

## SIGNIFICANT QUANTITY PRODUCTION RATES IN SMALL MODULAR REACTORS

**S. E. Bays**

Idaho National Laboratory

**G. A. Reyes**

Idaho National Laboratory

**M. J. Schanfein**

Idaho National Laboratory

**R. H. Stewart**

Idaho National Laboratory

**N. P. Martin**

Idaho National Laboratory

### ABSTRACT

This work assesses the time necessary to create a significant quantity of weapons-grade plutonium, given the reactor size over a broad range of small modular reactor (SMR) technology families. Given the avant-garde nature of current SMR development, it is impractical to consider every technological nuance. However, some meaningful contrasts can be made based on basic reactor physics characteristics. The rate at which plutonium is produced in a uranium-fueled reactor is primarily a function of the conversion ratio, power level, and plant availability factor. Currently, many SMR concepts considered as “advanced”, are being designed with at least one of the following traits: high conversion ratio, high power density, or multiyear to decades-long cycle lengths. These characteristics are generally thought to increase proliferation resistance, as they reduce the frequency at which a state can access in-situ bred plutonium. However, this premise does not apply to the breakout scenario. When the state is no longer bound to wait until the end of the declared cycle length, the time necessary to achieve one significant quantity of weapons-grade plutonium becomes highly relevant. In this study, four reactor technology families are explored: pressurized-water reactors, sodium-cooled fast reactors, high-temperature gas reactors, and molten-salt reactors. A 3D neutronics model for each reactor type was created based on available preconceptual design data found in the open literature. The plutonium production rate for each concept is contrasted as a function of the conversion ratio, power level, and plant availability factor.

### INTRODUCTION

This work assesses the time to create one significant quantity (SQ) of weapons-grade (WG) plutonium given the reactor size for a broad range of small modular reactor (SMR) technologies. Given the avant-garde nature of current SMR development, it is impractical to consider every technological nuance. However, some meaningful contrasts can be made based on basic reactor physics characteristics. The rate for which plutonium is produced in a uranium-fueled reactor is primarily a function of conversion ratio, power level, and the plant’s availability factor [1]. The conversion ratio (CR) is a function of the neutron spectrum and fissile-to-fertile isotopes ratio. For a given reactor size, the power level is a function of the fuel design’s thermal limits. The availability factor is a function of the reactivity- and burnup-limited cycle length and the time required to perform necessary off-power maintenance and refueling operations.

In this study, only advanced reactor concepts having cycle lengths in the multiple years were considered. Four reactor technology families are considered: pressurized-water reactor (PWR), sodium-cooled fast reactor (SFR), high-temperature gas reactor (HTGR), and molten-salt reactor (MSR), see Table 1. Advanced reactor concepts that fit the definition for SMR were selected (i.e., <300 MWe). With the exception of the HTGR where fuel shuffling has no meaning, the fuel cycle was considered to be one batch. That is, all concepts considered do not require a shutdown to shuffle or replace fuel assemblies. The plutonium production rate for each concept is contrasted as a function of CR, power level, and availability factor. A 3D neutronics model for each of these reactor types is created based on available preconceptual design data found in open literature.

It is important to note that not all advanced reactor concepts require high-assay low-enrichment uranium

(HALEU) (i.e.,  $5\% < ^{235}\text{U}/\text{U} < 20\%$ ). The NuScale SMR PWR is not designed for uranium enrichments  $> 5\%$ . Similarly, the MSR does not require HALEU, though the use of HALEU is not technologically excluded. Generally, the criticality requirement drives the enrichment level. As shown here, fast systems require a greater fissile density in order to be critical over the course of the cycle length. Thermal systems employ moderation to increase the probability of nuclear fission and thus require significantly less fissile inventory.

These scenarios are analyzed through the lens of reactor misuse to create plutonium and excludes the material diversion of HALEU (or low-enrichment uranium [LEU]). For perspective, we assessed the number of HALEU SQs comprising the fresh core. Because HALEU (or LEU) have enrichments of  $^{235}\text{U}/\text{U} < 20\%$ , one SQ is 75 kg  $^{235}\text{U}$ . Also, one SQ of plutonium is considered to be 8 kg Pu, see Table 2. If the number of HALEU SQs in the fresh fuel is greater than the number of Pu SQs in the irradiated fuel, the SMR concept can truly be considered “fully” proliferation resistant. This is because reactor misuse would be less attractive than simply diverting the original HALEU (or LEU) fuel.

**Table 1: List of concepts for which literature review indicate sufficient data to create neutronics model.**

Genre	<100 MWe
PWR	NuScale [2][3] (160 MWth / 50Mwe)
SFR	ARC-100 [4][5] (260 MWth / 100 Mwe)
HTGR	Xe-100 (X-Energy) [6] (200 MWt/ 75 Mwe)
MSR	IMSR [7][8] (423 MWt / 100 Mwe)

**Table 2. Time to acquire 1 SQ of WG material by diversion of the fresh HALEU core, or conversion of the irradiated fuel [9].**

	SQ (kg)	Time to divert fresh core	Time to convert irradiated fuel
WG Pu $^{233}\text{U}$	8	—	Irradiation time + 3 month
$^{235}\text{U}^\dagger$	75	~1 year	—

<sup>†</sup>note: This is the definition of one SQ of  $^{235}\text{U}$  when  $^{235}\text{U}/\text{U} < 20\text{ wt}\%$ .

