

Article



# **Reducing Irradiation Damage in a Long-Life Fast Reactor with Spectral Softening**

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Abstract: Long-life fast reactors receive considerable attention for their potential of using uranium efficiently, and because they can operate for extended periods without refueling. However, the main obstacle to achieving maximum operating times and fuel burnup is the neutron radiation damage that accumulates in the cladding and structural materials. Simulations of metal-fueled high-burnup fast reactors showed that the damage in these reactors' cladding material reached 200 displacements per atom (dpa) long before the maximum burnup was achieved. One possibility for overcoming this problem is spectral softening, which would reduce the kinetic energy imparted to reactor materials when neutrons collide with them. In this work, we compared the peak irradiation damage in metal-and oxide-fueled fast reactors with that in equivalent reactors containing beryllium in the fuel and reflectors. We showed that the peak damage to the cladding in a metal-fueled reactor was reduced from 273 dpa to 230 dpa when beryllium was included in the core. In an oxide-fueled reactor, the peak damage to the cladding was reduced from 225 dpa to 203 dpa. All four reactors were operated with a core-average burnup of 112 MWd/kg of initial heavy metal (IHM), without reshuffling or refueling, and contained the same initial actinide mass profiles.

Keywords: fast reactor; long-life; irradiation damage; displacements per atom (dpa)

# 1. Introduction

The sustainability of nuclear power depends in large part on its economic competitiveness and its long-term impact on the environment. Fast-neutron spectrum reactors can achieve high levels of fuel burnup, which increases both the amount of electricity that a unit mass of fuel can produce, and the burnout of transuranic nuclides which give nuclear waste its long-term radioactive signature. Long-life fast reactors have been of interest for decades, in part because of their potential to run for extended periods without refueling, and to operate as a "nuclear battery" which could reduce both the cost and the proliferation risks associated with nuclear energy [1].

Advanced fast-reactor designs can achieve peak burnup values of over 500 MWd/kg of initial heavy metal (IHM) before the accumulation of fission products stops the chain reaction [2]. However, the main limiting factor in achieving maximum fuel burnup in fast reactors is the neutron damage to core structural and cladding materials. High-energy neutrons collide with atoms in these materials, and displace them from their equilibrium lattice positions. Over time, this leads to macroscopic changes in the materials, such as swelling and embrittlement, which can cause the core to deform, and the cladding to rupture. Studies of proposed Generation-IV fast-reactor systems showed that operating these cores produces damage levels ranging from 200 displacements per atom (dpa) to 250 dpa [3] in the cladding. At present, the best candidate material for these applications, HT-9 steel, was demonstrated to have damage levels of only ~200 dpa at temperatures below 873 K [4,5].

Advanced materials that could withstand higher dpa levels, and maintain their mechanical properties are in development [6]; however, they are unlikely to be available for practical use anywhere in the near term.

Past work showed that spectral softening could be used to reduce irradiation damage in a fission wave reactor [7]. The softer spectrum reduces the kinetic energy imparted to reactor materials when neutrons collide with them. Here, we compared the dpa levels in long-life fast-reactor cores with and without beryllium in the fuel and reflectors. Beryllium was proposed as an additive to uranium dioxide fuel for light-water reactors in order to improve its thermal conductivity [8], and beryllium reflectors were used in studies of liquid-metal-cooled fast reactors to improve their safety [9,10]. We showed that the addition of beryllium reduced the peak dpa in a mixed-oxide-fueled core from 225 dpa to 203 dpa, and in a metal-fueled core from 273 dpa to 230 dpa. In all cases, the average burnup level was 112 MWd/kgIHM, and was achieved without the refueling or reshuffling of fuel. All cores had the same initial mass of actinides, and fuel densities were adjusted where appropriate to accommodate the beryllium. A high degree of core-average burnup was achieved by axially and radially grading the initial plutonium content of the fuel so as to flatten the power profile. The cores included diluent regions of sodium to reduce neutron flux in the regions that otherwise experienced the greatest irradiation damage.

#### 2. Results

The biggest obstacle to the practical implementation of a long-life fast reactor is the high degree of irradiation damage to in-core materials. However, spectral softening, and the use of diluent regions to reduce the peak neutron fluence to materials can be used in combination to overcome both problems. In the core designs presented in this work, a reduction in the relative population of high-energy neutrons was achieved through the use of beryllium-doped oxide fuel and beryllium reflectors. The axial and radial grading of the fresh fuel, along with the beryllium reflectors and a diluent region, flattened the power profile and allowed for a high core-average burnup before maximum allowable dpa levels were reached.

