

Neutronenphysikalische Berechnungen zur Umstellbarkeit von Forschungsreaktoren auf niedrig angereichertes Uran

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Hintergrund

Forschungsreaktoren, allgemein

IAEA Datenbank: ca. 550 Reaktoren in mehr als 60 Ländern, heute ca. 280 in Betrieb
Besondere Bedeutung in der Grundlagenforschung: Neutronenstreuung

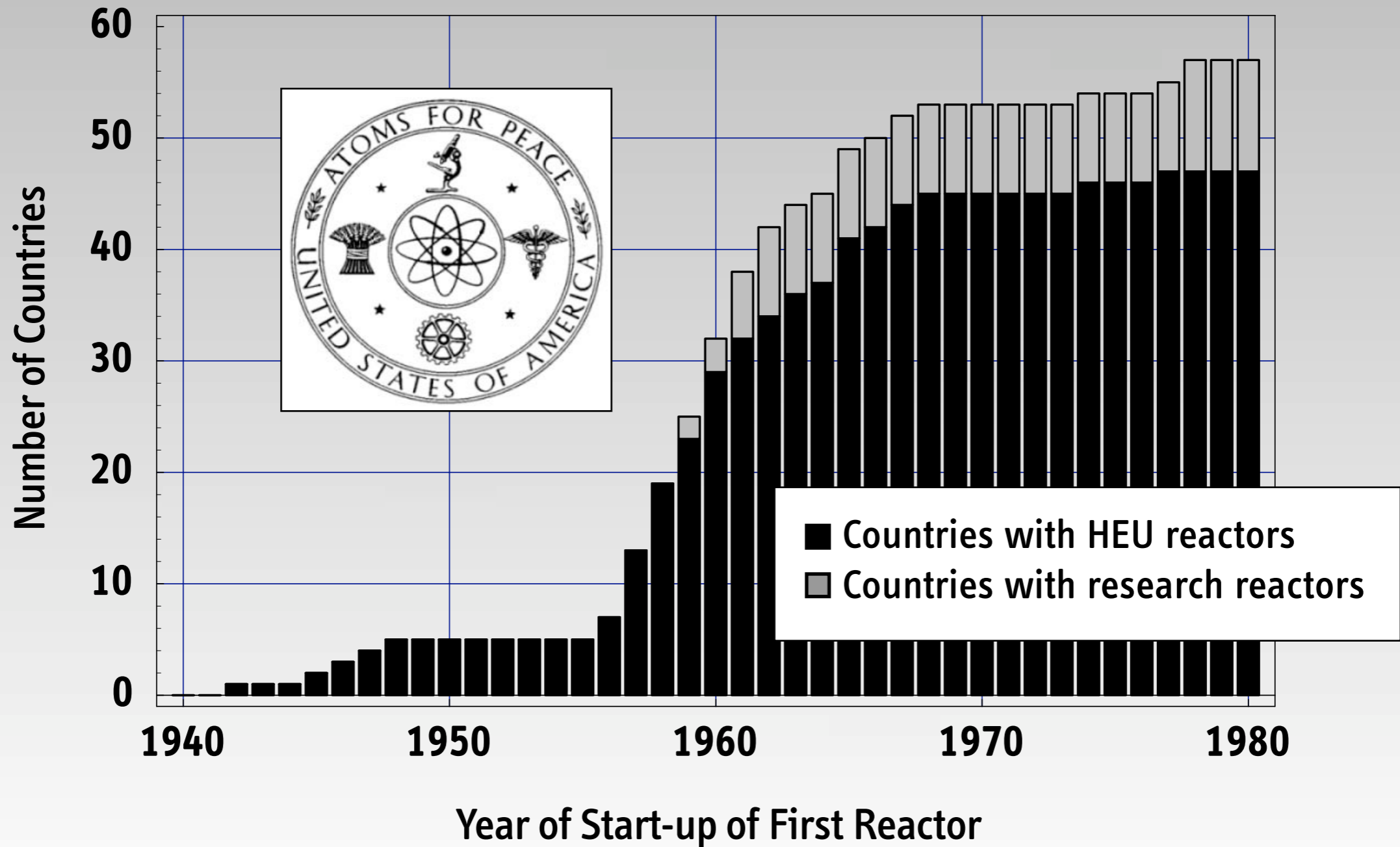
Hochangereichertes Uran und Proliferationsrisiko

HEU ist (neben Plutonium) für Kernwaffen geeignetes Material
ca. 50 Reaktoren (mit mind. 1 MW) weltweit nutzen hochangereichertes Uran

Umstellung von Reaktoren auf niedrig angereichertes Uran

Problemstellung: Wie kann dies ohne wesentliche Einbussen
in der Nutzbarkeit der Anlagen geschehen?

Number of Countries with Research Reactors



Research Reactor Applications

| Application | Reactors |
|--------------------------------|----------|
| 1. Neutron activation analysis | 71 |
| 2. Teaching | 68 |
| 3. Training | 63 |
| 4. Materials or fuel tests | 53 |
| 5. Isotope production | 48 |
| | |

| Application | Reactors |
|-------------------------------|----------|
| 6. Material structure studies | 34 |
| 7. Neutron radiography | 32 |
| 8. Transmutation | 21 |
| 9. Geochronology | 14 |
| 10. Neutron capture therapy | 9 |
| Other uses | 47 |

Data based upon a survey among operators
Table adapted from Dodd et al., Physica B (311), 2002

High-Flux-Reactor Grenoble

Institut-Laue-Langevin, 1972



Thermal power: 58 MW, max. thermal neutron flux: ca. $1.5 \cdot 10^{15}$ n/cm²s

HEU-Fueled Research Reactors

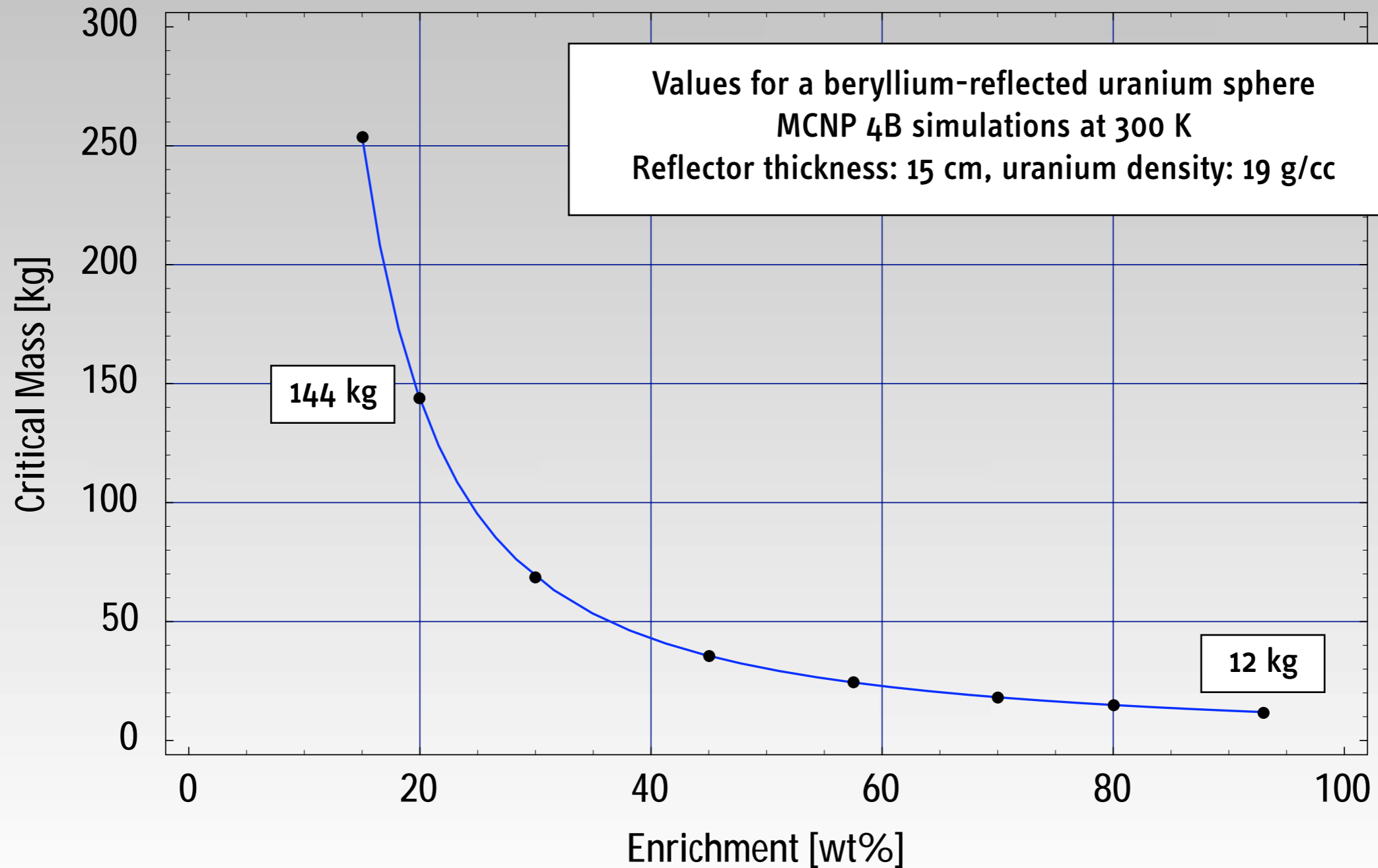
(Highest Relevance Group, 2004)