The time to convert 1 SQ of HALEU (or LEU) to usable metallic components of a nuclear explosive device is assumed by the International Atomic Energy Agency (IAEA) to be one year, see Table 2. This implies that the time for fresh uranium fuel to be converted into a form suitable for enrichment, enriched to greater than 20%, and fabricated into metallic device components is assumed to be one year. Similarly, the time to convert the plutonium found in irradiated fuel into WG usable metallic components is three months [9]. This implies that the time to divert used fuel, reprocess the plutonium, and fabricate the plutonium into device components is assumed to take three months. Thus, if the time to breed 1 SQ Pu plus three months is less than one year, the critical path to weaponization is shorter using the reactor than it is for an undeclared uranium enrichment facility. In order to have usable metallic Pu components in one year, the 1 SQ Pu would need to be created in  $12-3 = 9$  months.

In the results section, the depletion results of the nuclear reactors considered in Table 1 are used to determine the point in the reactors’ fuel cycles for which the plutonium SQ is equivalent to the uranium SQ. For the purpose of the following discussion, this will be coined the significant quantity equivalence time (SQET). In all of these cases in Table 1, the SQET is greater than one year. However, working backward from the CR, one can solve for the minimum reactor power that would produce at least 1 SQ Pu in nine months. This is the calculation performed by Lamarsh to determine the minimum size of the graphite pile using measured physics parameters from the Brookhaven Graphite Research Reactor [1]. Performing this calculation, the SQET is re-evaluated. From the above discussion, if the minimum SQET

that produces one SQ Pu is less than nine months, misuse of the reactor technology to produce plutonium is a greater proliferation concern than diverting the fresh fuel. Consider a developing state that receives assistance from developed nuclear countries to install the SMR, then decided to construct a primitive copy with a minimalistic power level just to create 1 SQ. The SQET analysis can be used to consider if the SMR technology should be exported to that state, and what design improvement could be made to enable such export.

## **BACKGROUND**

The SMR concepts discussed here were selected purely as representatives of a broader family of technologies. They are not an endorsement or critique. Also, there was adequate public information for which to produce a simulation model. In all cases, except for the Integral Molten Salt Reactor (IMSR), even the  $^{235}\text{U}$  and  $^{239}\text{Pu}$  depletion results are published in the open literature. The IMSR salt composition, uranium enrichment, and reactor vessel inventory were “best guessed” based on other similar sources describing other MSRs.

### NUSCALE

The NuScale reactor is a self-contained nuclear steam supply system comprised of a reactor core, a pressurizer, and two steam generators integrated within the reactor pressure vessel [10]. The reactor model is loosely based on the papers by Hines et al. and Suk et al. [11][3].

The core is comprised of 37 fuel assemblies arranged in a rectangular lattice. Each fuel assembly is a  $17\times 17$  square-pitch array of fuel rods. Each fuel assembly has 24 guide tubes for control rod movements and one instrument tube that displaces a fuel rod within the  $17\times 17$  array. The NuScale fuel assembly is essentially a standard PWR fuel geometry with the exception that the fuel rod length is approximately half that of a typical PWR assembly. The fuel rod consists of a stack of enriched uranium oxide encapsulated in the zirconium alloy M5. The uranium enrichment is varied by assembly and rods within assemblies to aid in suppressing localized power peaking. The burnable absorber,  $\text{Gd}_2\text{O}_3$ , is also mixed with the  $\text{UO}_2$  in select locations to manage power peaking as well as aid excess reactivity suppression. The nominal cycle length is two full-power years. The thermal power is 160 MWth. The uranium enrichment in standard (no  $\text{Gd}_2\text{O}_3$  rods) ranges from 1.95% to 3.6%. The  $\text{Gd}_2\text{O}_3$  concentration ranges from 2.5% to 3.0% in select pins where the uranium enrichment is only 1.8% to 1.5%, respectively.

### XE-100

The Xe-100 is an advanced modular pebble-type HTGR. The uranium enrichment is 15.5% HALEU. The reactor model constructed for this work is loosely based on the paper by Mulder and Boyes [6].

The core is essentially a neutron reflected bin containing randomly placed spherical fuel pebbles, with each pebble being approximately the size of a 60-mm-wide billiard ball. Dispersed within the graphite of each fuel pebble are thousands of coated fuel particles, called tristructural isotropic (TRISO) particles. The core consists of approximately 223,000 pebbles. Each pebble contains approximately 19,000 TRISO particles. The Xe-100 fuel pebble and TRISO fuel system is based on the German Arbeitsgemeinschaft Versuchsreaktor (AVR) research test reactor [12]. A TRISO particle consists of a uranium oxy-carbide fuel kernel, which is then coated in a porous carbon layer, then an inner pyrocarbon layer, then a silicon carbide layer, and finally an outer pyrocarbon layer. The TRISO particle has a 0.855-mm diameter. The TRISO particles are dispersed in graphite with a packing fraction of approximately 10%. Said differently, the dispersion zone of the fuel pebble is 10% TRISO particles and 90% graphite by volume. The TRISO dispersion zone (a 50-mm diameter) is surrounded by more graphite to constitute the fuel pebble. The packing fraction of the core is approximately 62%. Each pebble passes through the core six times on average. The cumulative in-core residency time over the course of these six passes is approximately 1,273 full-power days (or 3.5 full-power years). The thermal power is 200 MWth.

### ARC-100

The Advanced Reactor Concept, ARC-100 is a an advanced modular SFR for which the pumps and heat exchangers all mounted (i.e., integral) within the vessel. The reactor model constructed for this work is loosely based on the paper by Wade and Walters [4].