Figure 1 shows the dpa levels in the metal-fueled core, with sodium in the reflector regions and no beryllium in the fuel. The maximum cladding dpa occurred close to the inner region, and reached a level of 273 dpa at 8.4 years. The maximum damage levels were lower close to the reflectors due to lower overall fluence away from the center of the core.



**Figure 1.** Radial and axial displacements per atom (dpa) in the unsoftened metal core. The depression in the center was due to the diluent region. The peak damage at 8.4 years was 273 dpa in assembly region 5 between 180 cm and 220 cm along the axial direction. The dpa was significantly lower near the sodium reflector regions (corresponding to regions 1, 2, and 17–20 in Figure 11) due to a lower overall fluence. The peak dpa in the fuel regions adjacent to the inner and outer reflectors were 232 and 99, respectively.

Figure 2 shows the dpa levels in the metal-fueled core, with beryllium rods in the reflector regions, and 10-atom-percent beryllium doping in the fuel. The maximum cladding damage occurred close to the inner region, and reached a level of 230 dpa at 8.4 years. The maximum damage levels were lower close to the reflectors due not only to lower fluence values, but also to a softer neutron spectrum.

Figure 3 shows the difference in dpa between the metal-fueled core, with beryllium reflectors and beryllium in the fuel, and the metal-fueled core with sodium in the reflector region and no beryllium in the fuel. The greatest reduction in dpa was near the axial and radial centerline where  $\Delta$ dpa was -75.8. The dpa increased by a maximum of 19.9 dpa in the outer radial regions due to the reflectors causing an increase in fission rate; however, in the outer radial regions, the dpa value remained well below 200, as shown in Figure 2.



**Figure 2.** Radial and axial dpa in the softened metal core. The depression in the center was due to the diluent region. The peak dpa at 8.4 years was 230 in assembly region 7 between 80 cm and 100 cm along the radial direction, and between 300 cm and 320 cm along the axial direction. The dpa was significantly lower near the beryllium reflector regions (corresponding to regions 1, 2, and 17–20 in Figure 11) due to both a softer neutron spectrum, and a lower overall fluence. The peak dpa in the fuel regions adjacent to the inner and outer reflectors were 161 and 117, respectively.



**Figure 3.** Changes in dpa in due to spectral softening in the metal core. The core centerline experienced the greatest reduction in dpa, with a corresponding axial position of 180 cm to 220 cm (assembly region 3 in Figure 11). The rise in dpa at the outer edge of the core was due to neutrons being reflected back into the core when a beryllium reflector was present.

Figure 4 shows the dpa levels in the mixed-oxide-fueled core, with sodium in the reflector regions and no beryllium doping in the fuel. The maximum cladding damage occurred close to the inner region, and reached a level of 225 dpa at 8.4 years. The maximum damage levels were lower close to the reflectors due to lower overall fluence away from the center of the core. The overall damage levels were lower relative to either of the metal-fueled cores due to the presence of oxygen atoms which produced a softer neutron spectrum.



**Figure 4.** Radial and axial dpa in the unsoftened mixed-oxide core. The depression in the center was due to the diluent region. The peak dpa at 8.4 years was 225 in assembly region 6 between 80 cm and 100 cm along the radial direction, and between 300 cm and 320 cm along the axial direction. The dpa was significantly lower near the beryllium reflector regions (corresponding to regions 1, 2, and 17–20 in Figure 11) due to a lower overall fluence. The peak dpa in the fuel regions adjacent to the inner and outer reflectors were 181 and 73, respectively.

Figure 5 shows the dpa levels in the mixed-oxide-fueled core, with beryllium rods in the reflector regions and a beryllium doping of 10 atom percent in the fuel. The maximum cladding dpa occurred in the vicinity of the diluent, and reached a level of 203 dpa at 8.4 years. The maximum damage levels were lower close to the reflectors due not only to lower flux values, but also a softer neutron spectrum. Here, the presence of oxygen and beryllium in combination lowered the average neutron energy even further.

Figure 6 shows the difference in dpa between the oxide-fueled core with beryllium reflectors and beryllium in the fuel, and the oxide-fueled core with sodium in the reflector region and no beryllium in the fuel. The greatest reduction in dpa was near the axial and radial centerline, where  $\Delta$ dpa was -61.6. The dpa increased by a maximum of 19.0 dpa in the outer radial regions due to the reflectors causing an increase in fission rate; however, in the outer radial regions, the dpa value remained well below 200, as shown in Figure 5.

