Molten Salt Reactors

Resource Requirements and Proliferation-Risk Attributes of Single-Fluid and Two-Fluid Designs

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ABSTRACT. Molten salt reactors (MSRs) are often advocated as a radical but worthwhile alternative to traditional reactor concepts based on solid fuels. This article builds upon the existing research into MSRs, most of it carried out at Oak Ridge National Laboratory until government-sponsored research was canceled in the 1970s, to model and simulate the operation of various types of MSRs using the MCNP5 Monte Carlo neutron transport code. Types of molten salt reactors considered include thorium-fueled single-fluid and two-fluid breeder reactors with a focus on those designs using denatured fuels. The paper analyzes specifically the resource utilization and basic proliferation-risk attributes compared to those of standard light-water reactors.[1]

Background

Molten salt reactors (MSRs), in which fissile and fertile nuclear materials are dissolved in a liquid carrier salt, were originally proposed in the late 1940s. Oak Ridge National Laboratory under Alvin Weinberg later spearheaded the research and development effort and first conceived MSRs as a compact, relatively light-weight reactor design to power airplanes.[2] Following the termination of the aircraft-propulsion project in 1961, Oak Ridge continued its research into molten salt reactors for electricity production, which led to the Molten Salt Reactor Experiment (MSRE, 8 MWt, 1965–1969) and the concept for a prototype Molten Salt Breeder Reactor (MSBR, 1000 MWe, never built), which would have been based on the thorium fuel cycle. Research on the technology was terminated in 1976 due to a lack of government funding. No other molten salt reactors were ever built in the United States or elsewhere after shutdown of the MSRE forty-four years ago, and no MSR ever used thorium fuel.

Several technical challenges would have to be resolved before molten salt reactors could be brought to a commercial level. There are currently few incentives for the nuclear industry, however, to embrace new types of reactors and fuels that are radically different from those designs currently licensed and used. Despite these obstacles, several research groups, private companies, and independent developers continue to explore MSRs given that they potentially offer important advantages vis-à-vis existing reactors. These include, for example, the iMSR, the Fuji MSR, the LFTR, and the Waste-Annihilating MSR.[3] These newer designs are often envisioned as small modular reactors with power levels of 200 MWe or less; these power levels are also considered in the analysis of notional designs below.

Design Principles and Options for Molten Salt Reactors

Molten salt reactor are typically (but not necessarily)[4] designed as thermal systems facilitated by the low-Z constituents of common salts. The core itself contains additional moderating material, typically graphite, which ensures that the salt reaches criticality only within the core. Since the fuel is in liquid form, there is great flexibility with regard to fuel choice and reactor configuration. Many types of fuel can operate within MSRs, but most designs envision a thorium fuel cycle, in which natural thorium-232 is used to breed fissile uranium-233, because of the excellent neutron economy of thermal uranium-233 systems.[5] Initially, designers sought to optimize the breeding ratio of MSRs in order to "compete" with sodium-cooled fast neutron reactors. As discussed in more detail below, additional design options become available, if a breeding ratio of less than one is accepted.

All MSR designs involve varying levels of online fuel processing. At a minimum, volatile gaseous fission products escape from the fuel salt during routine reactor operation and must be captured. Most designs also call for the removal of rare earth metals from the core since these metals act as neutron poisons. Some designs envision more complex processing schemes, including the temporary removal of protactinium from the salt or other adjustments of the actinide inventory in the fuel.

The great design flexibility of MSRs, combined with the remaining technology gaps, has contributed to a reluctance of designers to publish detailed specifications of core geometries for the proposed designs.[6] More so than perhaps for any other type of reactor, there is uncertainty in the broader energy debate about the potential of molten salt reactors—and, more generally, about the potential of thorium as a reactor fuel. The following discussion highlights the main design features and options for MSRs.

Single-Fluid and Two-Fluid Designs

There are two fundamental classes of molten salt reactors: single-fluid and two-fluid designs. In a single-fluid design, a single type of molten salt flows through the core. This salt contains both the fissile material and any fertile material for breeding. In contrast, in a two-fluid design, the fertile material is separated into a second molten salt. The two salts need to be placed in close proximity to achieve adequate breeding ratios. Fissile material that is produced in the blanket salt is quasi-continuously extracted and transferred to the fuel salt, while new fertile material is added to the blanket. Typically, the two fluids are separated by a graphite structure, which serves as a barrier and a neutron moderator. Single-fluid and two-fluid designs both have tradeoffs in terms of complexity, performance, and proliferation risk.