| Country | IAEA Code | Name | Criticality | Power | Enrichment | HEU Demand |
|--------------|-----------|---------|-------------|-----------|------------|----------------|
| USA | US-0070 | ATR | 1967/07 | 250 MW | 93% | 120–175 kg/yr |
| USA | US-0137 | HFIR | 1965/08 | 85–100 MW | 93% | 91–150 kg/yr |
| Russia | RU-0024 | SM-2 | 1961/10 | 100 MW | 90% | 43–110 kg/yr |
| China | CN-0004 | HFETR | 1979/12 | 125 MW | 90% | 75 kg/yr |
| Russia | RU-0013 | MIR-M1 | 1966/12 | 100 MW | 90% | 62.2 kg/yr |
| Kazakhstan | KZ-0003 | EWG-1 | 1972/01 | 60 MW | 90% | ? |
| France | FR-0017 | HFR | 1971/07 | 58.3 MW | 93% | 54.8 kg/yr |
| Germany | DE-0051 | FRM-II | 2004/03 | 20 MW | 93% | 40.5 kg/yr |
| Netherland | NL-0004 | HFR | 1961/11 | 45 MW | 93% | 38.3 kg/yr |
| Belgium | BE-0002 | BR-2 | 1961/06 | 80–100 MW | 74–93% | 29 kg/yr |
| USA | US-0204 | MURR | 1966/10 | 10 MW | 93% | 23.5 kg/yr |
| Germany | DE-0006 | FRJ-2 | 1962/11 | 23 MW | 80–93% | 19.2 kg/yr |
| Poland | PL-0004 | MARIA | 1974/12 | 17–30 MW | 36–80% | ? |
| France | FR-0022 | ORPHEE | 1980/12 | 14 MW | 93% | 15.8 kg/yr |
| Russia | RU-0008 | WWR-M | 1959/12 | 18 MW | 90% | 3.7–14.4 kg/yr |
| USA | US-0126 | NBSR | 1967/12 | 20 MW | 93% | 13 kg/yr |
| South Africa | ZA-0001 | SAFARI | 1965/03 | 20 MW | 87–93% | 12.6 kg/yr |
| USA | US-0120 | MITR-2 | 1958/07 | 4.9–10 MW | 93% | 1.6–12 kg/yr |
| Romania | RO-0002 | TRIGA-2 | 1979/11 | 14 MW | 20–93% | 11.8 kg/yr |
| Russia | RU-0010 | IVV-2M | 1966/04 | 15 MW | 90% | 3.5–9 kg/yr |
| Kazakhstan | KZ-0002 | IGR | 1961/01 | 10 MW | 36–90% | ? |
| Australia | AU-0001 | HIFAR | 1958/01 | 10 MW | 60% | 8.1 kg/yr |
| Russia | RU-0014 | IRT-T | 1967/07 | 6 MW | 90% | 5.6 kg/yr |

Hochangereichertes Uran und Proliferationsrisiko

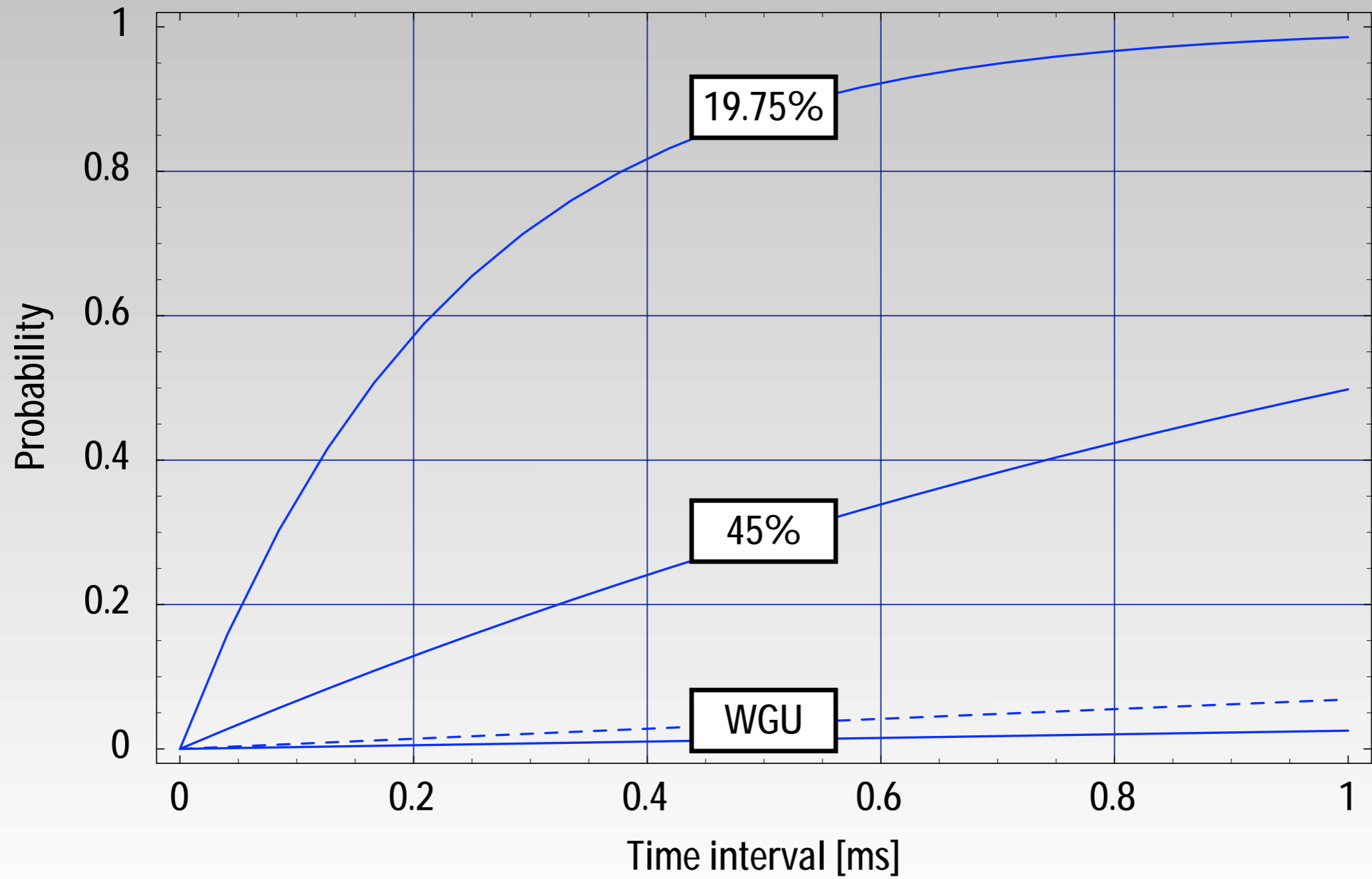
(Waffentauglichkeit, Mengen und geographische Verbreitung)

Critical Mass of Uranium



Spontaneous Fission in Uranium

Probability for at least one SF event in one bare critical mass



“With modern weapons-grade uranium, the background neutron rate is so low that terrorists, if they had such material, would have a good chance of setting off a high-yield explosion simply by dropping one half of the material onto the other half. Most people seem unaware that if separated U-235 is at hand it's a trivial job to set off a nuclear explosion ...”

Luis Alvarez, *Adventures of a Physicist*, Basic Books, 1987, p. 125

Umstellung von Forschungsreaktoren auf niedrig angereichertes Uran

(und die Entwicklung hochdichter Brennstoffe)

Internationaler Kontext

RERTR Programm (seit 1978) und Unterstützungsprogramme

(Reduced Enrichment for Research Reactor Test Reactors)

Kooperation zwischen dem U.S. RERTR Programm und dem entsprechenden russischen Programm seit 1996

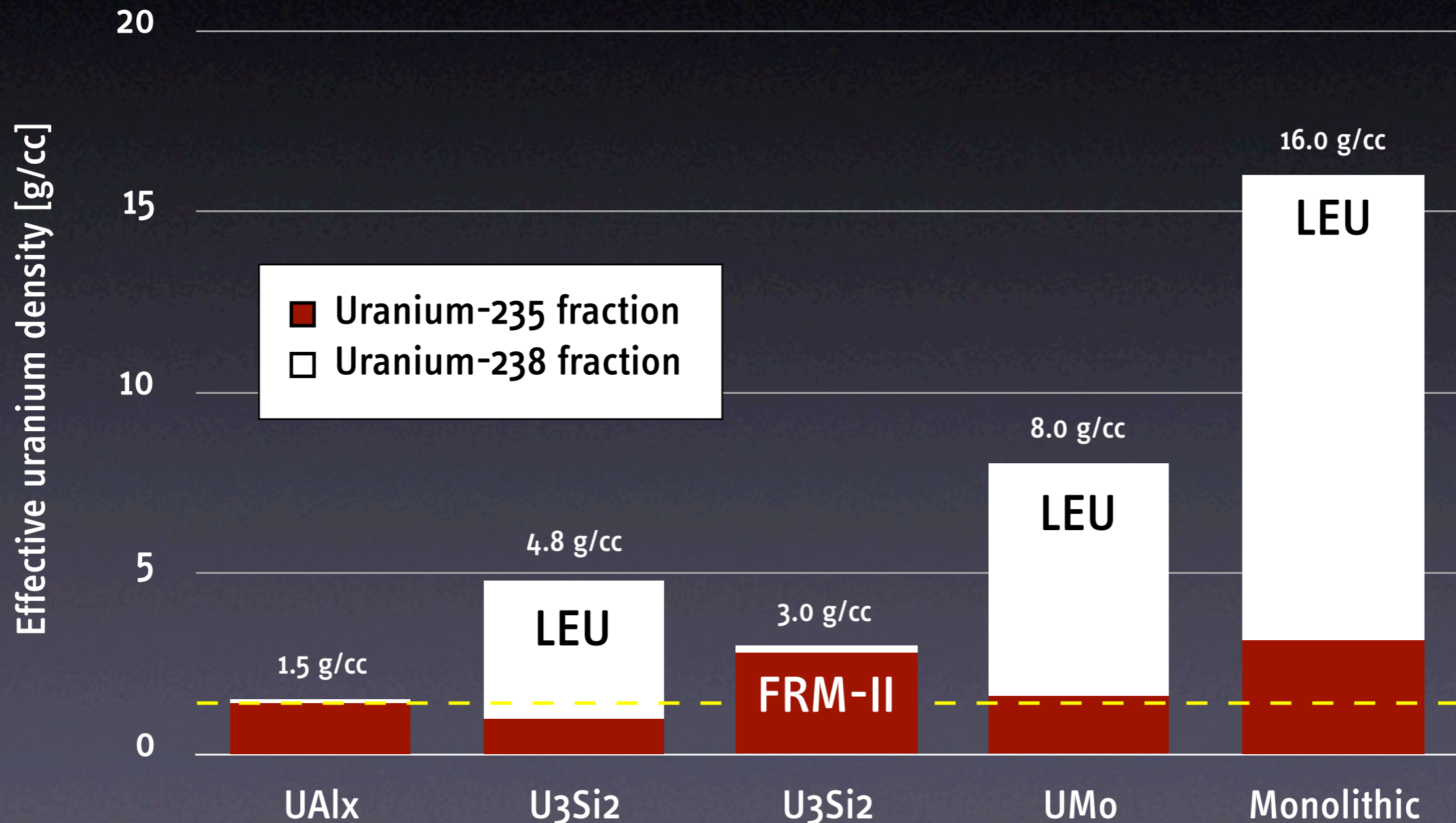
INFCE Konferenz (1978-1980)