Figure 7 shows the neutron flux in the mixed-oxide-fueled core, with beryllium rods in the reflector regions and a beryllium doping of 10 atom percent in the fuel. The profiles are shown at t = 0 years, and t = 8.4 years, the time at which peak cladding damage hit 203 dpa. The increase in flux in the central region of the reactor was clearly evident at 8.4 years, and was due to an increase in transuranic content in that region of the reactor. The axial and radial plutonium grading, along with the diluent and reflectors, produced a shifting flux distribution that produced a more even burnup profile. This allowed for a high core-average burnup before maximum dpa levels were reached, without the need for reshuffling or reforming the fuel. Plots of neutron flux and dpa levels, including error bars reflecting Monte Carlo statistical uncertainty, can be found in Supplementary Note 1.



**Figure 5.** Radial and axial dpa in the softened mixed-oxide core. The depression in the center was due to the diluent region. The peak dpa at 8.4 years was 203 in assembly region 7 between 80 cm and 100 cm along the radial direction, and between 300 cm and 320 cm along the axial direction. The dpa was significantly lower near the beryllium reflector regions (corresponding to regions 1, 2, and 17–20 in Figure 11) due to both a softer neutron spectrum, and a lower overall fluence. The peak dpa in the fuel regions adjacent to the inner and outer reflectors were 130 and 91, respectively.



**Figure 6.** Changes in dpa in due to spectral softening in the oxide core. The core centerline experienced the greatest reduction in dpa, with a corresponding axial position of 180 cm to 220 cm (assembly region 3 in Figure 11). Again, the rise in dpa at the outer edge of the core was due to neutrons being reflected back into the core when a beryllium reflector was present.



**Figure 7.** Three dimensional (3D) neutron flux profiles in the softened mixed-oxide core. (**a**) Neutron flux at the beginning of life (BOL). Peak flux was  $2.76 \times 10^{15} \text{ n/cm}^2$ -s in assembly region 7 between 80 cm and 100 cm along the radial direction, and between 300 cm and 320 cm along the axial direction. (**b**) Neutron flux at the end of life (EOL), 8.4 years. Peak flux was  $2.79 \times 10^{15} \text{ n/cm}^2$ -s in assembly region 7 between 180 cm and 220 cm along the axial direction. The flux gradually changed shape due to the increasing concentration of Pu in the inner region of the core.

Figure 8 shows the position-dependent burnup profile, as well as the uranium utilization at 8.4 years in the mixed-oxide-fueled core, with beryllium rods in the reflector regions and a beryllium doping of 10 atom percent in the fuel. The maximum fuel burnup at this point was 163.4 MWd per kg of initial heavy metal, which occurred in the vicinity of the diluent. The core-average burnup at this time was 112 MWd per kg of initial heavy metal, with a minimum of 22.3 MWd per kg of initial heavy metal in the upper and lower axial locations, and outermost radial locations. The center region also experienced the greatest uranium burnup—21.6 atom percent of the initial depleted uranium. The core-average burnup was 14.7 atom percent of the initial uranium, fixed by the power,

burn duration, and initial uranium mass of the core. This diminished to 2.7 atom percent in the upper and lower axial regions.



**Figure 8.** Position-dependent fuel burnup at 8.4 years in the softened mixed-oxide core. (**a**) Overall burnup. Peak burnup was 163.4 MWd/kg of initial heavy metal (IHM) in assembly region 7 between 80 cm and 100 cm, and between 300 cm and 320 cm along the axial direction. Core-averaged burnup was 112 MWd/kgIHM. (**b**) Uranium burnup. The peak uranium burnup fraction was 21.6 a/o in assembly region 8 between 100 cm and 120 cm along the radial direction, and between 280 cm and 300 cm along the axial direction. The core-averaged uranium burnup fraction was 14.7 a/o. The diluent is shown in the empty region in both figures where no burnup was experienced.

Figure 9 shows the neutron multiplication factor as a function of time in the mixed-oxide-fueled core, with beryllium rods in the reflector regions and a beryllium doping of 10 atom percent in the fuel. Axial and radial grading of the fresh core, along with the use of reflectors, had a significant effect on the reactivity of the reactor. The reactor experienced a reactivity swing because of breeding and burning, but it stayed above critical levels throughout the 8.4 years. The core started its life with  $k_{eff} = 1.015$ , peaked at 1.028, and was 1.001 at 8.4 years. The beginning of life  $k_{eff}$  was smaller than that for some conventional fast-reactor designs which run oxide fuel, as was the reactivity swing from the beginning to the end of life [11].



