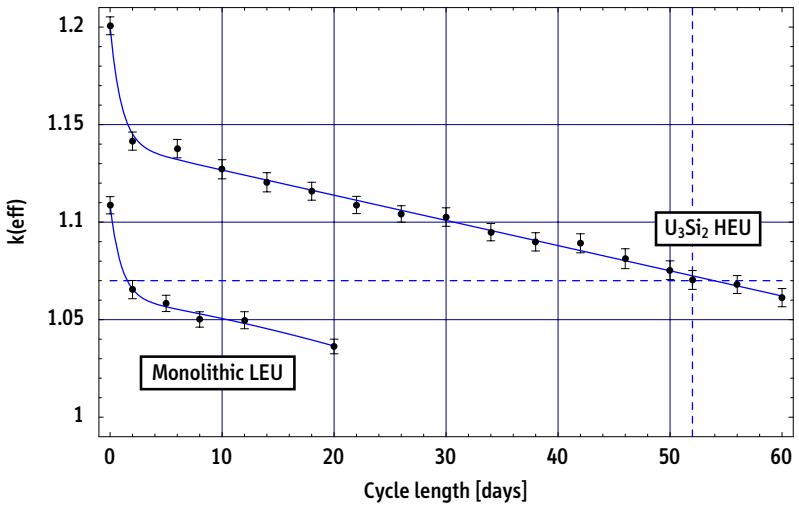

Figure 3: Cycle length achievable with monolithic LEU fuel in original FRM-II HEU geometry.

[1]The reactor has recently been renamed to *Heinz Maier-Leibnitz Neutron Source*.

[2]The inner section of the plate, in which the original uranium density was 3.0 g/cc, would now contain fuel with an uranium density of 16 g/cc. In the outer section of the plate, a reduced effective uranium density of 8 g/cc of same enrichment would be used. In practice, various strategies are conceivable to lower the effective uranium-235 density in the fuel, which include both reduced meat thickness and reduced enrichment.

The cycle length for the LEU option is unacceptably low (about 2 days). As can be inferred from the figure, the reason for this behavior is not due to the reactivity loss rate, which is nearly equivalent to the original HEU design, but due to the low initial reactivity of $k_{\text{eff}} = 1.109 \pm 0.005$. Once the xenon equilibrium is reached, the core reactivity has almost dropped to the pre-defined EOL criterion of $k_{\text{eff}} = 1.07$. For this reason, strategies to increase the initial reactivity of the core are key when exploring fuel with reduced enrichment in single element reactors that were originally designed for HEU.

Design variables — Strategies to increase initial core reactivity

The following set of independent design variables $\vec{x} = (x_1, \ldots, x_9)$ is used to describe the main characteristics of the FRM-II core.[3] Due to the given constraints imposed by the current reactor design, some of these variables will be assumed constant in the considerations below.

x_1 :	Meat thickness	x_4 :	Inner core radius	x_7 :	Fuel enrichment
x_2 :	Cladding thickness	x_5 :	Outer core radius	x_8 :	Transition radius
x_3 :	Coolant channel	x_6 :	Active core height	x_9 :	Density ratio

Variation of the fuel plate and coolant channel dimensions (x_1 through x_3) are particularly relevant in the present context, because both affect the H/HM ratio in the core. With monolithic fuel, the average neutron spectrum in the plate can be expected to harden significantly, which is due to the substantial increase of the heavy metal inventory in the core and requires re-optimization for reduced enrichment.

For obvious reasons, fuel enrichment x_7 plays a unique role in the above set of design variables: higher enrichment will always yield higher initial reactivity, but the lowest possible value is preferred for nonproliferation reasons. Fuel enrichment is therefore fixed prior to the following optimization process and the best reactor performance is subsequently determined for a particular enrichment level.

The sensitivity of core reactivity to separate variables may be studied as a function of enrichment to identify an initial model for further analysis. As an example, Figure 4 shows the initial k_{eff} of the core for the standard geometry as well as for an elongated fuel element with an active height of 80 cm. Generic M^3O burnup calculations suggest that a minimum value of $k_{\text{eff}} = 1.17$ is needed for monolithic fuel to achieve the target cycle length of 52 days in the FRM-II geometry. To meet this criterion, a minimum enrichment of 32.5% is required for the original fuel element (70 cm) and of 26.0% for an active core height of 80 cm.