Deutsches AF Programm (1979-1989)

51.5 Millionen DM zur Umstellung aller deutschen Reaktoren

Entscheidendes Element: Brennstoffentwicklung

Effective Uranium Densities in Research Reactor Fuels



Neutronics Calculations for Single Element Reactors

(Steady state and burnup calculations)

Methodik und Ziele

Entwicklung von Mathematica-Modulen zur Erzeugung von detaillierten 3D-Modellen von Forschungsreaktorgeometrien
(zur Analyse mit Monte Carlo Neutronentransportcodes)

Entwicklung des Programmsystems "M³O" für neutronenphysikalische Berechnungen basierend auf diesen 3D-Modellen
(Mathematica, MCODE, MCNP, ORIGEN)

Bestimmung wesentlicher Leistungsmerkmale von Forschungsreaktoren

Anwendungsbeispiele:
"Generic Single Element Reactor" (GSER) und FRM-II

Implementierung eines Algorithmus zur neutronenphysikalischen Optimierung von Forschungsreaktorgeometrien

Beurteilung der wissenschaftlichen Nutzbarkeit von Neutronenquellen

Nuclides in Burnup Calculations

130 Actinides
 850 Fission products
 720 Activation products

| | | | | | | | | | | |
|--------------------------------|-------------------------------|----------------------|----------------------|-------------------------------|--------------------------------|--------------------------------|-------------------|-------------------|--------------------------------|-------------------|
| | | | | | | | | Cm-243 29.1 a | Cm-244 18.10 a | Cm-245 8500 a |
| | | | | | | Am-240 50.8 h | Am-241 432.2 a | Am-242 141 a | Am-243 7370 a | Am-244 10.1 h |
| | | | | Pu-237 45.2 d | Pu-238 87.74 a | Pu-239 $2.411 \cdot 10^4$ a | Pu-240 6563 a | Pu-241 14.35 a | Pu-242 $3.750 \cdot 10^5$ a | Pu-243 4.956 h |
| | | | | Np-236 $1.54 \cdot 10^5$ a | Np-237 $2.140 \cdot 10^6$ a | Np-238 2.117 d | Np-239 2.355 d | Np-240 65 m | | |
| U-232 68.9 a | U-233 $1.592 \cdot 10^5$ a | U-234 0.0055 | U-235 0.7200 | U-236 $2.342 \cdot 10^7$ a | U-237 6.75 d | U-238 99.2745 | U-239 23.5 m | | | |
| | | $2.455 \cdot 10^5$ a | $7.038 \cdot 10^8$ a | | | $4.468 \cdot 10^9$ a | | | | |
| Pa-231 $3.276 \cdot 10^4$ a | Pa-232 1.31 d | Pa-233 27.0 d | Pa-234 6.70 h | | | | | | | |

Burnup Equations I

$$\frac{d}{dt} N_i(\vec{r}, t) = - \left[\int_0^\infty dE \sigma_a^i(E) \phi(E, \vec{r}, t) + \lambda_i \right] N_i(\vec{r}, t) \\ + \sum_{j \neq i} \left[\int_0^\infty dE f_{j \rightarrow i} \sigma_a^j(E) \phi(E, \vec{r}, t) + l_{j \rightarrow i} \lambda_j \right] N_j(\vec{r}, t)$$

$\phi(E, \vec{r}, t)$ itself is a function of $N(\vec{r}, t)$

**Complex structure of $\phi(E, \vec{r}, t)$
due to spatial and spectral resonance-self-shielding**

Burnup Equations II

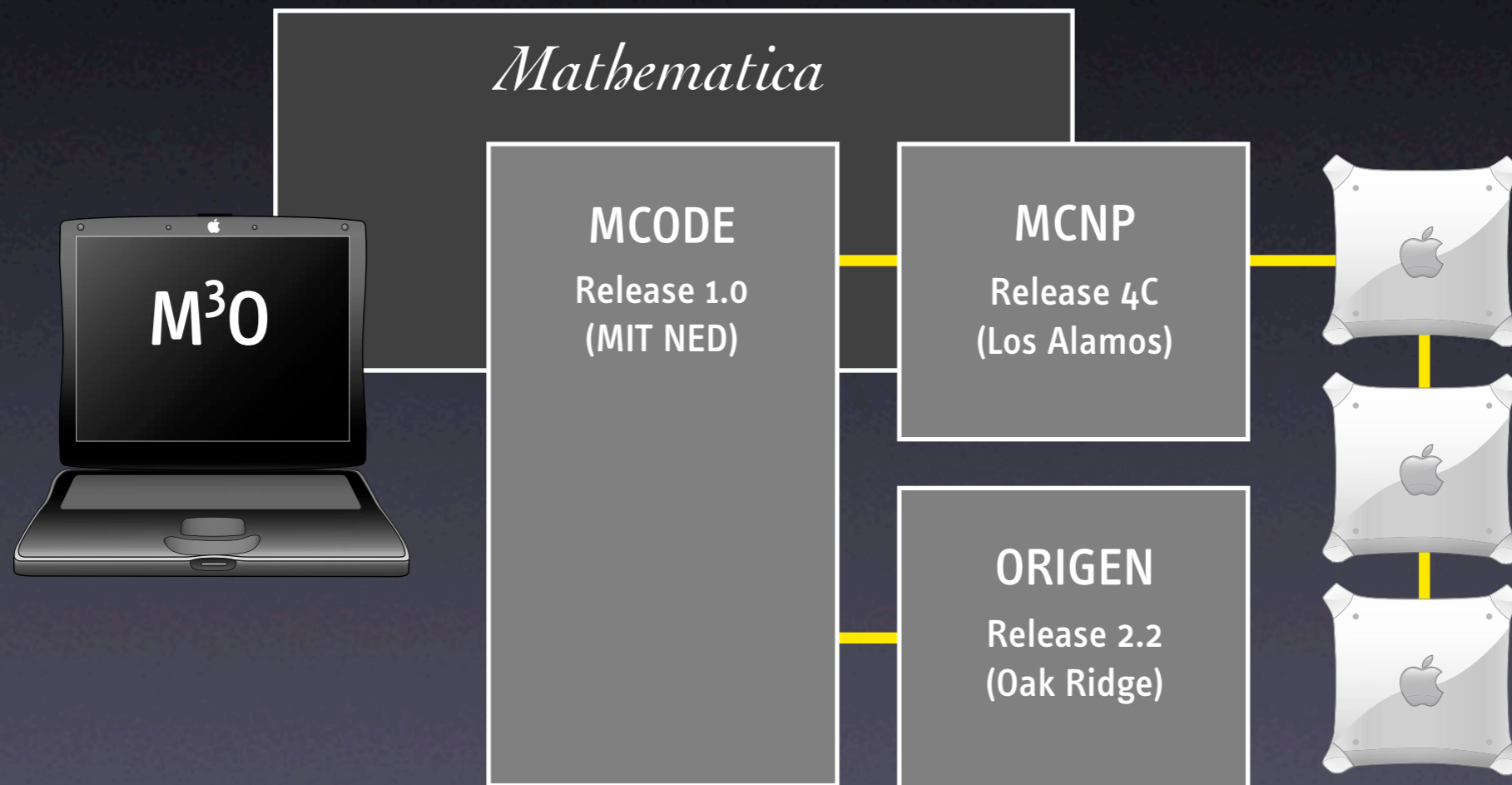
$$\phi_{\text{tot}}(\vec{r}, t) = \int_0^\infty dE \phi(E, \vec{r}, t) \quad \sigma(\vec{r}, t) = \frac{\int_0^\infty dE \sigma(E) \phi(E, \vec{r}, t)}{\int_0^\infty dE \phi(E, \vec{r}, t)}$$

Total neutron flux and spectrum-averaged cross-sections

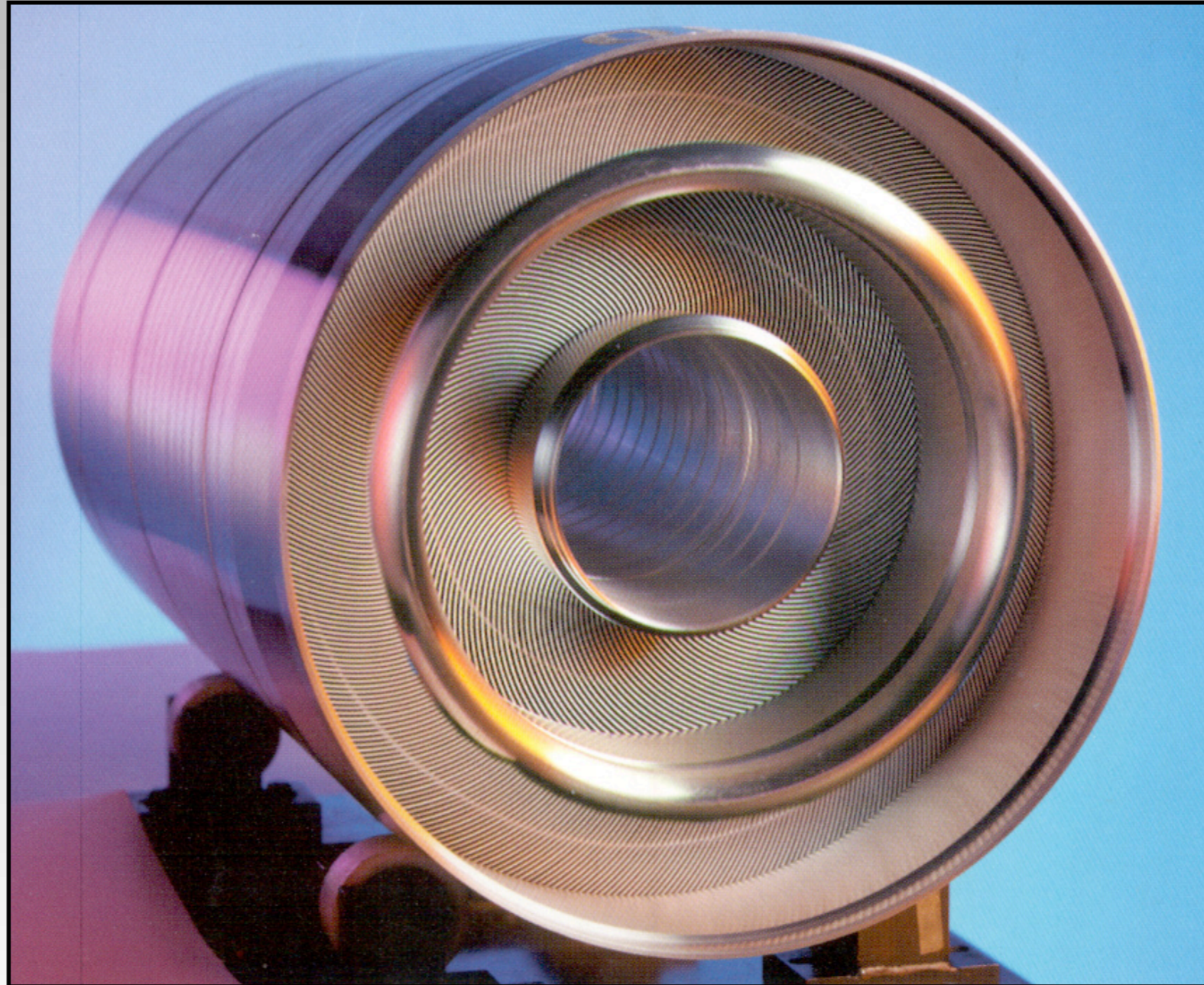
$$\begin{aligned} \frac{d}{dt} N_i(t) = & - \left[\sigma_a^i(t) \phi_{\text{tot}}(t) + \lambda_i \right] N_i(t) \\ & + \sum_{j \neq i} \left[f_{j \rightarrow i}(t) \sigma_a^j(t) \phi_{\text{tot}}(t) + l_{j \rightarrow i} \lambda_j \right] N_j(t) \end{aligned}$$