The core is sealed for 20 years before refueling. The core is comprised of 92 fuel assemblies arranged in a hexagonal lattice. Each fuel assembly contains 127 fuel rods. The large diameter (1.3 cm) of the fuel rods as well as their tight spacing (pitch-to-diameter of 1.1) allows for significant internal fissile breeding. The high fissile conversion ratio enables the long cycle-length of 20 years. The ARC-100 metallic fuel system as well as reactor design are based on the USA Experimental Breeder Reactor (EBR-II) research test reactor [13]. Metallic slugs of U-10Zr metallic alloy are cast and placed inside cladding tubes of HT-9 ferritic/martensitic stainless steel. Because the core fuel is designed to last 20 years between refueling, power density was selected sufficiently low that the average discharge burnup and fast neutron fluence to structural materials is consistent with past irradiation experience in EBR-II and Fast Flux Test Facility (FFTF). The core thermal power is 260 MWth. The ARC-100 has three enrichment zones. The inner, middle, and outer uranium enrichments are 10.1%, 12.1%, and 17.2% HALEU, respectively. Since the core is designed for breeding, its designers explain that the HALEU core will breed plutonium sufficient to fuel other ARC-100 cores.

### IMSR

The IMSR is an advanced MSR for which the pumps, heat exchangers, and control rods are all mounted (i.e., integral) in a single primary vessel. This primary vessel is fabricated offsite and installed as a sealed unit [14]. The reactor model constructed for this work is loosely based on the paper by Carter and Borrelli [7] with some inputs (fuel enrichment and fuel burnup) taken from the IAEA Advanced Reactor Information System (ARIS) on-line database [15].

The cycle length is seven years before the sealed primary vessel is replaced. The core consists of 133 hexagonal graphite blocks containing flow channels to allow fuel salt to flow. The neutron moderation provided by these graphite blocks allows for the flowing fuel salt to be critical. When the fuel salt is outside the graphite blocks, it is subcritical. There are 19 flow channels per block. The IMSR has three different radial zones of graphite blocks, an inner, middle, and outer zone. Each zone has its own unique diameter of flow channels in its graphite blocks. The middle zone fuel channels are zoned to a nominal 85 v/o carbon moderator and 15 v/o molten-fuel salt. The inner zone fuel channels 10% more carbon and less fuel. The outer zone has 10% less carbon and more fuel. The fuel salt for the IMSR is not in the public domain. Thus, for the purpose of this analysis, a typical FLiBe composition of LiF-BeF<sub>2</sub>-UF<sub>4</sub> (71.75-16-12.25 mol%) is chosen [16]. The lithium is assumed to be <sup>7</sup>Li (i.e., enriched lithium). This composition is based on the LiF-BeF<sub>2</sub>-ThF<sub>4</sub>-UF<sub>4</sub> (71.75-16-12-0.25 mol%) molten-salt breeder reactor composition but assuming UF<sub>4</sub> takes the place of ThF<sub>4</sub>. The uranium enrichment is assumed to be 5% LEU. The thermal power is 423 MWth.

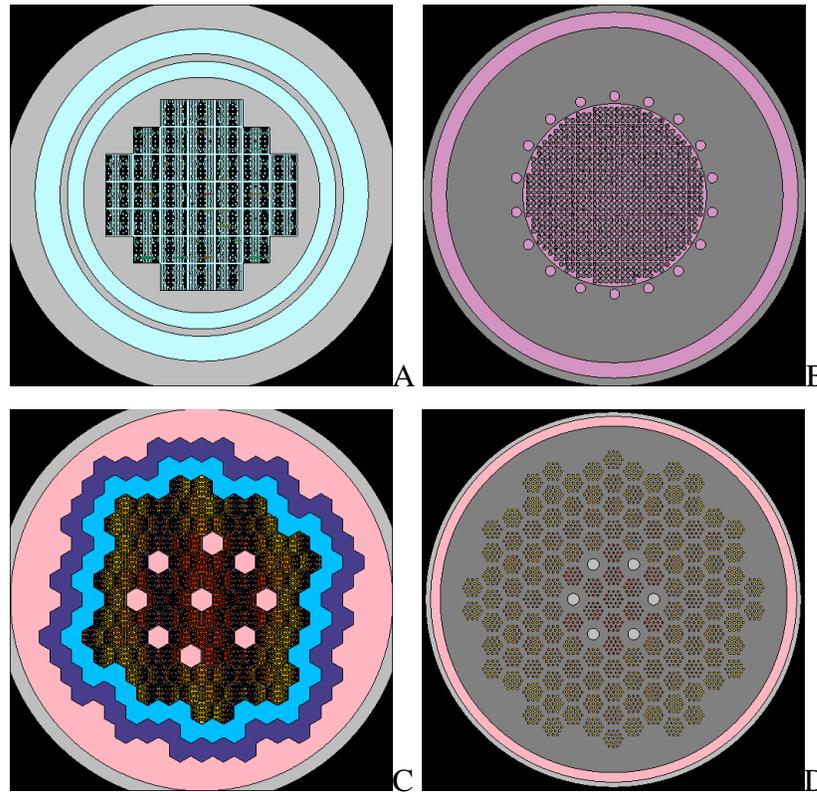
### **COMPUTATIONAL MODEL**

Serpent, Version 2.1.31, is produced by VTT Technical Research Centre of Finland. The code has a widespread user community and has emerged as one of the leading Monte Carlo codes for reactor physics applications over the last few years. Serpent uses the Monte Carlo method for solving particle (e.g., neutron and photon) transport in a continuous energy, angle, and 3D space representation of the reactor core [17]. Serpent uses an ACE-format library based on ENDF/B-VII.1 data, similar to the Monte Carlo N<sup>th</sup> Particle produced by Los Alamos National Laboratory. The code is optimized for solving reactor criticality and fuel depletion models at either the assembly or core level and is routinely used to create nuclear data “cross-sections” datasets from these simulations for use in modern deterministic codes.