**Figure 9.**  $k_{eff}$  as a function of time in the softened mixed-oxide core. The neutron multiplication factor increased for the first 2.8 years as plutonium was bred in. The peak  $k_{eff}$  was 1.028 at t = 2.8 years. At t = 8.4 years, corresponding to 203 dpa,  $k_{eff}$  = 1.001. The  $k_{eff}$  shown is the average of five burnup simulations of the softened mixed-oxide core using different random number seeds. The error bars are shown for one standard deviation, corresponding to the 68% confidence level. More details on the calculation of uncertainties are given in Supplementary Note 8.

Figure 10 shows the axial temperature profiles of the fuel, the cladding, and the coolant in the hottest radial position at the beginning of life in the mixed-oxide-fueled core, with beryllium rods in the reflector regions and a beryllium doping of 10 atom percent in the fuel. The temperatures in each region remained well below material limits. These temperature profiles were perturbed to estimate the reactivity coefficients at the beginning, middle, and end of life of the core, corresponding to 0 years, 4.4 years, and 8.4 years of operation.

The reactivity coefficients are listed in Table 1. All reactivity coefficients quoted were isothermal, corresponding to spatially uniform changes in fuel or coolant temperatures. The large positive void and the coolant reactivity coefficients were in part mitigated by a large negative Doppler reactivity feedback. The restructuring of oxide fuel with burnup introduced large uncertainties in the estimates of the behavior of the fuel as a function of temperature [12]. Because of this, thermal expansion of the fuel was an unreliable mechanism for additional negative reactivity feedback, and was not included in this analysis.



**Figure 10.** Temperature profiles of the fuel, cladding, and coolant. Blue, green, and red lines represent the temperatures of the fuel, inner cladding, and coolant, respectively. Data represent the axial temperature in assembly region 8 of the reactor which reached the highest fuel temperature—1423 K. The profiles were computed at the beginning of life. The central dip feature was due to the diluent at the *z* = 2 m axial position.

Core	Metal, Unsoftened	Metal, Softened	Mixed Oxide, Unsoftened	Mixed Oxide, Softened	
$\begin{array}{c} \text{Beginning of life} \\ \alpha_{\text{Doppler}} \left( K^{-1} \right) \\ \alpha_{\text{coolant}} \left( K^{-1} \right) \\ \alpha_{\text{void}} \left( \text{per } \% \text{ void} \right) \end{array}$	$\begin{array}{c} -2.7\pm 0.2\times 10^{-6}\\ 9.7\pm 0.2\times 10^{-6}\\ 3.3\pm 0.006\times 10^{-4}\end{array}$	$\begin{array}{c} -5.0\pm 0.2\times 10^{-6}\\ 1.1\pm 0.02\times 10^{-5}\\ 3.5\pm 0.006\times 10^{-4}\end{array}$	$\begin{array}{c} -8.2\pm 0.2\times 10^{-6}\\ 5.3\pm 0.2\times 10^{-6}\\ 1.8\pm 0.007\times 10^{-4}\end{array}$	$\begin{array}{c} -9.5\pm 0.2\times 10^{-6}\\ 7.1\pm 0.2\times 10^{-6}\\ 2.1\pm 0.007\times 10^{-4}\end{array}$	
Middle of life $\alpha_{\text{Doppler}}$ (K <sup>-1</sup> ) $\alpha_{\text{coolant}}$ (K <sup>-1</sup> ) $\alpha_{\text{void}}$ (per % void)	$\begin{array}{c} -2.0\pm 0.2\times 10^{-6}\\ 1.2\pm 0.06\times 10^{-5}\\ 4.1\pm 0.006\times 10^{-4}\end{array}$	$\begin{array}{c} -4.0 \pm 0.2 \times 10^{-6} \\ 1.3 \pm 0.02 \times 10^{-5} \\ 3.8 \pm 0.006 \times 10^{-4} \end{array}$	$\begin{array}{c} -6.0 \pm 0.2 \times 10^{-6} \\ 7.7 \pm 0.2 \times 10^{-6} \\ 2.7 \pm 0.007 \times 10^{-4} \end{array}$	$\begin{array}{c} -6.9 \pm 0.2 \times 10^{-6} \\ 7.7 \pm 0.2 \times 10^{-6} \\ 2.6 \pm 0.007 \times 10^{-4} \end{array}$	
End of life $\alpha_{\text{Doppler}} (\text{K}^{-1})$ $\alpha_{\text{coolant}} (\text{K}^{-1})$ $\alpha_{\text{void}} (\text{per }\% \text{ void})$	$\begin{array}{c} -2.3\pm 0.2\times 10^{-6}\\ 1.3\pm 0.02\times 10^{-5}\\ 4.4\pm 0.007\times 10^{-4}\end{array}$	$\begin{array}{c} -2.7\pm 0.2\times 10^{-6}\\ 1.3\pm 0.02\times 10^{-5}\\ 4.0\pm 0.007\times 10^{-4}\end{array}$	$\begin{array}{c} -4.7 \pm 0.2 \times 10^{-6} \\ 8.5 \pm 0.2 \times 10^{-6} \\ 3.0 \pm 0.007 \times 10^{-4} \end{array}$	$\begin{array}{c} -5.5\pm0.2\times10^{-6}\\ 8.9\pm0.2\times10^{-6}\\ 2.9\pm0.007\times10^{-4} \end{array}$	