Burnup equations for point-depletion (cell burnup equations)

Computational System for Research Reactor Analysis

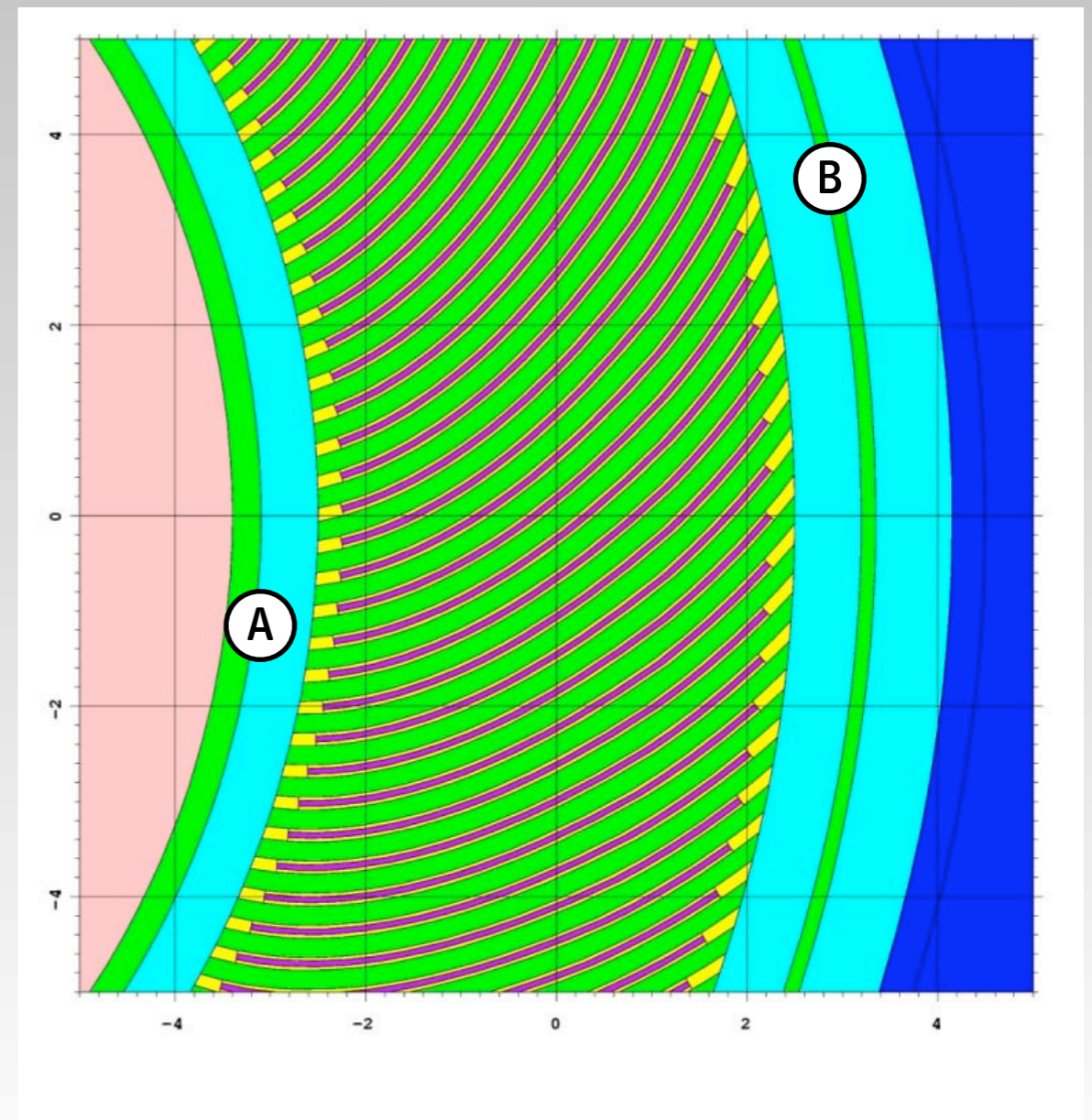
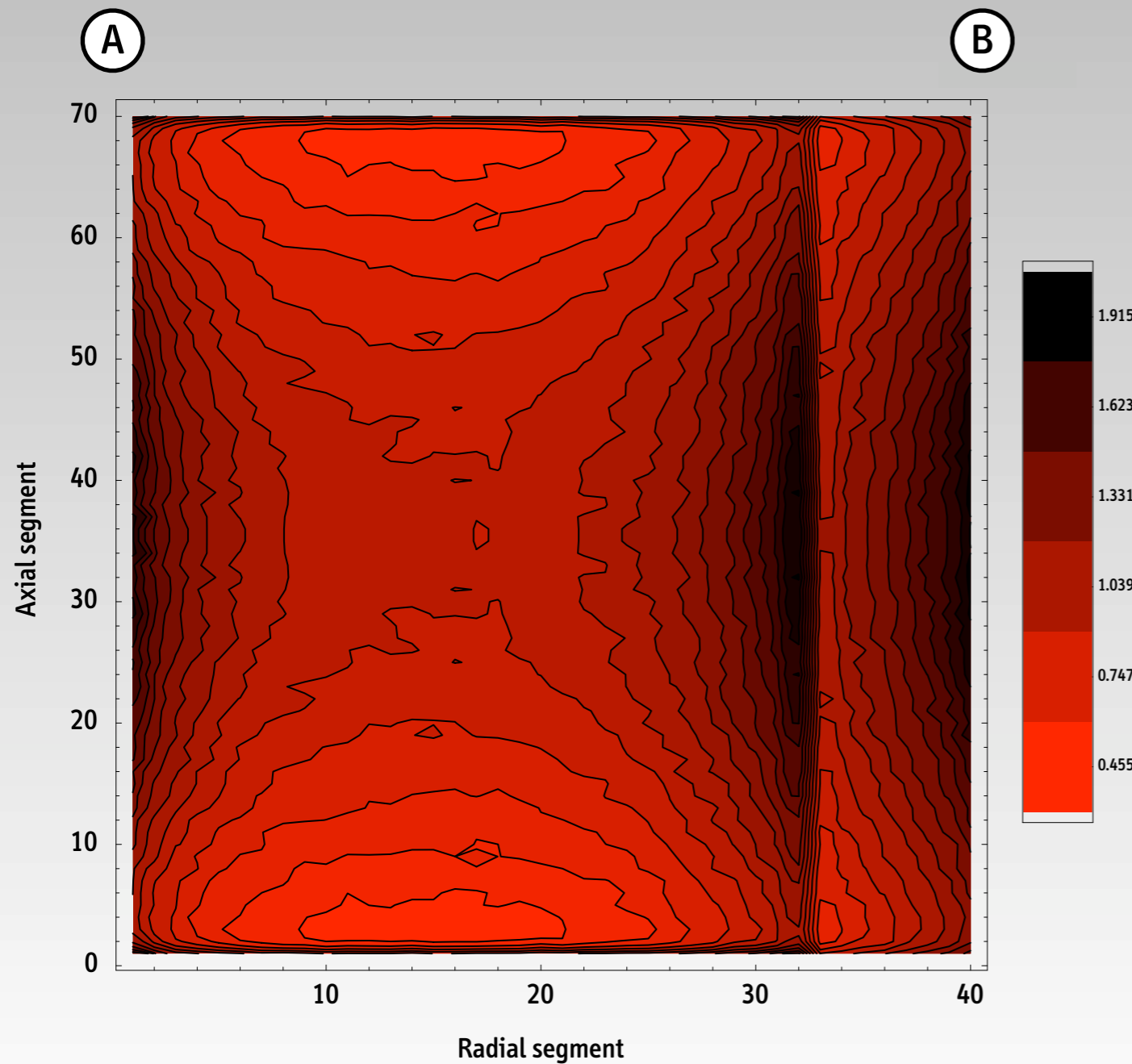