A 3D full-core model of each reactor core in Table 1, is modeled in Serpent. A plan view through the core midplane for each core is shown in Figure 1. A high-level description of relevant modeling parameters is provided in Table 3. Control rods are modeled as fully withdrawn. Since no fuel assembly shuffling is involved in the NuScale<sup>1</sup> or ARC-100, these are simply depleted at full-power until the end-of-cycle.

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<sup>1</sup> The model by Hines et al. and Suk et al. [11][3] assumed no shuffling. However, in some NuScale fuel cycles, shuffling is performed [10].



**Figure 1. Plan view through core midplane of the Serpent models: (A) NuScale, (B) Xe-100, (C) ARC-100, and (D) IMSR.**

For the Xe-100, the Cartesian coordinates of the 19,000 TRISO particles are randomly sampled within the dispersion zone of a fuel pebble. The sampling is done such that the outer diameter of each particle does not overlap with neighboring particles. Given that the packing fraction is very small,  $\sim 10\%$ , a brute-force sampling method is adequate. However, solving the random sphere-pack problem for the high packing fraction of the fuel pebbles is significantly more difficult. There are algorithms to perform this problem solving, which were considered but ultimately deemed unnecessary for the purpose of capturing basic core reactivity, CR, and depleted uranium and plutonium nuclide densities. Instead, the fuel pebbles were modeled in a repeating body-centered-cubic (bcc) lattice type within the active core volume. The separation between pebbles was defined by the packing fraction as reported in the paper by Mulder and Boyes [6]. The location of fresh, once, twice, thrice, fourth, and fifth burned pebbles was randomly sampled in the core-wide bcc lattice. Fuel pebble burnup was achieved iteratively in five versions of the full-core model. First, the full-core is depleted for  $1/6^{\text{th}}$  of the average pebble life (i.e.,  $3.5/6 = 0.583$  years). The fuel nuclide densities for fresh designated pebbles are then mapped to those pebble locations designated as twice-burned pebbles. Then the  $2^{\text{nd}}$  iteration of the full-core model is depleted. This process is repeated until the pebbles designated as the sixth pass receive a burned fuel composition.

The IMSR model involves depletion and mixing. Formally, one would add the in-flow and out-flow of each fuel isotope (i.e., both actinides and fission products) to the solution of the Bateman equations, thus coupling the solution of the spatially dependent mass continuity equation with time-dependent isotope transmutation, buildup, and decay. Since this is a basic fuel-cycle study, this level of detail is not warranted. Mixing is handled by depleting the actinide compositions over very small 3-day time steps and then by performing the mixing calculations over the entire vessel's fuel-salt inventory similar to the study by Rykhlevskii et al. [16]. The current model has six depletable zones: inner, middle, outer in-core zones, as well as an outer downcomer annulus and a top and bottom plenum. These plenums account for the remainder of the reactor vessel fuel-salt inventory. After the three-day depletion, the six depleted compositions are averaged (i.e., volume-weighted average), and then remapped back to the six depletion

zones for the next three-day depletion. This process is repeated over the seven-year fuel cycle. The size of the plenums was adjusted such that the total salt inventory would reach 26 MWd/kg if burned at 423 MWth for seven years, per the fact sheet on the IAEA ARIS website [15].

**Table 3. Relevant modeling parameters.**

	NuScale	Xe-100	ARC-100	IMSR
Fuel Pellet/Slug/Kernel/Channel Diameter (cm)	0.81	0.0425	1.04	2.3/2.4/2.6 (inner/middle/outer)
Fuel Rod/Pebble/Channel Outer Diameter (cm)	0.95	6.0	1.30	
Rod/TRISO/Channel Pitch (cm)	1.26	n/a	1.43	5.0
Fuel Assembly/Pebble/Block Pitch (cm)	21.5 (Square)	7.2 (bcc)	17.0 (Hex)	26.2 (Hex)
Rods/TRISOs/Channels	264	19,000	127	19
Assemblies/Pebbles/Blocks	37	223,000	92	133
Moderator per Fuel Volume Ratio	1.7	140	n/a	5.7
H (or C) per <sup>235</sup> U Atom Ratio	148	3,480	n/a	4,366
Cladding/Coating	M5	SiC	HT9	n/a
Coolant	Water	Helium	Sodium	FLiBe/UF <sub>4</sub>
Moderator	Water	Graphite	n/a	Graphite
Reflector Material	Steel	Graphite	Steel	Graphite
Active Core Height (cm)	200	893	150	415
Active Core Diameter (cm)	148	240	179	317
Reflector Outer Diameter (cm)	186	440	247	390
Reactor Power (MWth)	160	200	260	423
In-Core Residency (Full-Power-Years)	2	3.5	20	7
Fuel Batches per Refueling Cycle	1	6 passes on average	1	1

## Results

### Nuclear Materials Accountability and Control

The fuel depletion analysis is presented in this section. Table 4 provides a comparison of fresh and used fuel uranium and plutonium compositions with comparisons to values reported in literature. To better appreciate the nuclear materials accountability and control (NMAC) of fuel storage for each of these fuel and reactor systems, the fresh and depleted special nuclear materials are normalized to a physical item representative of past ex-core storage technologies.