**Table 1.** Reactivity coefficients. The coefficients were computed by perturbing the fuel temperature only, the coolant temperature, and the coolant density only.

# 3. Discussion

Simulations of long-life fast-reactor cores were done using Serpent 2.1.28, and confirmed with MCNPX 2.7.0. The metal-fueled core without beryllium experienced the greatest peak damage to cladding (273 dpa), which was brought down to 230 dpa when the fuel was doped with beryllium, and when beryllium reflectors were added. The mixed-oxide-fueled core without beryllium experienced a peak cladding damage level of 225 dpa, which was brought down to 203 dpa when the fuel was doped with beryllium, and when beryllium, and when beryllium reflectors were added. This showed that, by configuring a fast-reactor core with light elements, the dpa could be reduced by 25%.

The reactor that experienced the least cladding damage was configured with an oxide fuel doped with 10 atom percent beryllium, and with plutonium content, graded axially and radially. When coupled with beryllium reflectors and a sodium diluent, this enabled the simulated reactor to operate for 8.4 years at a thermal output of 4740 MW, with a peak cladding damage of 203 dpa.

The reactor developed an average uranium burnup of 14.7 atom percent of the initial uranium after 8.4 years. The average overall burnup was 112 MWd per kg of initial heavy metal. This was achieved without reshuffling or recladding the fuel, and without any additional input of fresh fuel. Crucially, this performance was for an unoptimized core, and was achievable with materials for which there was operational history. By using an optimized fuel shuffling strategy, the overall and uranium burnup values would be increased significantly. The results assumed that all fission products remained in the fuel. In reality, a significant fraction of the fission product gas would either escape into a plenum, or leave the core altogether in vented-fuel-pin designs. The partial removal of fission products would produce a small increase in criticality. While venting fission products to the sodium coolant with subsequent removal was proposed for high-burnup fast reactors [13], the implications for safety and licensing should be addressed for any design using this operating strategy.

The simulations which are shown here indicated that Doppler broadening in the fuel could compensate for the positive coolant temperature reactivity effect at the beginning of life in spectrally softened cores. However, for the simulations shown here, this was no longer the case after the middle of life. Additionally, the results in Table 1 show that the core suffered from a large void reactivity. One approach to mitigating these issues would be to use a gas expansion module system, which injects neutron poison into the core when the coolant heats up. These types of control devices were successfully demonstrated at the Fast Flux Test Facility [12]. A similar approach using a lithium injection device was proposed for reactivity control in breed-burn fast reactors [14], and transient analyses showed that this type of device could enable fast reactors with large positive coolant temperature reactivity coefficients to operate safely [15]. Additionally, mechanisms such as grid-plate expansion and axial fuel expansion could help reduce the large coolant temperature reactivity coefficient in both metal- and oxide-fueled cores. However, these mechanisms were not included in this work as their effect on the reactivity coefficients was found to be small relative to Doppler broadening, and coolant thermal expansion or voiding, for metal-fueled sodium-cooled fast reactors with low leakage [14]. Axial fuel-rod expansion was also understood to be an unreliable mechanism for reactivity control in oxide-fueled cores, due to the lack of structural integrity at high levels of burnup [12]. However, these effects, as well as other secondary effects such as control-rod driveline expansion and assembly bowing, should be investigated, as they could produce a strong negative reactivity feedback when combined.