The main advantages of two-fluid designs are higher breeding ratio and simplified fuel processing. Most importantly, since breeding occurs outside the fuel salt, protactinium-233 produced from neutron capture in thorium-232 does not have the same poisoning effect as it does in a single-fluid design. Therefore, two-fluid designs do not require the removal of protactinium from the blanket salt; it can simply be left in the salt to decay into uranium-233. In contrast, in the original single-fluid reactor aiming for the highest possible breeding ratio, the protactinium is immediately removed after it is produced following neutron capture in thorium and left to decay to uranium-233 outside the core ($T_{1/2} \approx 27$ days) before it is reintroduced into the salt. Essentially, two-fluid designs can decrease the fuel processing requirements of MSRs over single-fluid designs, at the expense of design complexity and cost.

In either case, a basic thorium-fueled MSR can run on virtually pure uranium-233 for its fuel, except for a startup period during which enriched uranium may be used. Uranium-233 is a weapon-usable material with a small critical mass and low neutron background. Under routine operating conditions, no stockpiling of uranium-233 is envisioned, but in principle, uranium-233 could also be extracted and set aside for weapons purposes while another make-up fuel is used to keep the reactor running. The presence of high-purity uranium-233 in a MSR fuel cycle therefore presents a credible proliferation concern and has led to the concept of denatured designs.

Denatured Designs

One method of preventing the buildup of pure uranium-233 in thorium-fueled reactors is to add depleted uranium to the fuel. Uranium is considered denatured if the following condition is met at all times:

$$N_{238} > 7 \times N_{233} + 4 \times N_{235} \quad \text{or} \quad (7 \times N_{233} + 4 \times N_{235})/N_{238} < 1 \tag{1}$$

Accordingly, for binary mixtures with uranium-238, the uranium-233 and uranium-235 content has to remain below 12.5% and 20% of total uranium, respectively. Such a modified MSR design is referred to as a denatured molten salt reactor (DMSR). Introducing uranium-238 to the fuel salt complicates matters as the isotope absorbs neutrons, which decreases the reactivity of the core and also produces relevant amounts of plutonium.

MSR Reference Models and Results

The analysis below focuses on denatured designs, which seek to avoid the presence of directly weapon-usable materials in the reactor core and fuel cycle. Results are based on neutronics calculations carried out with the MCODE computer code system, which links the Monte Carlo neutron transport code MCNP5 with the ORIGEN2 point-depletion code and permits reactor burnup calculations with regularly updated neutron flux distributions and one-group cross sections.[7] All numbers are scaled to a total power output 500 MW thermal, which is consistent with an electric output of 200 MWe, i.e., a power level proposed for many small modular reactors. A particular challenge for MSR neutronics calculations is the possibility of adjusting the fuel composition of liquid-fuel reactors quasi-continuously, for example, to add fissile material or to remove fission products. Existing burnup codes, however, have been developed primarily for solid-fuel reactors, where fuel depletion between refuelings is significant.

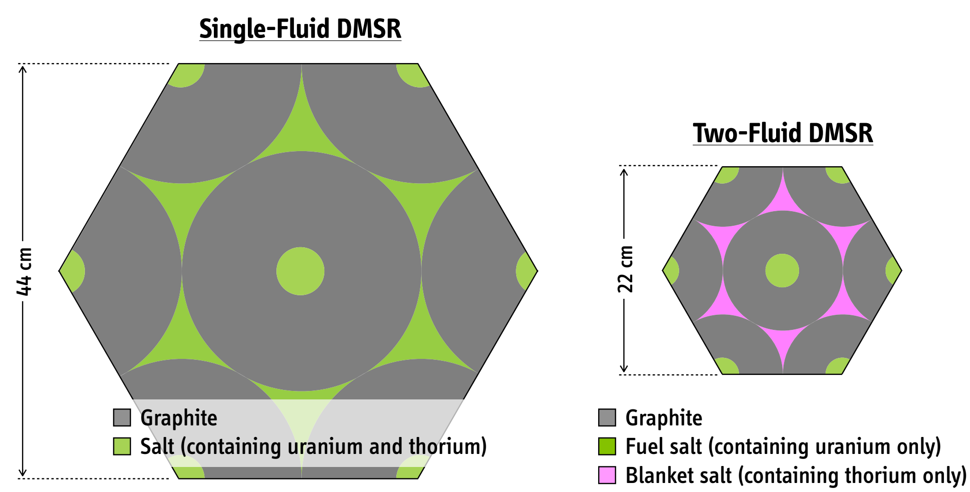

Figure 1: Unit cells of the denatured single-fluid design and the interlaced two-fluid design.