Fuel Element of HFIR

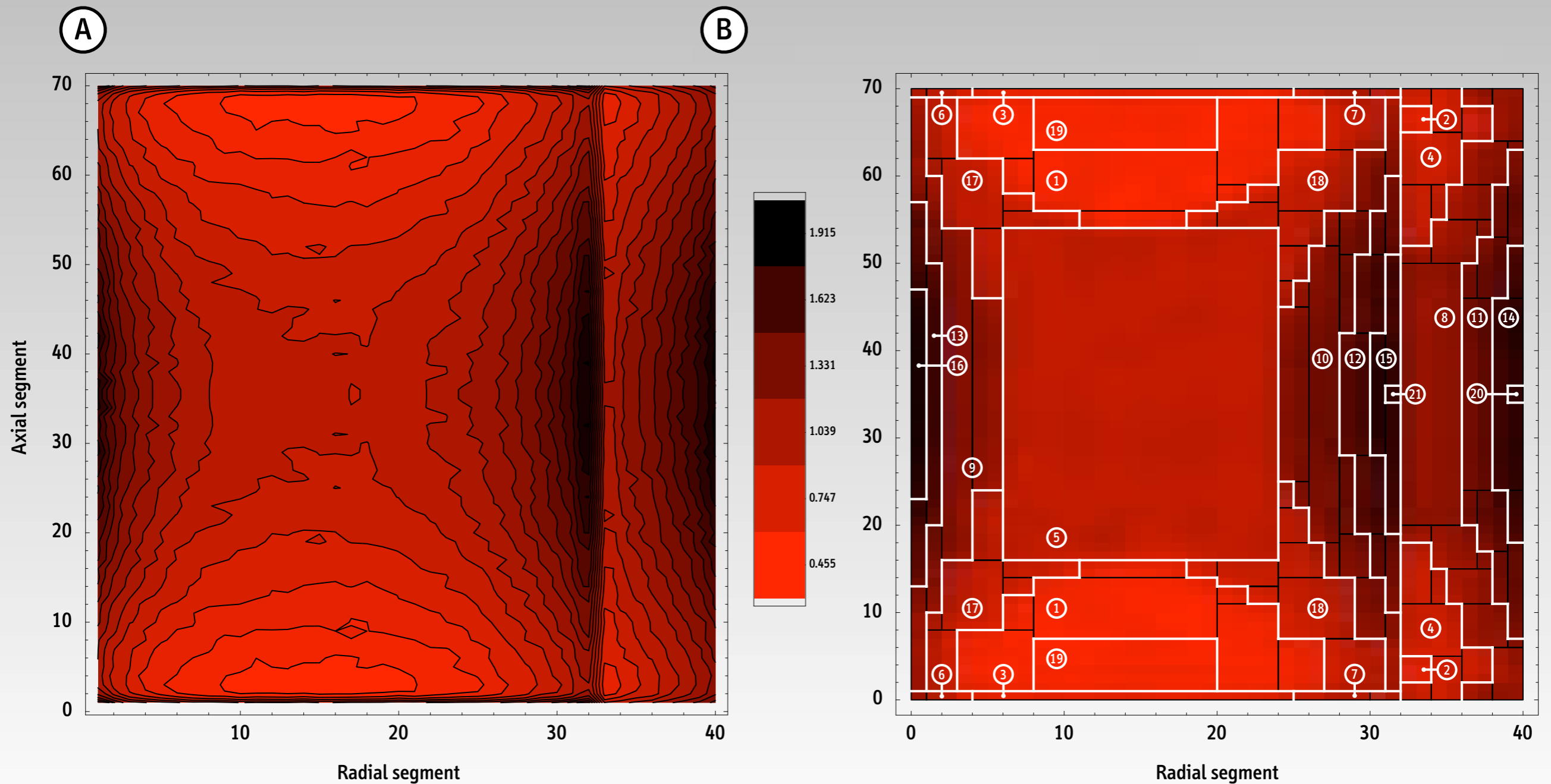


Source: BWXT

Adaptive Cell Structure for FRM-II for Burnup Calculations



Adaptive Cell Structure for FRM-II for Burnup Calculations

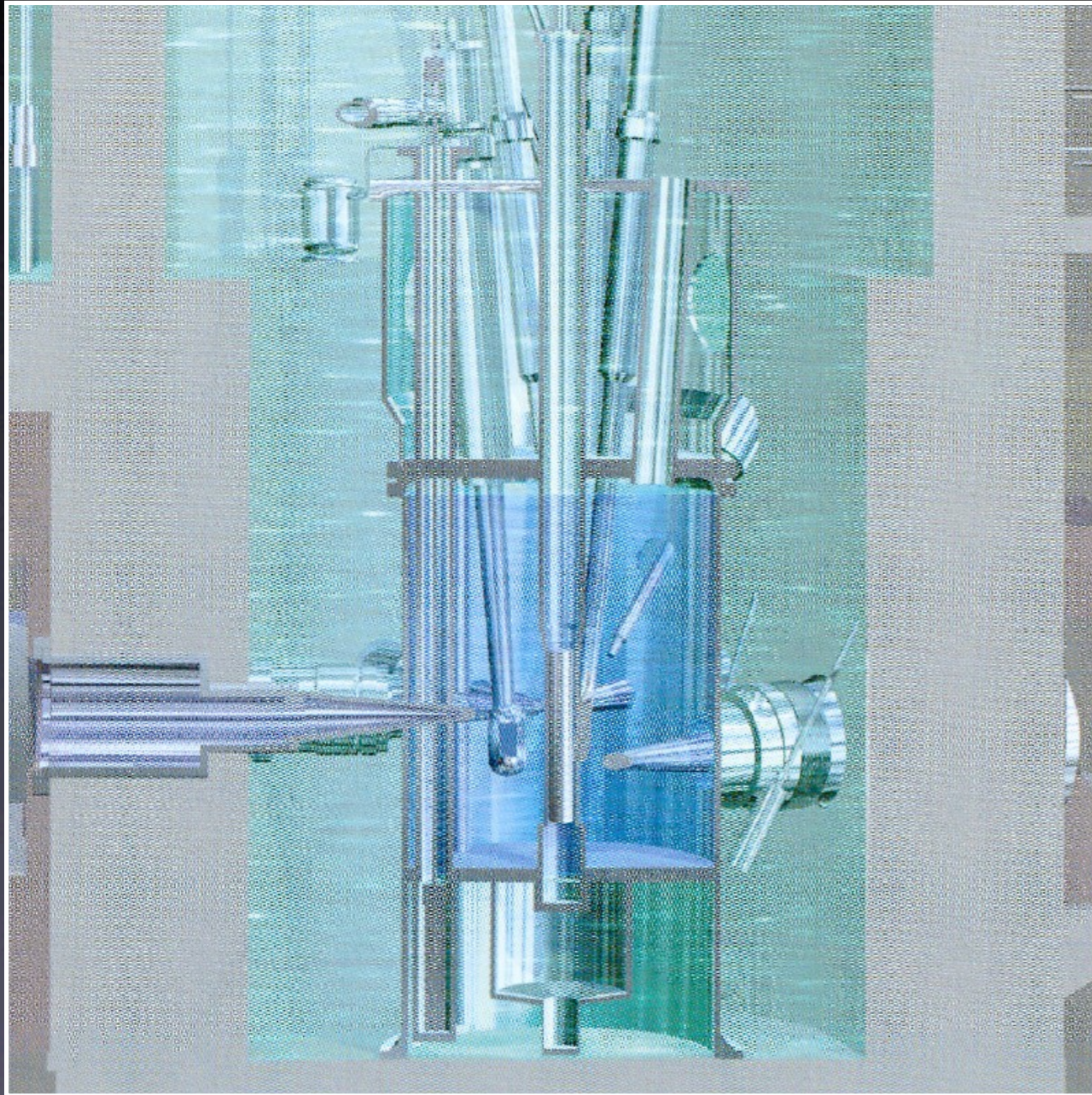


Fallbeispiel FRM-II



Forschungsreaktor München II (FRM-II), 2004

Technische Daten FRM-II



Thermische Leistung: 20 MW

Kühlmittel: H₂O, Moderator: D₂O

Brennstoffinventar: 8.1 kg Uran (93% U-235)

Durchmesser des Reaktorkerns: 24.3 cm

Höhe des Reaktorkerns: 70.0 cm

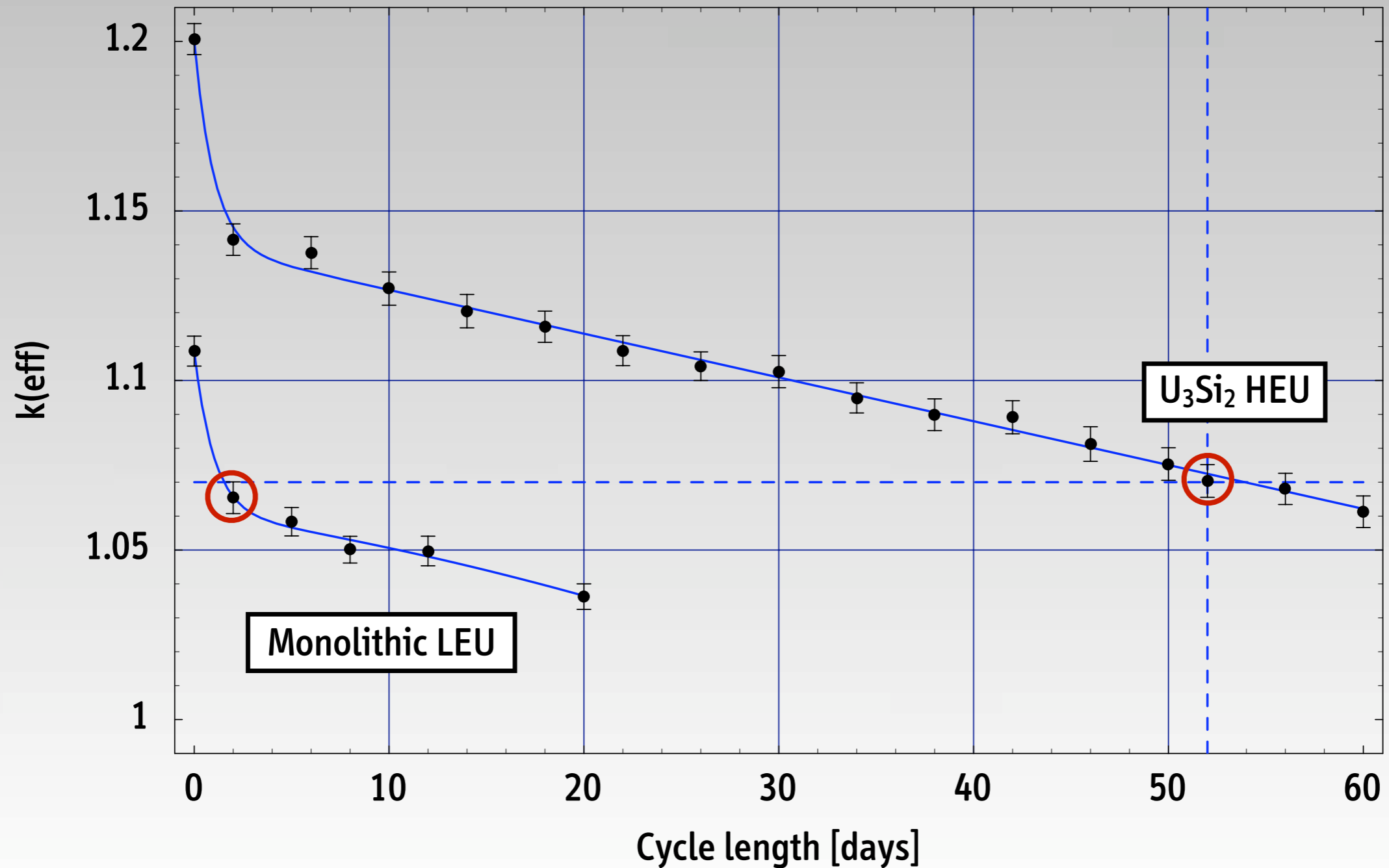
Wesentliche Kenndaten:

Zykluslänge: 52 Tage

Maximaler th. Neutronenfluss: $8.0 \cdot 10^{14} \text{ n/cm}^2\text{s}$

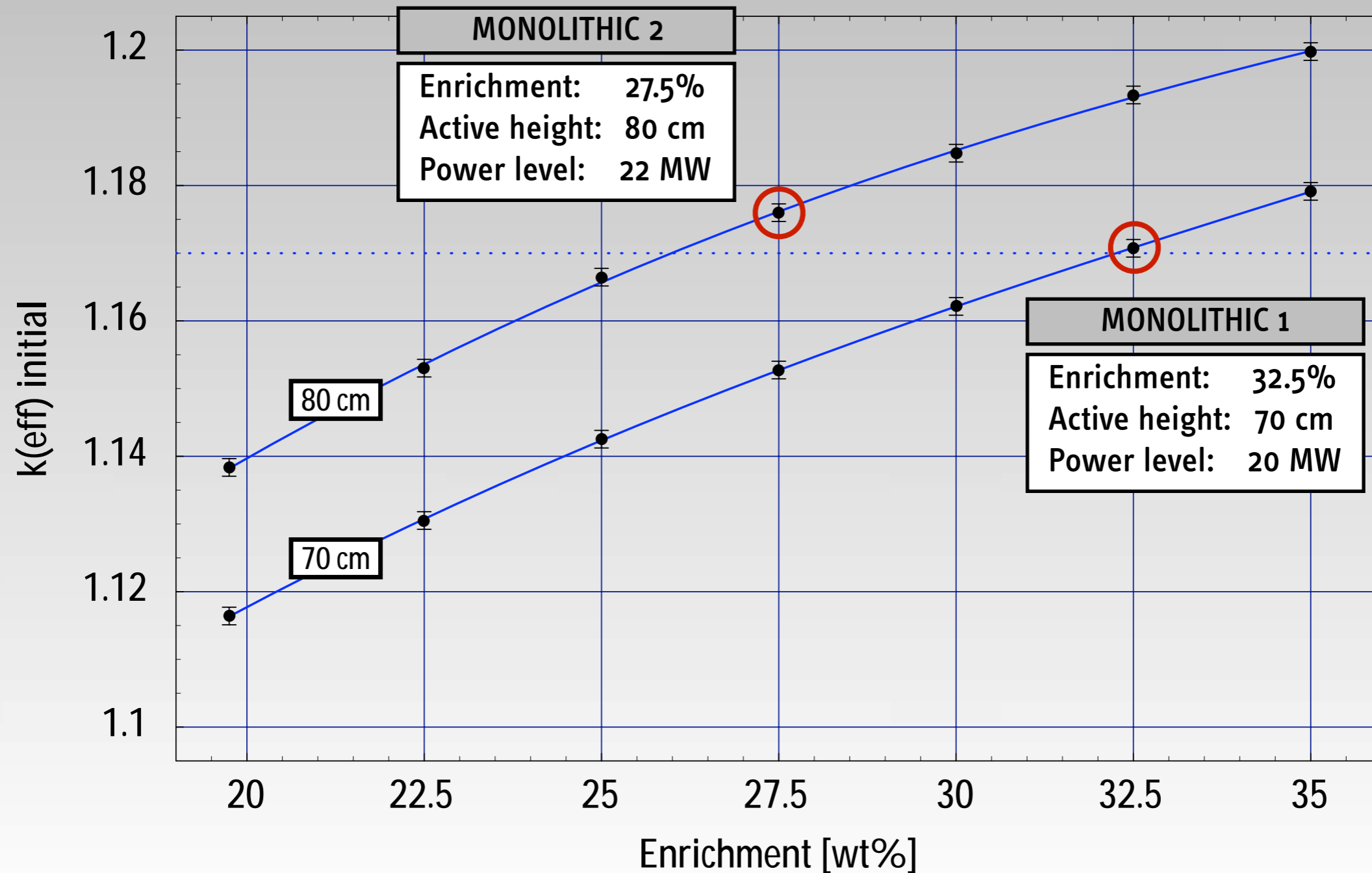
FRM-II Cycle Length

Standard HEU fuel (93%) vs monolithic LEU in original geometry



Initial $k(\text{eff})$ versus Enrichment

Modification: Height of fuel element



Optimization of Single Element Reactor Performance Using the Linear Programming Technique

Linear Programming Technique

(n independent variables and m constraints)

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

First suggested for research reactor optimization by S.C. Mo, RERTR 1991

Solution with Simplex algorithm

Design Variables and Constraints

x1 : Meat thickness

x2 : Cladding thickness

x3 : Coolant channel width

x4 : Inner core radius

x5 : Outer core radius

x6 : Active height of fuel plate

x7 : Fuel enrichment

x8 : Transition radius

x9 : Discontinuity factor

ϕ : Max. thermal neutron flux

C1 : Cycle length

C2 : Average power density

C3 : Average heat flux

C4 : Power peaking factor 1

C5 : Power peaking factor 2

Objective Function and Constraints

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

Linearization of thermal neutron flux (objective function)

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

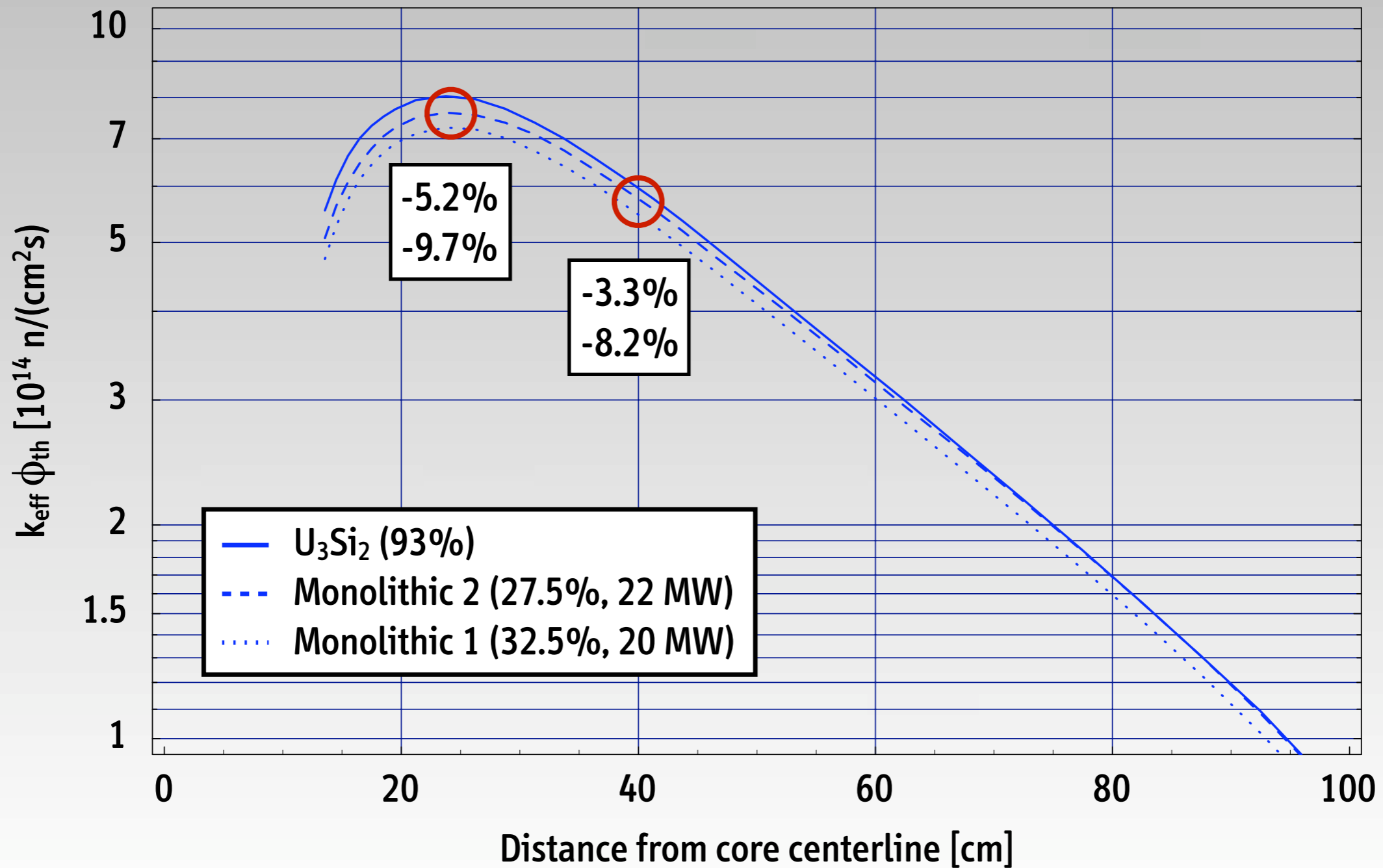
Linearization of constraints

Partial derivatives determined with MCNP (or exactly)

Main Results for Option 2

| | FRM-II Monolithic 2 | | |
|--------|---|-----------------------------|-----------------------------|
| | Enrichment: 27.5 wt% | | |
| | Active core height: 80 cm; Power level: 22 MW | | |
| | Start | LP Solution | Verification |
| x_1 | 0.60 mm | 0.60 mm | |
| x_3 | 2.20 mm | 2.70 mm | |
| x_8 | 10.56 cm | 10.48 cm | |
| x_9 | 0.50 | 0.49 | |
| Plates | 113 | 100 | |
| k(eff) | 1.174 ± 0.001 | 1.182 | 1.180 ± 0.001 |
| ϕ | 7.56E14 n/cm ² s | 7.57E14 n/cm ² s | 7.62E14 n/cm ² s |
| C_1 | 52 days | 52 days | 52 days |
| C_2 | 985 kW/cc | 985 kW/cc | 985kW/cc |
| C_3 | 175 W/cm ² | 200 W/cm ² | 200 W/cm ² |
| C_4 | 1.73 | 1.67 | 1.66 |
| C_5 | 2.08 | 2.00 | 1.99 |