[3]Obviously, with adequate design-specific modifications, this or a similar set of variables can also be used to describe any other single element reactor, too. The transition radius x_8 and the density ratio x_9 are FRM-II-specific variables to describe the graded uranium density in the plate.

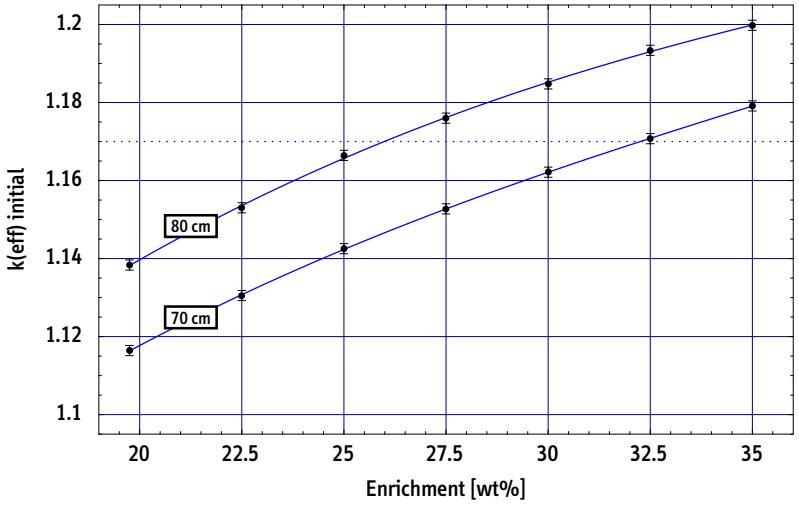

Figure 4: Initial reactivity of the FRM-II core as a function of enrichment using monolithic fuel. Original core height and elongated version with 80 cm. Minimum initial k_{eff} value to achieve target cycle length is approx. 1.17 for given fuel type and core geometry. MCNP 4B/C calculations.

A detailed sensitivity analysis for all variable parameters goes beyond the scope of this article.[4] Instead, the two candidate options (from the figure) are used in the following to demonstrate the effectiveness of further optimization. In the calculations for the elongated fuel element, the power level of the reactor is increased by 10% to 22 MW in order to reproduce the original average power density in the core, while enrichment is set at 27.5% to account for a higher expected reactivity loss rate.[5]

Optimization of reduced enrichment options

The linear programming (LP) technique outlined in the Appendix is applied to improve and optimize the initial models (see corresponding columns in Table 1).[6] The objective of this process is both to satisfy a set of additional constraints and to maximize the thermal neutron flux ϕ. Several core design parameters are pre-defined in the optimization process: only the meat thickness x_1, the width of the cooling channel x_3, the

[4]For instance, equivalently to Figure 4, k_{eff} versus enrichment could be plotted for variable meat thickness or coolant channel width.

[5]An increased power level may or may not be acceptable in the case of FRM-II.

[6]The use of this method for research reactor optimization has first been proposed in [Mo, 1991].

transition radius x_8, and the discontinuity factor x_9 are allowed to vary.[7] According to the terminology introduced in the Appendix, the linear programming problem can be formulated as follows: Maximize the objective function represented by the thermal neutron flux $\phi(x_1, \ldots, x_9)$ subject to the constraints:

Cycle length:	$C_1(x_1, \ldots, x_9) \geq$ 52 days
Average power density:	$C_2(x_1, \ldots, x_9) \leq$ 1100 W/cm^3
Average heat flux:	$C_3(x_1, \ldots, x_9) \leq$ 200 W/cm^2
Power peaking factor #1:	$C_4(x_1, \ldots, x_9) \leq$ 2.0
Power peaking factor #2:	$C_5(x_1, \ldots, x_9) \leq$ 2.0

This set of constraints $(C_1, \ldots, C_5)$ is a representative one and can be modified or extended as needed. In the present case, the power peaking factors #1 and #2 are measured at the uranium-density transition and at the periphery of the fuel plate.