Notional Single-Fluid Denatured Molten Salt Reactor (SF-DMSR)

Oak Ridge's denatured molten salt reactor with once-through fueling represents the laboratory's attempt to design a reactor with maximum proliferation resistance in mind.[8] In the early 1980s, a breeding ratio greater than one was no longer considered necessary for a viable reactor design. The notional SF-DMSR examined here is inspired by this design and uses a hexagonal unit cell that consists of graphite logs with molten salt flowing through and around them (Figure 1, left).[9] The reference fuel salt contains 10 mol% ThF_4 and 3.0 mol% UF_4 enriched to 20% uranium-235 at startup. The reactor is operated at an average power density in the fuel salt is 37.5 kW per liter. To achieve the target power output of 200 MWe, the SF-DMSR would have an active core diameter and height of about 7.75 meters. At startup, the reactor core contains about 16,000 kg of thorium and 5,000 kg of uranium enriched to 20% U-235.

The main results of the burnup calculations are shown in Figure 2. Over time, uranium-233 builds up in the core and makes a significant contribution to the energy release, but periodic additions of 20%-enriched uranium are necessary to sustain criticality; as discussed below, these additions also keep the fuel denatured at all times despite the buildup of uranium-233 in the salt. The increasing uranium-238 content in the salt also leads to a significant buildup of plutonium, which reaches about 430 kg after 30 years. MCODE calculates an average breeding ratio of about 0.76 for the reactor, which is comparable to the breeding ratio of 0.80 listed in the original design document. Table 1 below summarizes additional resource and fuel-cycle requirements for this SF-DMSR.

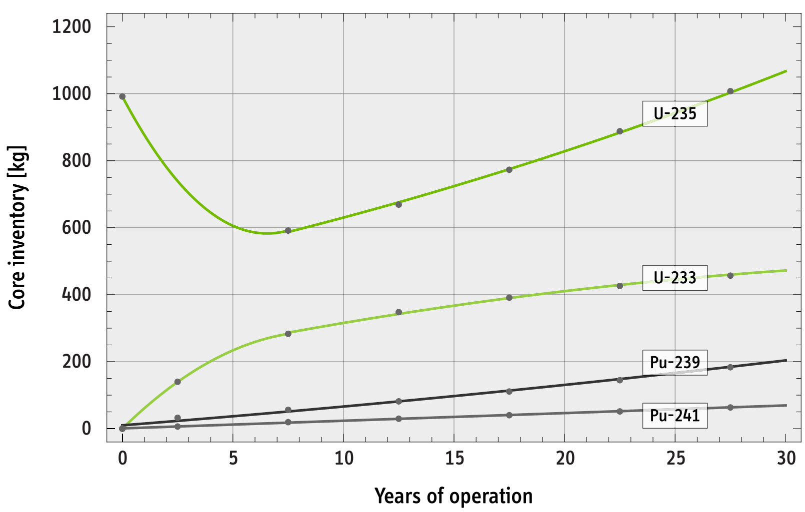

Figure 2: Fissile content of the Single-Fluid Denatured Molten Salt Reactor.

MCODE (MCNP5, ORIGEN2) calculations

Notional Two-Fluid Denatured Molten Salt Reactor (TF-DMSR)

Although two-fluid designs present greater engineering challenges than single-fluid designs, they can achieve higher conversion ratios, which would further reduce resource requirements of MSRs. The analysis below seeks to quantify the performance of a notional two-fluid design compared to the single-fluid variant. To meet basic proliferation resistance requirements, the fuel salt will remain denatured throughout the lifetime of the reactor; as discussed further below, there is no viable strategy though to also denature the blanket salt.