Thermal Neutron Flux



Wissenschaftliche Nutzbarkeit

Fallbeispiel FRM-II

Relative Acquisition Time

Signal-to-noise ratio: $f = \frac{N_{\text{sig}}}{N_{\text{noi}}} = \frac{\phi_{\text{th}}}{C + \phi_{\text{fast}}}$

Proposed by Axmann et al., TUM, 1999

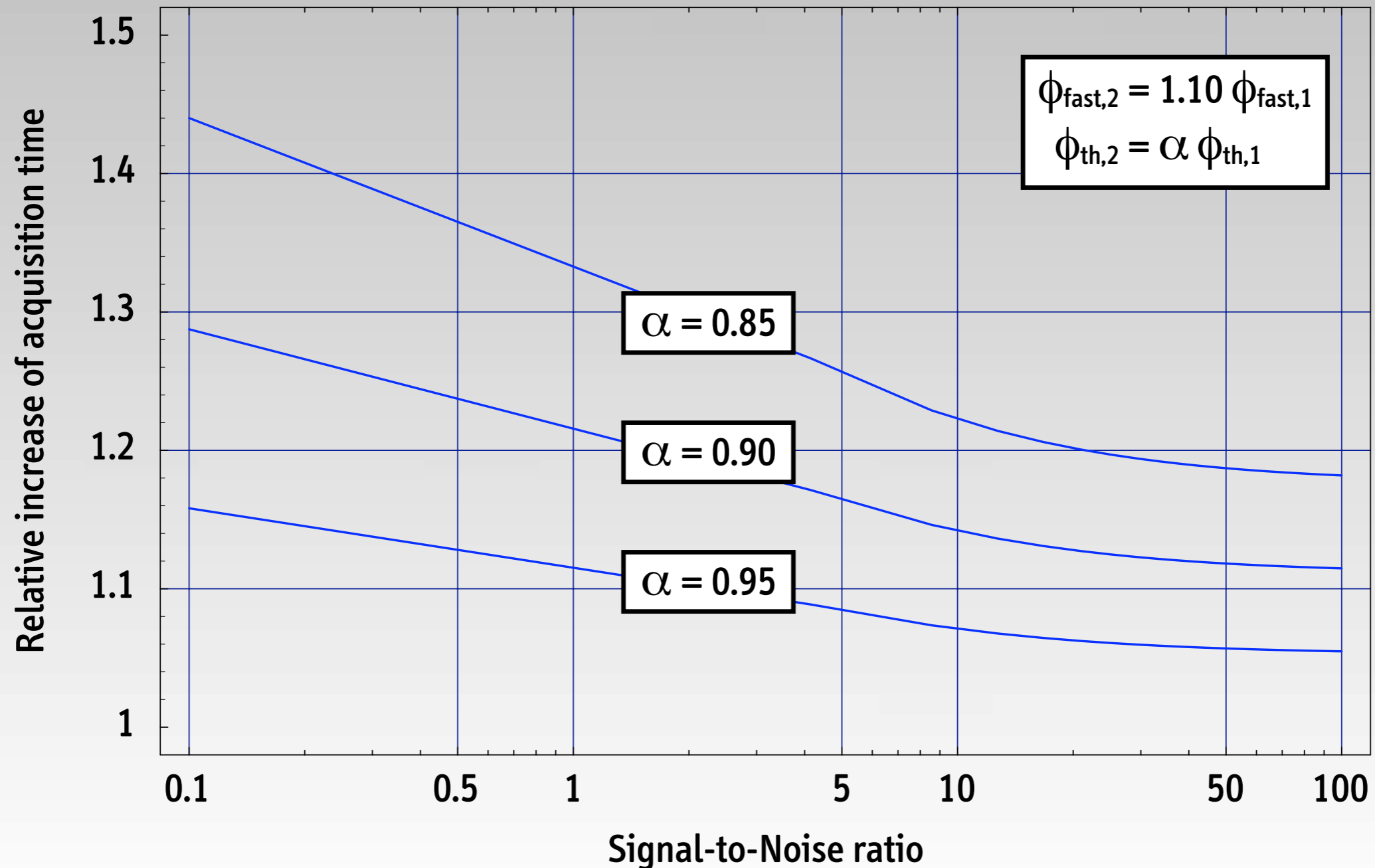
$$\phi_{\text{th},2} = \alpha \phi_{\text{th},1} \quad \text{and} \quad \phi_{\text{fast},2} = \beta \phi_{\text{fast},1}$$

$$\frac{t_2}{t_1} = \frac{1}{\alpha} \left[1 + \left(\frac{1 + \beta}{\alpha f} \right) \right] \cdot \left[1 + \left(\frac{2}{f} \right) \right]^{-1}$$

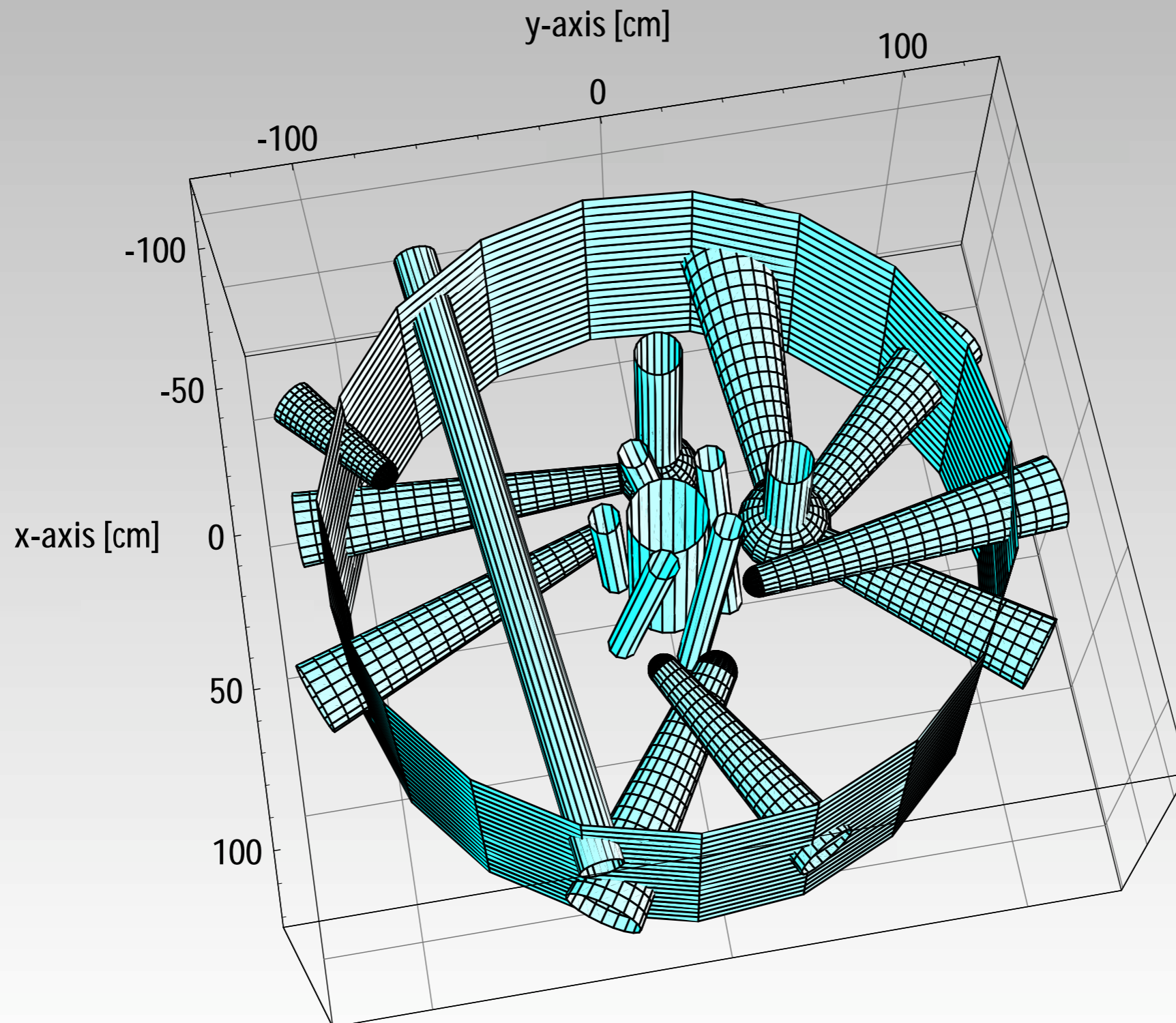
Relative acquisition time to achieve the same relative error of data