Using the initial core model to formulate the linear programming problem, the LP algorithm yields a new model, whose main data and results are summarized in Table 1. The code suggests increased coolant channel widths for both cases to soften the neutron spectrum,[8] while reducing the total number of fuel plates in the core. Table 1 also includes results of a final MCNP simulation that verifies the data for the new model.[9] In general, the predicted and verified data are in excellent agreement. Most importantly, all constraints are now satisfied, particularly the power peaking in the plate, and the neutron flux has increased further by several percent. Figure 5 shows thermal neutron flux levels in the moderator tank for all design options.

Incidentally, Table 1 and Figure 5 also demonstrate the potential of monolithic fuel for FRM-II conversion. Using monolithic fuel enriched to 32.5% in the original fuel element geometry, implies a relatively modest loss in maximum thermal neutron flux of 9.7% relative to the HEU design (7.3 versus 8.0×10^{14} n/cm^{2}s). At a distance of 40 cm from the core centerline, which corresponds to the central position of the cold neutron source, the loss reduces to 8.2%. The second conversion option candidate envisions modifications to the core geometry *and* to the power level of the reactor, but reduces the relative performance loss even further: it amounts to 5.2% (7.6×10^{14} n/cm^{2}s) at maximum and to 3.3% at the position of the cold neutron source, respectively.

[7] While the cladding thickness is assumed constant (0.38 mm), the minimum value of the meat thickness is set at the original value of 0.60 mm to provide a minimum plate thickness and to guarantee mechanical and thermohydraulical stability of the fuel plate. Similarly, inner and outer core radii (x_4 and x_5) are fixed at their respective original values.

[8] More precisely, due to the high uranium density in the fuel, thermalized neutrons re-entering the core from the moderator tank are mainly absorbed in the periphery of the plate. Wider coolant channels increase the relative importance of the central zones of the core, which is preferable for overall neutronics.

[9] In principle, the model of the first iteration could be used to set-up an execute a second iteration.

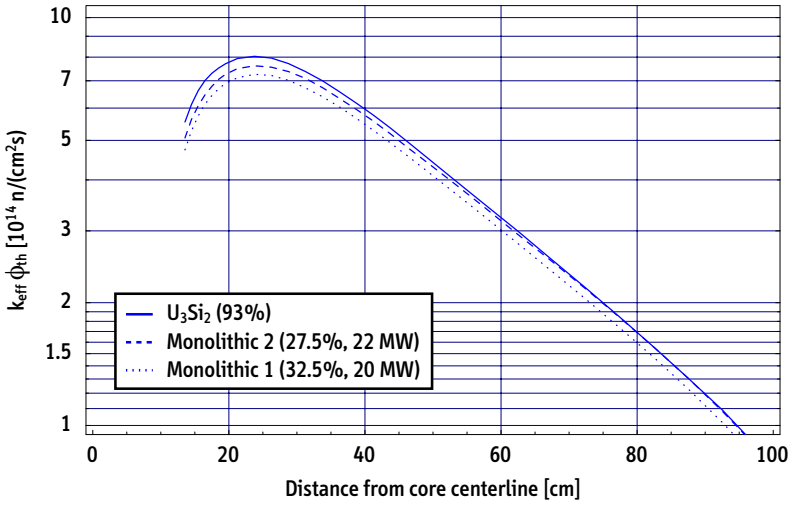

Figure 5: Thermal neutron flux for various FRM-II core variants.

	FRM-II Monolithic 1			FRM-II Monolithic 2		
	Enrichment: 32.5 wt% Active core height: 70 cm; Power level: 20 MW			Enrichment: 27.5 wt% Active core height: 80 cm; Power level: 22 MW		
	Start	LP Solution	Verification	Start	LP Solution	Verification
x_1	0.60 mm	0.62 mm		0.60 mm	0.60 mm	
x_3	2.20 mm	2.53 mm		2.20 mm	2.70 mm	
x_8	10.56 cm	10.48 cm		10.56 cm	10.48 cm	
x_9	0.50	0.45		0.50	0.49	
Plates	113	104		113	100	
k(eff)	1.169 ± 0.001	1.173	1.172 ± 0.001	1.174 ± 0.001	1.182	1.180 ± 0.001
ϕ	7.15E14	7.25E14	7.26E14	7.56E14	7.57E14	7.62E14
C_1	52–54 days	52 days	52–56 days	52 days	52 days	52 days
C_2	1024 kW/cc	1024 kW/cc	1024 kW/cc	985 kW/cc	985 kW/cc	985kW/cc
C_3	182 W/cm^2	200 W/cm^2	200 W/cm^2	175 W/cm^2	200 W/cm^2	200 W/cm^2
C_4	1.67	1.68	1.66	1.73	1.67	1.66
C_5	2.13	2.00	1.99	2.08	2.00	1.99