In the case of the NuScale and ARC-100 reactors, the item is the fuel assembly. The NuScale fuel assembly storage will be in water pools just as in traditional light-water reactors [18]. Fast reactor fuels similar to that of ARC-100 have been historically stored in water-filled cans in water pools after the core sodium is drained and washed from the assembly by steam. This is not the universal approach over all SFRs, but this is the process at the Joyo and Monju SFRs in Japan. These two reactors were monitored using the IAEA's containment and surveillance methods as part of the international safeguards program at those facilities. In the Japanese experience with the Joyo reactor, fresh fuel was stored in a room adjoining the reactor building [19][20]. Spent fuel assemblies were cleaned of sodium, and then canned in their own airtight water-filled canisters prior to being placed in the spent fuel pool for interim storage. Sodium cleaning is accomplished by steam and "desalted" water. In Japan's Monju SFR, fresh fuel was loaded into a fresh fuel storage rack before use. From here, it was transported via an "under-floor transporter," to a sodium pool adjacent to the reactor building, called the Ex-Vessel Storage Tank [20][21]. Spent fuel

assemblies were also stored in this pool prior to being cleaned of sodium and placed in a nearby water pool.

Pebble storage could take several forms, ranging from small canisters to large bins. In the German experience at the AVR and Thorium Hochtemperatur Reaktor, pebbles that had reached their full burnup potential, were discharged from reactor, and loaded into small canisters, called the AVR-K canister. The AVR-K canister could store 50 pebbles each. After sufficient cooling in the water pool, the pebbles were transferred from AVR-K canisters to the larger AVR-TLK [22]. The pouring of pebbles from the AVR-K into AVR-TLK canisters was conducted in a hot-cell. Two AVR-TLK canisters were loaded into a small cask, the CASTOR shipping/storage cask, and stored in an onsite vault-type room. The CASTOR cask, containing two AVR-TLK canisters, could store 1,900 pebbles. When these reactors were decommissioned, the CASTOR containers were loaded onto railcars and moved to a centralized storage facility where they now await final geologic disposal.

After the Molten-Salt Reactor Experiment (MSRE), the fuel salt containing most of the uranium and fission products was divided and drained into two separate tanks. The fuel salt was divided in order to ensure criticality safety. A third tank contained the flush salt. The volume of flush salt was relatively equal to that of the fuel salt, but only contained 1–2% of the uranium and fission products, thus allowing it to be stored in a single tank. After draining and filling these tanks, the salt was allowed to freeze and was surveilled until either the salt was reused or disposed. The contents of the MSRE fuel salt at room temperature was reported to be 66.4 ft<sup>3</sup> for the fuel salt and 69.9 ft<sup>3</sup> for the flush salt [23]. The density of frozen LiF-BeF<sub>2</sub>-ZrF<sub>4</sub>-UF<sub>4</sub> salt stored in these tanks was 2.5 g/cm<sup>3</sup>. Multiplying the fuel-salt volume by density and dividing by the two tanks, the mass of salt per tank is 2,350 kg. For the UF<sub>4</sub>/FLiBe composition chosen for the purpose of this study, the weight fraction of <sup>235</sup>U per fuel salt is 2.25 wt%. Thus, if this notional salt is stored in an MSRE-like drain tank, each tank would hold approximately 2,350 × 0.0225 = ~53 kg <sup>235</sup>U. As a reference point, the <sup>233</sup>U inventory of MSRE (both tanks) was 36.46 kg [23].

**Table 4. Fresh and used fuel compositions per NMAC item.**

	NuScale	Xe-100	ARC-100	IMSR
Item Type	Assembly Average	AVR-TLK (950 pebbles/container)	Assembly Average	One MSRE Fuel-Salt Drain Tank
Items/Core	37	~235	92	~40
<b>Fresh Fuel</b>				
<sup>235</sup> U (gm)	6.97E+03	1.11E+03	3.03E+04	5.31E+04
<sup>238</sup> U (gm)	2.47E+05	5.90E+03	1.95E+05	1.01E+06
<b>Used Fuel</b>				
<sup>235</sup> U (gm)	4.13E+03	2.14E+02	1.40E+04	2.69E+04
<sup>238</sup> U (gm)	2.45E+05	5.48E+03	1.75E+05	9.97E+05
<sup>239</sup> Pu (gm)	8.84E+02	5.48E+01	1.06E+04	4.98E+03
<sup>240</sup> Pu (gm)	2.17E+02	3.24E+01	7.52E+02	1.79E+03

Normalizing used fuel mass per discharged assembly is natural for assemblages of fuel rods. Normalizing per storage canister and drain tank provides a means to gain a perspective of these fuel systems that will require elements of bulk and item NMAC practices. The <sup>235</sup>U mass of an LEU NuScale fuel assembly is of the same order magnitude as that of an AVR-TLK container that currently stores AVR and Thorium Hochtemperatur Reaktor pebbles in interim dry storage. The ARC-100 fuel assembly is the smallest item but has the greatest fissile loading. This is intuitive from the perspective of critical mass. In the fast spectrum, the nuclear cross-section is smaller than for thermalized (moderated) neutrons. Thus, fissile material needs to be configured in a more concentrated arrangement for criticality. The MSRE drain tank has a fissile loading of the same order of magnitude even though it is an LEU fuel.