Importantly, the insertion of moderating materials into a fast-reactor core introduced side effects, such as disturbing the power profile of the core, as well as significantly reducing the neutron multiplication factor. These effects were consistent with past studies on fast reactors operating with additional moderating materials (e.g., References [16] and [17]). However, the usual limiting factor in achieving high burnup in sodium-cooled fast reactors is not reactivity, but rather, damage to cladding [12,18]. Figure 9 shows that  $k_{eff}$  remained above 1 when the cladding damage reached 200 dpa. More details on these specific effects are provided in Supplementary Note 2. Additionally, while the densities of fuel in this work were adjusted to accommodate mixing in beryllium, the effect of this on other thermo-physical properties of the fuel, as well as fuel performance under irradiation and burnup, was not addressed. Although experimental data on the thermo-physical properties of fresh UO<sub>2</sub>-BeO pellets exist [8], the optimal moderator composition and geometry need to be explored. Moderator materials, such as boron carbide (enriched to 99% <sup>11</sup>B to limit neutron capture) and zirconium hydride, are options with significant operational experience, and are known to improve the safety characteristics of sodium-cooled fast reactors [16,17].

#### 4. Materials and Methods

Four core designs were simulated: (i) a metal-fueled (U-Pu-Zr) core with sodium reflectors (an unsoftened metal core); (ii) a metal-fueled, beryllium-doped (U-Pu-Zr-Be) core with beryllium reflectors (a softened metal core); (iii) a mixed-oxide-fueled core (an unsoftened mixed oxide core); and (iv) a mixed-oxide-fueled, beryllium-doped core with beryllium reflectors (a softened mixed oxide core). Each core had an identical geometry and mass of actinides in order to compare the effect of spectral softening on an even basis. The main design parameters of the simulated cores are given in Table 2.

**Table 2.** Design parameters for all four cores. The fuel densities shown were the actual densities used in the simulation. The fuel density of the softened mixed oxide core was computed by adding mixed-oxide fuel at a density of  $11 \text{ g/cm}^3$  with beryllium at  $1.85 \text{ g/cm}^3$  in a density-weighted linear fashion, given 9.675 a/o Be and assuming a 95% smear density. The fuel densities of the softened metal core and the unsoftened cores were computed using the softened mixed-oxide core fuel as a basis, and by requiring that the overall mass of actinides in each core was the same.

Core	Metal, Unsoftened	Metal, Softened	Mixed Oxide, Unsoftened	Mixed Oxide, Softened		
Power (MWth)	4740					
Number of assemblies	1039					
Fuel form	U-Pu-Zr	U-Pu-Zr-Be	Mixed oxide (MOX)	MOX-Be		
Beryllium a/o, relative to actinides	0%	9.675%	0%	9.675%		
Zirconium w/o, overall	10%	10%	0%	0%		
Fuel density (g/cm <sup>3</sup> )	9.95	9.98	10.16	10.26		
Fuel/coolant/void/structural volume ratios	42.18/25.24/7.44/25.13%					
Inlet coolant temperature (K)	659					
Core flat-to-flat diameter (cm)	401.66					
Assembly flat-to-flat diameter (cm)	11.5917					
Duct thickness (cm)	0.1					
Pins per assembly	127					
Cladding thickness (cm)	0.038					
Fuel pellet diameter (cm)	0.754					
Helium gap thickness (cm)	0.01					
Helium void diameter (cm)	0.2435					
Assembly pitch (cm)	11.7917					
Pin pitch (cm)	1.0175					
Pin diameter (cm)			0.85			
		238	Pu 0.8 atom %			
	<sup>239</sup> Pu 63.18 atom %					
Relative abundance of plutonium nuclides	<sup>240</sup> Pu 32.71 atom %					
	<sup>241</sup> Pu 1.24 atom %					
		242]	Pu 2.07 atom %			

The simulated reactors were 400 cm long, with a flat-to-flat diameter of 401.67 cm. The thermal power (4740 MW) was chosen to keep the maximum inner cladding temperature of the softened mixed-oxide core below 823 K [19]. In the simulations, the cores were divided into ten axial regions of 20 cm in height, and 20 radial regions ranging from 36 to 66 assemblies each. The cores were radially and axially heterogeneous. The plutonium content of the fuel was radially and axially graded to flatten the power and flux profiles during the lifetime of the cores. The innermost 109 assemblies and the outermost 204 assemblies made up the reflectors, which were pins filled with either beryllium (in the case of the softened cores) or sodium (in the case of the unsoftened cores). A sodium diluent was incorporated into the regions situated  $\pm 100$  cm from the axial center of the cores, in order to further flatten the flux and reduce the dpa in regions that would have otherwise experienced the most damage. Figure 11 shows a schematic of the core to scale, with individual material regions numbered. The specific positions and compositions of the fuel, reflector, and diluent regions, corresponding to the numbered schematic in Figure 11, are listed in Table 3. Top-down diagrams of the core composition at every axial location are given in Supplementary Note 3, Figures S10–S19. In all four core designs,

the hexagonal assemblies contained 127 annular fuel, reflector, and diluent rods clad in HT-9 steel, with sodium coolant flowing between the pins. More details on the pin-level core geometry and the materials can be found in Supplementary Note 4.