Table 1: Basic results for two candidate core options with reduced enrichment using monolithic fuel. Variable core parameters recommended by linear programming algorithm (LP Solution). All neutronics calculations (Start and Verification) performed with M^{3}O.

Conclusion and Outlook

If monolithic fuel could be successfully qualified, it would offer a tremendous potential for the conversion of the remaining HEU-fueled research reactors worldwide. Nonetheless, conversion of high-flux reactors using HEU in very compact cores (single element reactors) would still be a challenging undertaking. A few typical problems have been identified and discussed in this article: these include, in particular, the requirement to guarantee sufficient initial core reactivity to achieve satisfactory cycle lengths as well as the requirement to address power peaking issues, which may result from the substantial increase of heavy metal inventory in the core.

The FRM-II has been analyzed as an example since its conversion to low-enriched fuel would be most difficult, even with monolithic fuel. Even though the primary focus of this article is on methodology, not on particular numerical results, preliminary data suggest that an enrichment level of not higher than 32.5% is sufficient for monolithic fuel in the current FRM-II geometry. Enrichment levels of less than 28–30% would be feasible, if certain core and reactor modifications were allowed. Simultaneously, such targeted modifications could be used to further reduce the relative performance loss to very low values compared to the original HEU design. Note that much higher enrichment levels (about 50%) would be necessary to obtain similar results using UMo-dispersion fuels in FRM-II, even if these fuels could be qualified for operational conditions encountered in high-flux reactors.

Re-optimization of research reactors for use of low-enriched monolithic fuel does strongly benefit from the availability of adequate optimization tools. The linear programming technique, which is outlined in an appendix to this paper, is one approach to address this problem. Such optimization strategies can be used both to guarantee important operational constraints, which may be violated when moving from HEU to high-density LEU fuel, and to re-optimize reactor performance for LEU fuel. The formalism suggested in this article, in conjunction with a specially-designed computer code environment to perform necessary neutronics calculations, may be the basis for further developments in that direction.

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A P P E N D I X

Linear Programming Technique and Research Reactor Optimization

The determination of adequate core parameters for a research reactor is a complex optimization process that typically depends upon a number of design variables and constraints. If this problem were a truly linear one, it could be formulated as follows:

$$\begin{aligned} \text{maximize} \quad & \sum_{j=1}^{n} c_j \, x_j \\ \text{subject to} \quad & \sum_{j=1}^{n} a_{ij} \, x_j \leq b_i \qquad (i = 1, 2, \ldots, m) \\ & x_j \geq 0 \qquad (j = 1, 2, \ldots, n) \end{aligned}$$

Here, one objective function of the independent variables x_j is maximized, while a set of additional functions of x_j has to be satisfied simultaneously (constraints). Mathematically, this type of problem is addressed with the *Linear Programming* (LP) technique. Typical LP problems are extremely underdetermined, i.e. there are much more independent variables than there are equations ($n \gg m$). Such systems can be solved very efficiently using the Simplex algorithm [Chvátal, 1983].

As with research reactor optimization, practical LP problems are, of course, seldom truly linear. In these cases, the fundamental equations of the LP problem may be linearized in the vicinity of an initial feasible point $\vec{x}^0$ and a solution identified in an iterative process [Reklaitis et al., 1983, Chapter 8].[10] Application of this method has been suggested and tested previously for research reactor performance optimization [Mo, 1991]. Here, a modified version is developed and specially designed for single element reactor analysis.

As usual, the maximum thermal neutron flux $\phi(\vec{x})$ is selected as the primary objective function to be maximized. As the functional dependency of $\phi(\vec{x})$ from the design variables is *a priori* unknown, the thermal neutron flux is linearized around an initial point $\vec{x}^0$.