Increase of Required Beam Time

for constant relative error, exemplary data

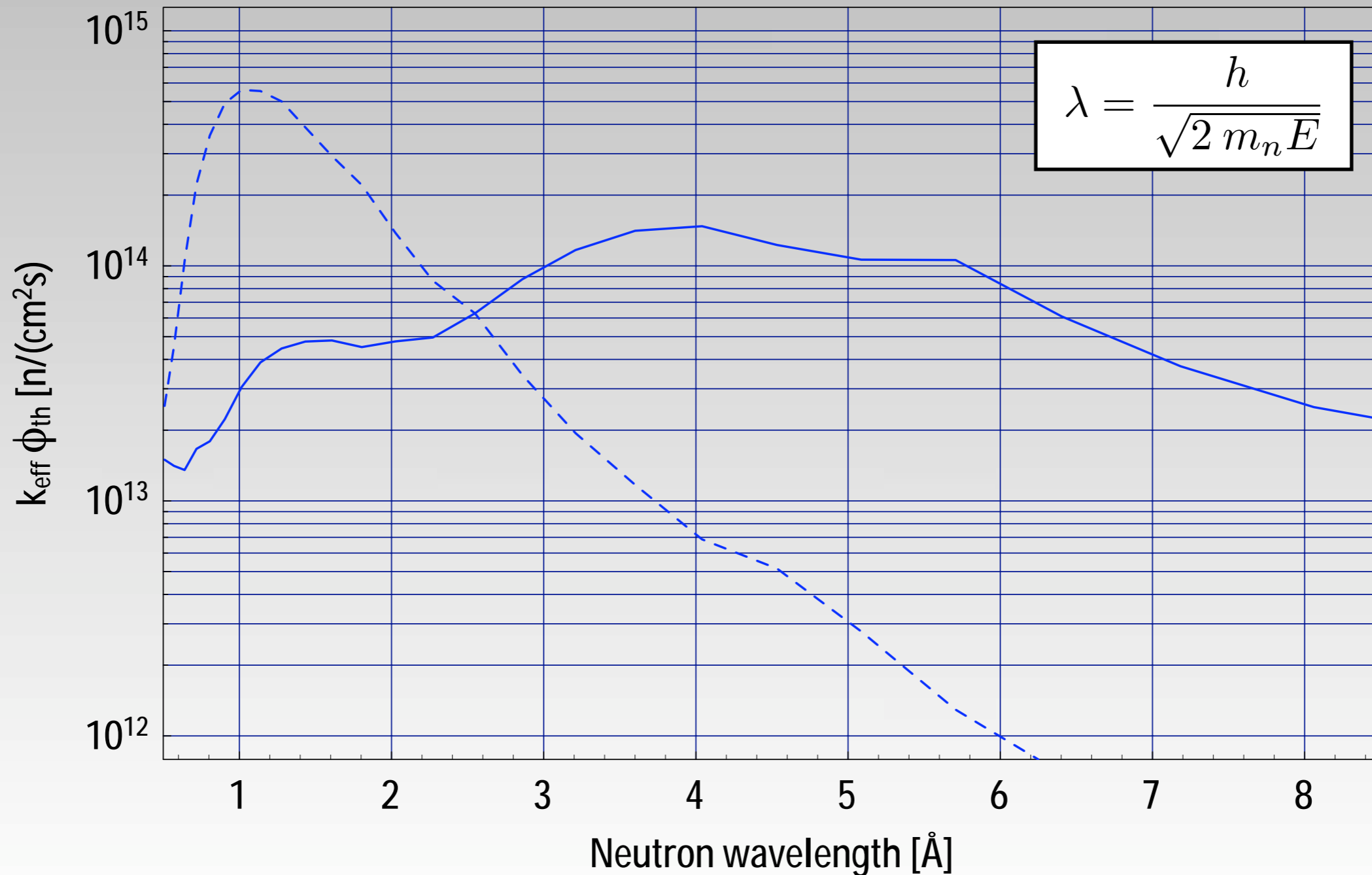


MCNP Model of FRM-II Tank



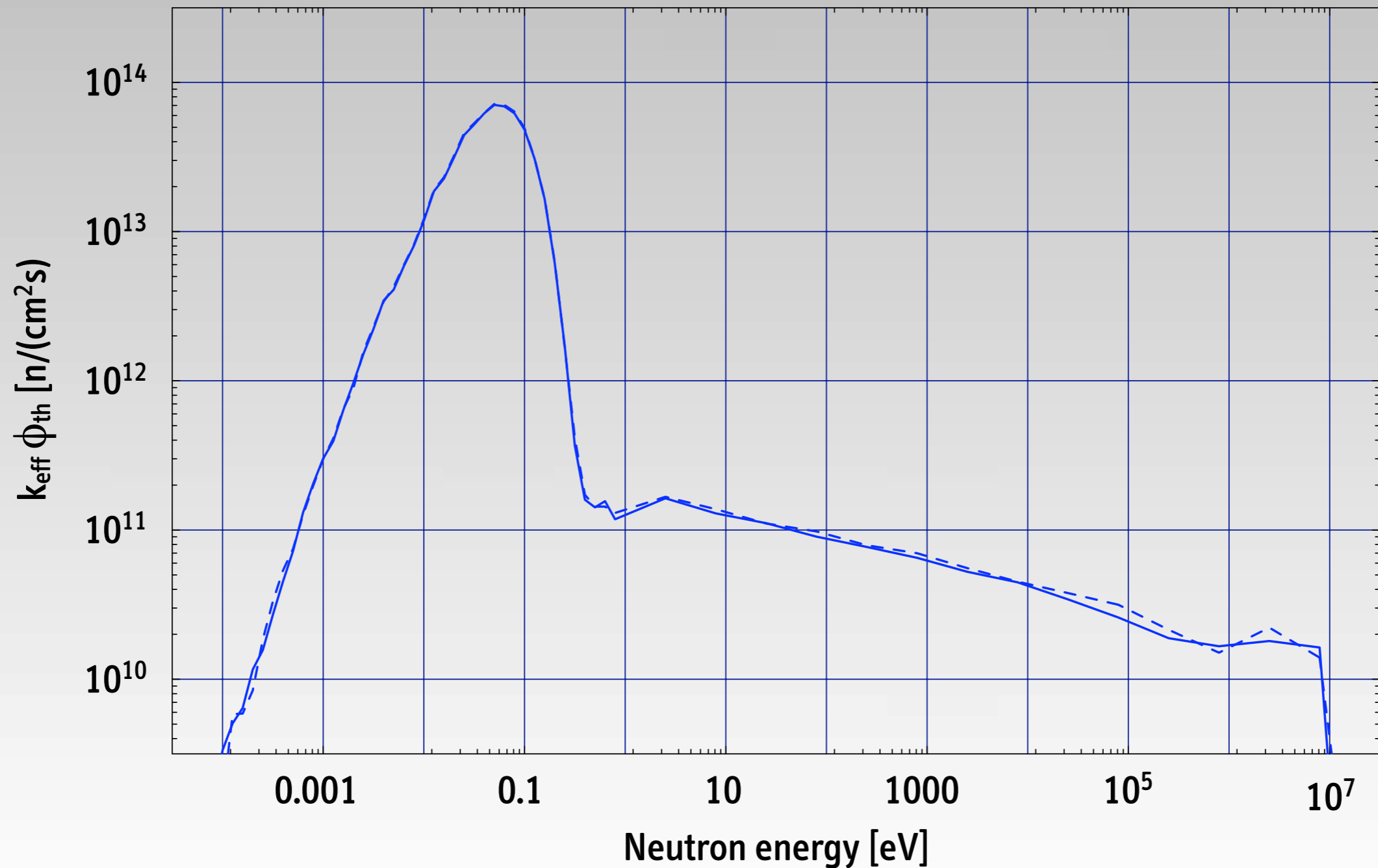
Neutron Wavelength in CNS

HEU design of FRM-II, liquid deuterium at 20 K



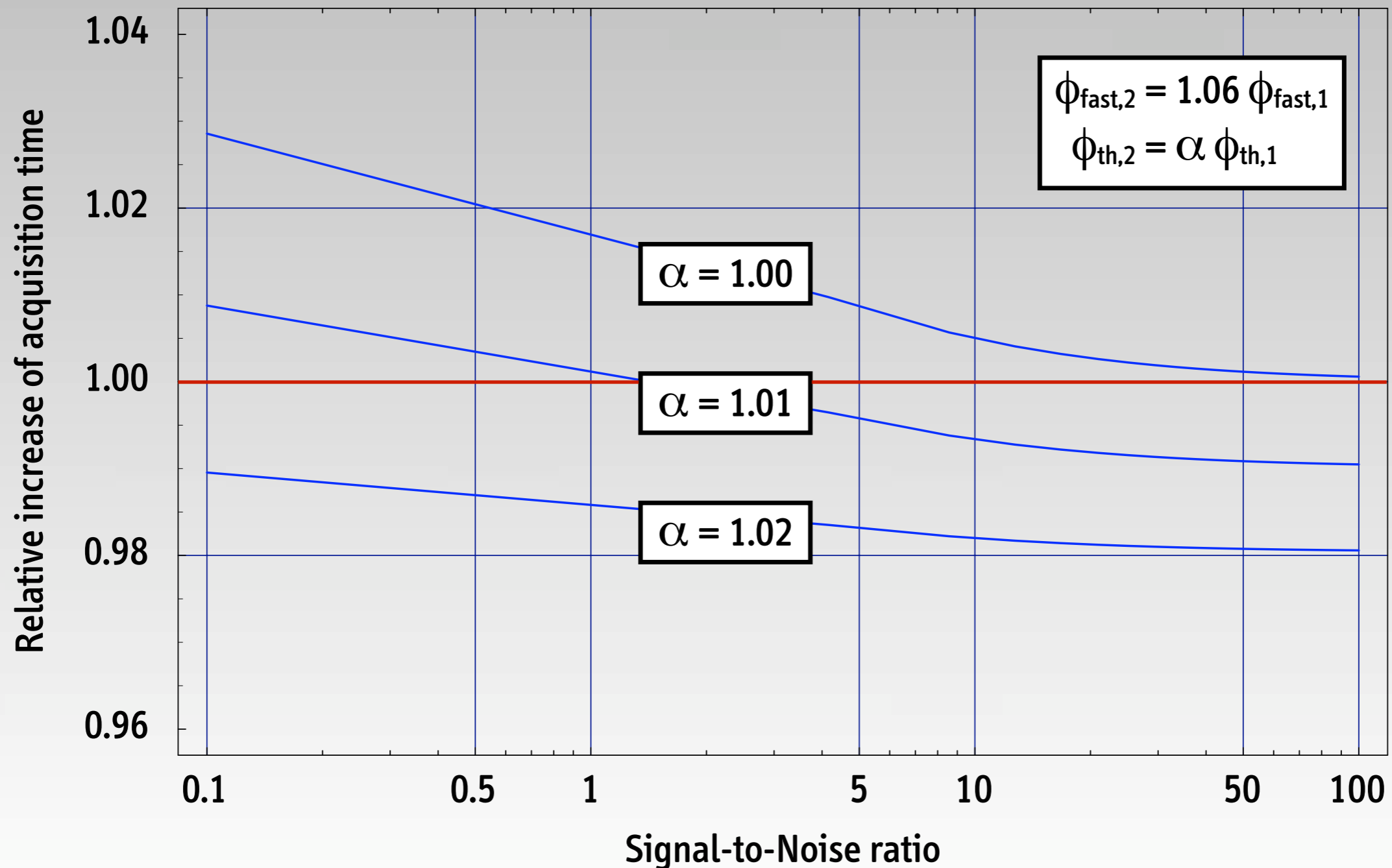
Neutron Spectrum in Beam Tube 1

(measured at -70 cm in beam direction, preliminary data)



Increase of Required Beam Time

for constant relative error, HEU vs Monolithic 2 in FRM-II



Schlussfolgerungen und Ausblick

Monolithische Brennstoffe würden enormes Potential zur Umstellung von Forschungsreaktoren bieten

FRM-II stellt die grösste Herausforderung bezüglich Anreicherungsreduktion dar, allerdings wäre Einsatz von monolithischem Brennstoff mit $< 30\%$ möglich
(Alle anderen HFR's sollten auf LEU umstellbar sein)

Typische Probleme bei der Umstellung auf monolithischen Brennstoff: Anfangsreaktivität und Power Peaking

Methoden der Linearen Programmierung kombiniert mit Monte Carlo "Störungsrechnungen" können mit zunehmender Rechnerleistung (weiter) an Bedeutung gewinnen