**Figure 11.** Reactor core structure. Numbered cells belong to a single material region. Material content varied with axial and radial position. Material regions 1, 2, 17, 18, 19, and 20 contained beryllium assemblies (softened cores), or sodium-filled assemblies (unsoftened cores) in all axial locations. Material regions 3 to 16 contained fuel, with each numbered cell belonging to a single burnup region, or sodium diluent, which varied axially. Specific contents of each radial and axial material region are listed in Table 3.

Serpent 2.1.28 [20] was used for the simulations, and the results were corroborated using MCNPX 2.70 [21]. Additional details can be found in Supplementary Note 5. The spatial mesh used in the simulations had an axial grid of 20 segments with  $\Delta z = 20$  cm. The materials were burned at a thermal power of 4740 megawatts with a time step of 114 days, until a peak irradiation damage of 203 dpa was reached. The simulations tracked a total of 19.2 million neutron histories per burnup step. A thermal resistor model was used to determine the approximate fuel and coolant temperatures in all radial and axial regions, and these were fed back into the Monte Carlo simulations at the beginning, middle and

end of life, in an effort to compute the isothermal reactivity coefficients. More information on the heat transfer model can be found in Supplementary Note 6.

**Table 3.** Radial and axial plutonium concentration. Column headings refer to axial positions. The axial concentration from 0 cm to -200 cm is the reflection of the concentration from 0 cm to 200 cm, and the core was reflected about z = 0 cm. Values are plutonium concentrations as a proportion of total heavy metal in each region, in atomic percent.

Axial Position	0–20	20–40	40–60	60-80	80–100	100–120	120–140	140–160	160–180	180-200
	cm	cm	cm	cm	cm	cm	cm	cm	cm	cm
Material Region										
3	12.00	12.44	12.89	13.33	13.78	14.22	14.67	15.11	15.56	16.00
4	12.31	12.72	13.13	13.54	13.95	14.36	14.77	15.18	15.59	16.00
5	12.62	12.99	13.37	13.74	14.12	14.50	14.87	15.25	15.62	16.00
6	12.92	13.26	13.61	13.95	14.29	14.63	14.97	15.32	15.66	16.00
7	13.23	Na	Na	Na	Na	14.77	15.08	15.38	15.69	16.00
8	Na	13.81	14.09	14.36	14.63	14.91	15.18	15.45	15.73	16.00
9	13.85	14.09	Na	Na	Na	15.04	15.28	15.52	15.76	16.00
10	14.15	14.36	14.56	14.77	14.97	15.18	15.38	15.59	15.79	16.00
11	14.46	14.63	14.80	14.97	15.15	15.32	15.49	15.66	15.83	16.00
12	14.77	14.91	15.04	15.18	15.32	15.45	15.59	15.73	15.86	16.00
13	15.08	15.18	15.28	15.38	15.49	15.59	15.69	15.79	15.90	16.00
14	15.38	15.45	15.52	15.59	15.66	15.73	15.79	15.86	15.93	16.00
15	15.69	15.73	15.76	15.79	15.83	15.86	15.90	15.93	15.97	16.00
16	16.00	16.00	16.00	16.00	16.00	16.00	16.00	16.00	16.00	16.00

#### 4.1. Irradiation Damage

The displacements per atom of cladding and structural materials were computed using

$$dpa = \int \int \sigma_D(E_i)\phi(E_i, t)dE\,dt.$$
(1)

Here,  $\sigma_D$  is the energy-dependent microscopic displacement cross section (barns),  $\varphi$  is the neutron flux (1/b-s), and  $E_i$  is the incident neutron energy (eV). The integrals are over both energy and time.