[10] It goes without saying that linearization methods have to be used with great caution and that the analyst has to guarantee the validity of the linearized problem. Adequate safeguards, such as step-size adjustment, may have to be taken. In general, there is no assurance that a true optimum is obtained in the process.

$$\phi(\vec{x}) = \phi(\vec{x}^{\,0}) + \sum_{i=1}^{n} \frac{\partial \phi}{\partial x_i}(x_i - x_i^0)$$

The partial derivatives of $\phi(\vec{x})$ are estimated in MCNP simulations for small perturbations $(x_j - x_j^0)$ of individual design variables with $\vec{x}^{\,j} = (x_1^0, x_2^0, \ldots, x_j, \ldots, x_n^0)$.

$$\left.\frac{\partial \phi}{\partial x_j}\right|_{\vec{x}=\vec{x}^{\,0}} \approx \frac{\phi(\vec{x}^{\,j}) - \phi(\vec{x}^{\,0})}{x_j - x_j^0} \qquad \text{with } \phi(\vec{x}^{\,j}),\ \phi(\vec{x}^{\,0}) \text{ from MCNP}$$

Analogous to the linearized function approximating the thermal neutron flux, the constraints too are expanded into first-order Taylor series.

$$C_j(\vec{x}) = C_j(\vec{x}^{\,0}) + \sum_{i=1}^{n} \frac{\partial C_j}{\partial x_i}(x_i - x_i^0)$$

The partial derivatives required to construct the linearized approximations of the constraint conditions fall into two categories: one subset can be directly derived from explicit functions of the design variables. For instance, using the notation of the main text for the design variables x_i and the constraints C_i, the average power density in the core C_2 and the average heat flux C_3 are given by the following expressions.[11]

$$C_2(\vec{x}) = \frac{P_{\text{th}}}{\pi} \frac{1}{x_7\,(x_6^2 - x_5^2)} \qquad \text{and} \qquad C_3(\vec{x}) \approx \frac{P_{\text{th}}}{2\pi} \frac{x_1 + 2\,x_2 + x_3}{x_7\,(x_6^2 - x_5^2)}$$

For a second subset, such functions are unavailable. In those cases, MCNP-based perturbation calculations are performed to acquire appropriate numerical values in the vicinity of the linearization point $\vec{x}^{\,0}$.

$$\left.\frac{\partial C_i}{\partial x_j}\right|_{\vec{x}=\vec{x}^{\,0}} \approx \frac{C_i(\vec{x}^{\,j}) - C_i(\vec{x}^{\,0})}{x_j - x_j^0} \qquad \text{with } C_i(\vec{x}^{\,j}), C_i(\vec{x}^{\,0}) \text{ from MCNP}$$

The most challenging constraint to process is the cycle length C_1: in order to execute the LP process in a reasonable time, the objective is to estimate C_1 without actually performing burnup calculations for a given $\vec{x}$. Only once a promising set of design variables has been identified, the cycle length is verified in a final M^3O burnup calculation.

[11] To obtain the expression for the average heat flux C_3, the upper limit of the number of fuel plates as well as the surface area of the involute-shaped plates have to be calculated first. The expression is an approximation, because an integer is ultimately chosen for the number of plates in the core.

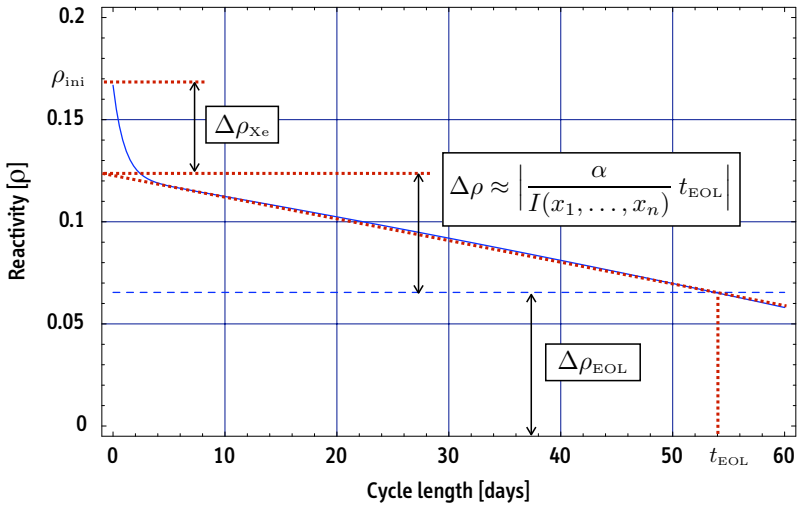

Figure 6: Typical reactivity loss during irradiation for a single element reactor.