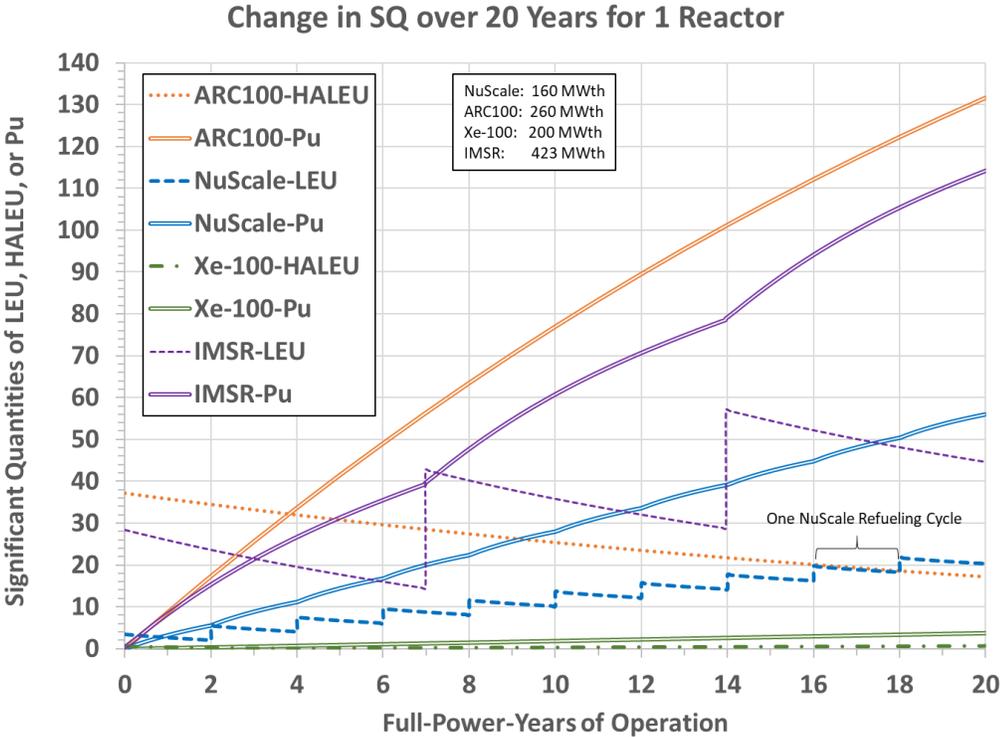
#### Significant Quantity Production

In this section, the fuel depletion data is converted to SQs of LEU/HALEU or Pu per Ref. [9]. Figure 2 shows these depletion results converted into SQs. The fuel depletion data is tabulated to represent the

evolving inventory, both in core and used fuel storage, of one reactor over 20 years. For NuScale and the IMSR, the whole cores are fueled, irradiated, and discharged (i.e., no-shuffling). Thus, the fresh fuel of Cycle N+1 is added to the discharged used fuel inventory of Cycle N. Since, ARC-100 has a full-core depletion of 20 years, no special cycle-to-cycle tabulation is needed.

For Xe-100, the HALEU inventory at time zero is approximated as the fresh fuel for 1/6<sup>th</sup> of the core. In practice, the first core for a pebble-bed HTGR is a combination of fresh fuel pebbles and inert graphite pebbles. The graphite pebbles are used to dilute the reactivity contribution of the fresh pebbles. As the fuel pebbles are depleted and ultimately replaced by fresh pebbles, the graphite pebbles are removed and sent to their own storage bin, until the core contains fuel pebbles with a distribution of burnups representing the six reactor passes. The SQs of HALEU and Pu for 3.5 full-power years represents the pebbles after an average core residency of 3.5 years. Significant quantities to the right are the accumulation of discharged pebbles. In practice, the core residency is stochastic, thus the accumulation of SQs will have a statistical uncertainty not represented in Figure 2.

To gain a general understanding of the special nuclear materials inventories that may evolve within a developing state, the SQ of Figure 2 should be normalized on an equal energy level. Consider a fleet of SMRs with thermal generation capacities of 10 GWth. Assuming a thermodynamic efficiency of 34%, this would equate to an electric production of 3.4 GWe. Producing electricity for one year, this fleet would generate ~30 TWh of electricity annually. This is the order of magnitude of electric generation for many developing world countries [24]. Figure 3 shows the LEU/HALEU and Pu SQ evolution normalized to 10 GWth. From this plot, it is apparent that at beginning-of-life (i.e., fresh fuel) the LEU/HALEU SQs for the ARC-100 and IMSR are roughly an order of magnitude greater than that of the NuScale and Xe-100 reactors. It is interesting that the ARC-100 uses HALEU, but the IMSR uses LEU. Thus, the relatively large fissile inventory of these two advanced reactors are more likely a result of provisioning the core with sufficient <sup>235</sup>U to remain critical for many years without refueling. The fact that the ARC-100 is HALEU is an attribute of the fast neutron spectrum. Conversely, the IMSR is a thermal spectrum, which enjoys greater <sup>235</sup>U fission cross-sections.