The displacement cross section took into consideration the different types of neutron–material interactions that were sufficient to dislodge an atom. For steels, the elastic scattering on <sup>56</sup>Fe was the dominant mechanism through which atoms were displaced, but neutron absorption, and inelastic scattering, (n, p),  $(n, \alpha)$ , (n, n), (n, 2n), etc., all contributed. The total microscopic displacement cross section was then the sum of the displacement cross sections for the individual mechanisms:

$$\sigma_D = \sigma_{D,s(elastic)} + \sigma_{D,s(inelastic)} + \sigma_{D(n,\gamma)} + \sigma_{D(n,n)} + \sigma_{D(n,2n)} + \dots$$
(2)

The output of the Serpent simulations was used to compute a flux in 494 energy bins ranging from  $10^{-4}$  to 20 MeV for each location and time step. We used the NJOY [22] code to compute  $\sigma_D$  in the same 494-group structure as the flux. More details can be found in Supplementary Note 7.

#### 4.2. Fuel and Cladding Temperature

Temperatures for the fuel, coolant, and cladding were computed as a function of axial and radial position in the reactor using a one dimensional (1D) thermal resistor model [12,23,24], and thermo-physical quantities were drawn from References [25–27]:

$$q' = \frac{\Delta T}{R'},\tag{3}$$

where q' is the linear power density of the fuel (W/m), and R' and  $\Delta T$  are the linear thermal resistance (m-K/W) and the temperature difference (K), respectively, between the fuel and the coolant.

In order to reduce peak temperatures, the fuel was comprised of annular pins, which were clad in HT-9 steel with a helium bond. While sodium bonds are normally used in combination with metal fuel, we chose a helium bond for all fuel types in order to keep as many design parameters constant between cores. The 1D thermal transport model assumed no radial heat transfer between coolant channels. Additional details can be found in Supplementary Note 6.

## 4.3. Reactivity Coefficients

The core-average Doppler coefficient for the fuel was computed by uniformly perturbing the temperature of the fuel in the reactor by +300 K, and by recomputing the neutron multiplication factor. All other variables remained the same in order to isolate the effect of Doppler broadening. The coefficient was computed according to Reference [28]:

$$a_{Doppler} = \frac{1}{k^2} \frac{\Delta k}{\Delta T_{fuel}}.$$
(4)

The core-average reactivity coefficient for the sodium coolant was computed by uniformly perturbing the coolant temperature by +300 K, by adjusting the sodium densities accordingly, and by recomputing the neutron multiplication factor:

$$a_{coolant} = \frac{1}{k^2} \frac{\Delta k}{\Delta T_{coolant}}.$$
(5)

The core-average void coefficients were computed using Reference [28] by uniformly reducing the coolant density by 90%, and by recomputing the neutron multiplication factor. All other variables remained the same to isolate the effect of coolant voiding:

$$a_{void} = \frac{1}{k^2} \frac{\Delta k}{\Delta x_{coolant}},\tag{6}$$

where *x* is the core-average void fraction. Reactivity coefficients were computed at the beginning, middle (t = 4.4 years), and end of life. In all cases, this was done by sampling the materials from the burnup simulation at 0 years, 4.4 years, and 8.4 years, by performing the relevant temperature or density perturbation, and by recomputing the neutron multiplication factor using 432 million total neutron histories.

The uncertainties of  $k_{eff}$  values of Figure 9 were computed by repeating the Monte Carlo simulation five times, each with a different random number seed, and by calculating the mean and standard deviation of the results. The uncertainties in the reactivity coefficients were computed using the error propagation formula on the standard deviations for k, quoted by the Monte Carlo code. The errors were, in many cases, extremely small, as they reflected the statistical uncertainties in the Monte Carlo neutron transport. Additional details can be found in Supplementary Note 8.

## 5. Conclusions

While the impact of spectral softening on both metal- and oxide-fueled long-life fast-reactor cores is significant, this paper only provided a demonstration of unoptimized core designs. Although the mixed-oxide-fueled, beryllium-doped core with beryllium reflectors presented in this paper achieved good performance, the unexplored parameter space for long-life fast-reactor design using a softened neutron spectrum is large. Additionally, the fuel densities in the metal-fueled cores, as well as in the mixed-oxide-fueled core without beryllium, were reduced in order to enable an even comparison with the mixed-oxide core containing beryllium. In reality, the fuel densities of the three higher-damage designs would be greater, which would give a bonus to neutron economy. This suggested that the optimal design of maximizing burnup while keeping dpa low might be achieved using a metal-fueled core with additional beryllium. Finally, a key goal would be to find the fuel-loading strategy

which enables long-life cores to be operated using recycled actinides from their own discharged fuel. This would significantly reduce the net production of transuranics, which are the nuclides responsible for giving nuclear waste its long-lived signature.

**Supplementary Materials:** The following are available online at http://www.mdpi.com/1996-1073/11/6/1507/s1. Supplementary-Document.pdf.

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