As shown in Figure 6, the typical reactivity loss during irradiation is nearly linear once the xenon equilibrium is reached after a few days. An estimate of the average reactivity loss rate can therefore be used to obtain an approximation of the maximum cycle length, which is achieved when the reactivity drops below $\Delta\rho_{\mathrm{EOL}}$. In the following, it is assumed that $\Delta\rho/\Delta t$ is inversely proportional to the initial uranium-235 inventory in the core.

$$\frac{\Delta\rho}{\Delta t} \approx -\frac{\alpha}{I(x_1,\ldots,x_n)}\,, \quad \alpha > 0$$

The characteristic constant α, which scales the reactivity loss rate, can be determined for the initial base design and is used during the iteration process. The initial U-235 inventory can be calculated directly from the set of design variables. Introducing the unfueled radius ϵ of the plate, the total U-235 inventory in the core is given by:

$$I(\vec{x}) = \pi\, \rho_{\mathrm{eff}}\, \frac{x_1\, x_7\, x_6}{x_1 + 2\,x_2 + x_3} \Big[x_8^2 - x_4^2 - 2\,x_4\,\epsilon + x_9\big(x_5^2 - 2\,x_5\,\epsilon - x_8^2\big) \Big]$$

End-of-life is reached when the core reactivity drops below a pre-defined value ρ_{EOL}. This margin is introduced to account for reactivity losses associated with experimental and other reactor devices not modeled at this stage. Figure 6 illustrates the corresponding reactivity balance.

$$\Delta\rho_{\mathrm{EOL}} \stackrel{!}{=} -\frac{\alpha}{I(\vec{x})}\, t_{\mathrm{EOL}} + \Big[\rho_{\mathrm{ini}}(\vec{x}) - \Delta\rho_{\mathrm{Xe}}\Big]$$

The achievable cycle length $C_1(\vec{x}) = t_{\mathrm{EOL}}$ can therefore be approximated by:

$$C_1(\vec{x}) \approx \frac{I(\vec{x})}{\alpha}\Big[\rho_{\mathrm{ini}}(\vec{x}) - (\Delta\rho_{\mathrm{EOL}} + \Delta\rho_{\mathrm{Xe}})\Big]$$

In practice, fixed values are used for xenon-poisoning and end-of-life reactivity reserve: these are $\Delta\rho_{\mathrm{Xe}} = 0.045$ and $\Delta\rho_{\mathrm{EOL}} = 0.065$ for the example discussed in the main text. The partial derivatives for all $C_1(\vec{x})$ can be calculated based on the preceding expression.

$$\left.\frac{\partial C_1(\vec{x})}{\partial x_j}\right|_{\vec{x}=\vec{x}^{\,0}} = \frac{1}{\alpha}\left[\left.\frac{\partial I(\vec{x})}{\partial x_j}\right|_{\vec{x}=\vec{x}^{\,0}}\Big[\rho_{\mathrm{ini}}(\vec{x}^{\,0}) - (\Delta\rho_{\mathrm{EOL}} + \Delta\rho_{\mathrm{Xe}})\Big] + I(\vec{x}^{\,0})\left.\frac{\partial\rho_{\mathrm{ini}}(\vec{x})}{\partial x_j}\right|_{\vec{x}=\vec{x}^{\,0}}\right]$$

The partial derivatives of $I(\vec{x})$ can be calculated directly from the definition, while the sensitivity of the initial reactivity $\rho_{\mathrm{ini}}(\vec{x})$ has to be determined in MCNP simulations using the perturbation method discussed above.

In practice, *Mathematica* generates all MCNP input decks for the perturbed models, extracts the tally data from the MCNP output, determines the required partial derivatives, and solves the linearized set of equations with an enhanced version of the Simplex algorithm [Wolfram, 2001, implementation notes, Section A.9.4].

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