**Figure 2. LEU/HALEU and Pu SQ trends for the four different reactor types.**

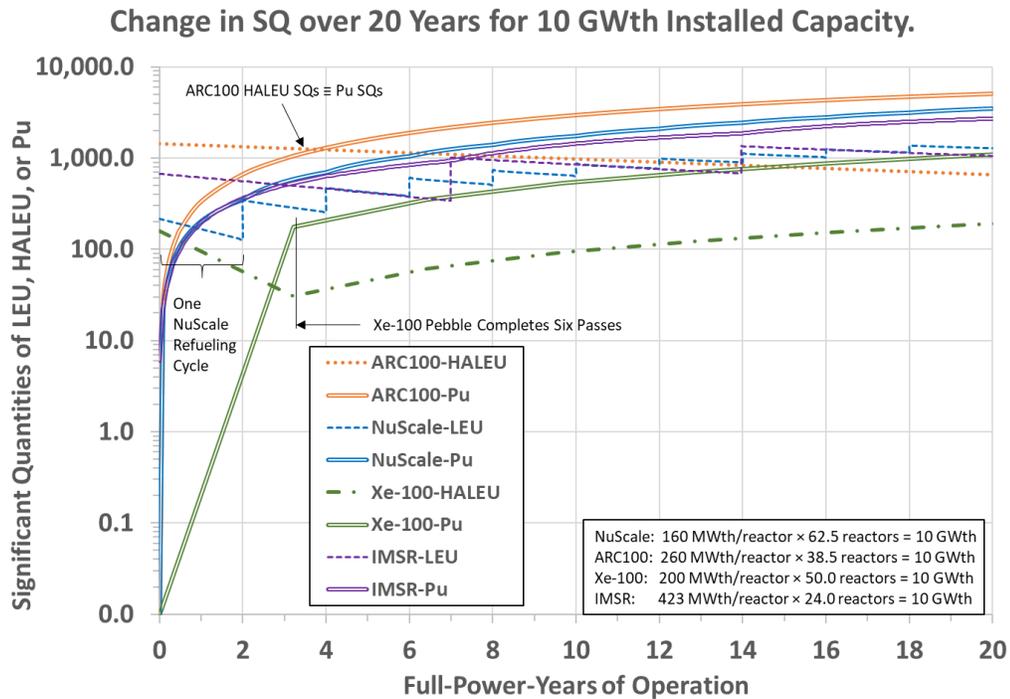
The time for which the LEU/HALEU SQ inventory equals the Pu SQ inventory always occurs in the first

cycle (or core residency for Xe-100). Controlling influences on the magnitude of SQs and the occurrence of SQET are the reactor’s criticality requirement to produce the prerequisite number of fissions between refueling (typically referred to as cycle energy) and CR. These values are summarized in Table 5.

**Table 5. Comparison of reactor power, cycle energy, CR, and SQET.**

	Reactor Power (MWth)	Cycle Energy (GWd)	Conversion Ratio (CR)	SQET (years)	SQs at SQET
NuScale	160	117	0.50	0.82	2.8
Xe-100 (1/6 <sup>th</sup> core)	200 ÷ 6	43	0.40	1.67	0.31
ARC-100	260	1899	0.75	3.8	32.2
IMSR	423	1082	0.45	3.0	21.4

The plutonium production rates are a consequence of CR. The CR is defined as the production rate of new fissile material (chiefly plutonium) versus fissile consumption (mostly <sup>235</sup>U, but some <sup>239</sup>Pu and <sup>241</sup>Pu is burned in situ). In order to create sufficient fissile plutonium to remain critical for 20 years, the ARC-100 is designed as a fissile breeder. It also has the largest fresh HALEU inventory. Thus, it is not surprising that it creates the largest plutonium inventory after 20 years. The NuScale has the next greatest CR. Its fuel cycle is also additive in the sense that fresh LEU is added to the inventory every two years. Similarly, the bred plutonium is cumulative. The IMSR and Xe-100 have CRs <0.5. The IMSR-bred plutonium is cumulative with every seven-year refueling. The Xe-100 is continuously refueled with pebbles; thus, the plutonium accumulation is also continuous.



**Figure 3. LEU/HALEU and Pu SQ trends for a fleet of reactors producing 10 GWth.**

Minimum Production Reactors

The production of plutonium per fission energy can be computed by following the arithmetic of J. Lamarsh found in a 1977 report to Congress by the Office of Technology Assessment [1]. Lamarsh used measured physics parameters from the Brookhaven Graphite Research Reactor (BGR) to estimate the <sup>239</sup>Pu production rate by both thermal and resonance neutron absorption in <sup>238</sup>U. The ratio of this absorption rate to the <sup>235</sup>U consumption rate is the CR. The BGR was a 30 MWth uranium-fueled, air-

cooled, graphite-moderated, pile-type research reactor. Used for civilian research purposes, the BGRR had basic physics parameters similar to the Hanford piles. Lamarsh computed the BGRR CR to be 0.806.

The amount of recoverable energy produced from one fission of  $^{235}\text{U}$  is 200 MeV. From this, the  $^{235}\text{U}$  burnup rate by fission is 1.05 gm/MWd. However, one must remember that not all  $^{235}\text{U}$  atoms are consumed by fission. Some are removed by radioactive capture with a ratio of approximately, 0.175 captures per fission. Thus, the total  $^{235}\text{U}$  consumption rate is  $1.05 \times 1.175 = 1.23$  gm/MWd. The  $^{235}\text{U}$  consumption rate will vary somewhat from reactor to reactor because the amount of recoverable energy per fission and the capture-to-fission ratio is partly a function of neutron energy and reactor configuration. However, the assumption of 1.23 gm/MWd is sufficiently accurate for the following comparison. Using the BGRR CR, the plutonium production rate is  $0.806 \times 1.23 \times 239/235 = 1.00$  gm/MWd. Therefore, the reactor power to produce 8 kg in one year is  $8,000 \text{ gm} / (1.00 \text{ gm/MWd} \times 365.25 \text{ days}) = 21.9 \text{ MWth}$ .

The Serpent code provides an estimated CR (see Table 5) based on the sum of *all* reactions, producing any fissile isotope divided by all neutron reactions (fission and capture) that remove any fissile isotopes, thus accounting for in-situ burning. This is more sophisticated than Lamarsh's calculation, which only considers the neutron absorptions in  $^{238}\text{U}$  for  $^{239}\text{Pu}$  production and absorptions, resulting in a  $^{235}\text{U}$  consumption.

Lamarsh's CR can be computed from the depletion rates deduced from the Serpent depletion results. The  $^{239}\text{Pu}$  production and  $^{235}\text{U}$  consumption rates per fission energy (gm/MWd) are tabulated from the Serpent depletion results as the net change in isotopes over one year, then divided by reactor power times one year, see Table 6. Lamarsh's CR is found by converting these rates to a molar basis then taking the ratio,  $CR \sim ({}^{239}\text{R}/239)/({}^{235}\text{R}/235)$ . Contrasting Table 5 and Table 6, there are significant differences in the CR stemming from differences in assumptions. Though the Serpent estimate of CR is more useful from a holistic perspective of core fissile management, the primary interest here is the production of  $^{239}\text{Pu}$  from the consumption of  $^{235}\text{U}$ . Thus, the depletion results over one year from the Serpent depletion calculation will be used to determine the minimum reactor size and associated SQET.

**Table 6. Plutonium production rates estimated using the Serpent computed CR as well as hand-calculated from the Serpent depletion result.**

	$CR \sim \frac{{}^{239}\text{R}/239}{{}^{235}\text{R}/235}$	From Serpent Depletion Result (gm/MWd)	
		${}^{239}\text{Pu}$ Production ( ${}^{239}\text{R}$ )	${}^{235}\text{U}$ Consumption ( ${}^{235}\text{R}$ )
NuScale	0.37	0.38	1.01
Xe-100	0.12	0.15	1.32
ARC-100	0.67	0.73	1.08
IMSR	0.32	0.39	1.20

The minimum reactor size to produce one SQ in one year, following nine months of irradiation for the plutonium route or one year following the enrichment route, is given in Table 7. For such a hypothetical reactor, it is assumed that the reactor can be made critical for nine months with the CR found for its SMR reference and fueled with 1 SQ of LEU/HALEU. This is a rather large assumption and should be considered for future work.

The SQET occurs when the consumed LEU/HALEU SQ, represented by  $y_1 = 1 - ({}^{235}\text{R}/75,000) \times t$ , is equal to the produced Pu SQ, represented by  $y_2 = ({}^{239}\text{R}/8,000) \times t$ . Thus, the SQET in units of MWd is  $t = 1 / ({}^{239}\text{R}/8,000 + {}^{235}\text{R}/75,000)$ . The SQET in units of full-power days is this value divided by the reactor power needed to produce the 1 SQ Pu in nine months. The SQET in units of days requires dividing by availability factor.

The availability factor (AF) is really a function of refueling (and maintenance) outage per time at-power. This is difficult to quantify for future reactors, but AFs for past demonstration reactors of PWR, HTGR, SFR, and MSR, can be found in historic literature. It is assumed that a proliferating state would encounter refueling and maintenance issues similar to that of historic demonstration reactors, not the planned SMRs.

An AF of 97% for demonstration PWRs are represented by the Shippingport PWR reactor [25]. This number is provided after discounting the time to change-out experiments. An AF of ~72% for HTGR is represented by AVR [26]. This value is compiled from a chart covering years of operation from 1967–1987, including a period from 1978–1980 where the plant experienced significant steam generator damage. An AF of >80% for demonstration SFRs is represented by the early years of EBR-II from 1976–1978, excluding down-time for experiment change out [27]. AF of 87% for demonstration MSRs is represented by MSRE [28]. Since in this section we are considering primitive copies of the SMRs considered in Table 1, we will use these AFs over that put forward by the designers of the SMRs.

The results shown in Table 7 highlight that the greater the CR, the smaller the reactor can be to produce one SQ Pu in a given amount of time. Conversely, lower CRs tend to drive an earlier equivalency between LEU/HALEU SQs and Pu SQs. The earlier the SQET occurs, the earlier the state is committed to the success of the clandestine plutonium production mission for weaponization.

**Table 7. Minimum reactor power, SQET, and SQs for the smallest conceivable plutonium production reactor having conversion ratios of known advanced reactor SMRs.**

Reactor Family	Availability Factor	Minimum reactor size (MWth)		SQET (Full-Power Days)	SQET (Days)	SQ at SQET
		9 months	1 year			
PWR	97% [25]	78	58	213	219	0.78
HTGR	72% [26]	189	142	143	199	0.52
SFR	80% [27]	40	30	237	296	0.86
MSR	87% [28]	74	56	207	238	0.75

## CONCLUSIONS

Keeping in mind that the results for minimum reactor size are general, and not representative of vendors’ specific technologies, some high-level observations can be made. SFRs can be designed for maximizing the CR, thus they enable the smallest reactor power to produce one SQ Pu in the shortest amount of time. However, they generally require significant core inventories of HALEU to be critical. The SQET is also very long. Thus, the SFR fuel’s HALEU SQ value towards weaponization is significantly longer than for other SMR types. Thus, the option to misuse the HALEU fuel for plutonium production as opposed to diverting it towards an enrichment route have similar value to a would-be proliferator.

HTGRs also generally assume HALEU as the fissile source. However, because of their small CR, they also have the greatest reactor power and shortest SQET. Thus, for a state to misuse the HTGR technology family for Pu weaponization, significant investment into enrichment (or procurement of enrichment services) in addition to reactor development (i.e., a reactor rated greater than 100 MWth) must be made.

For PWRs, only LEU is needed in addition to modest power levels (i.e., a reactor rated less than 100 MWth). Similarly, for MSRs, only LEU is needed, and only modest power levels are required. These two cases tend to be a middle-ground compared to the SFR and HTGR cases.

The effect of plant availability should not be ignored. Pressurized-water technology was considered mature even during the early days of nuclear development. Advanced reactor technology (e.g., pumps, valves, material science, instrumentation and controls) may be considered mature for new SMR builds by established vendors. However, redeveloping this technology by a nascent nuclear state can be expected to experience less plant availability similar to past demonstration plants. Lack of plant availability can essentially level the playing field in terms of which reactor technology family can proliferate one SQ the fastest.

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