The denatured two-fluid molten salt reactor (TF-DMSR) proposed here is derived from the original single-fluid design. The blanket salt is "interlaced" with the fuel salt, meaning that the two salts flow past each other in the core in order to maximize blanket breeding rates. For this analysis, the unit cell of the SF-DMSR was modified for two-fluid use so that the fuel salt flows through the central bores of the graphite logs and the blanket salt in between the logs (Figure 1, right). To reproduce neutronics comparable to the single-fluid design, the size of the unit cell has been reduced by a factor of two so that the distance between to fuel channels remains about the same. *It has to be pointed out that this particular interlaced design may be impractical from a reactor-engineering perspective and should be understood only as a conceptual approach to determine the neutronics performance of a TF-DMSR.*

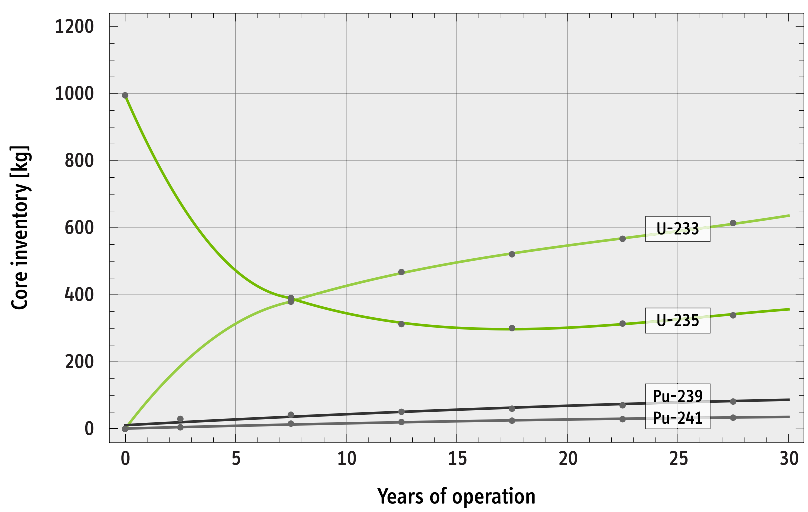

Figure 3: Fissile content of the Two-Fluid Denatured Molten Salt Reactor.

MCODE (MCNP5, ORIGEN2) calculations

In a two-fluid reactor, the uranium bred in the blanket salt would be constantly removed in an effort to keep its concentration as close to zero as possible and prevent it from fissioning in the blanket.[10] This cannot be easily modeled with existing burnup codes. We therefore pursue a two-step approach: In a first MCODE reference run, the equilibrium protactinium concentration in the blanket salt is determined for the proposed TF-DMSR, which then also determines the effective uranium-233 production rate in the blanket. This production rate is used for all subsequent calculations. In the main run of the simulation, the blanket composition is kept constant, i.e., the blanket salt is no longer considered in the burnup calculations except for its influence on the neutron economy, in particular due to the presence of protactinium. The main run of the simulation can now focus on the evolution of the fuel-salt composition, which changes due to burnup, pre-calculated periodic additions of uranium-233 recovered from the blanket, and additions of low-enriched make-up fuel.

In the proposed design, the blanket and fuel salts initially contain 15 mol% ThF_4 and 4.5 mol% UF_4, respectively. The reactor is operated at a power density of 74 kW per liter in the fuel salt and, just as the single-fluid design, has an initial in-core uranium inventory of 5,000 kg. A 500 MW thermal denatured two-fluid reactor of the proposed design reaches an in-core equilibrium inventory of protactinium-233 of about 13 kg, which corresponds to a uranium-233 production rate of 350 grams per day. Results for the TF-DMSR are summarized in Figure 3 and Table 1. Most importantly, the two-fluid design breeds uranium-233 more effectively, which reduces the need for low-enriched make-up fuel and therefore also reduces plutonium buildup in the fuel.

Resource Requirements and Proliferation-Risk Attributes

The results from the neutronics calculations for these notional DMSRs can be used to determine basic resource and fuel requirements and to compare them to reference values for other types of reactors. Here, we use a notional 200-MWe integral light-water reactor (iPWR) for this purpose. These reactors are typically designed to operate for about three years without refueling and can reach a burnup on the order of 30 MWd/kg for an initial fuel enrichment of five percent.[11] These characteristics translate into an inventory of 18,000 kg of 5%-enriched fuel for one 200-MWe iPWR.

Table 1 summarizes the average resource and fuel-cycle requirements of the three reactors considered for this analysis. Combining the requirements for the initial core with the annually required services and materials, one can estimate and compare the lifetime requirements of the iPWR, the SF-DMSR, and the TF-DMSR. Based on our simulations, the notional denatured single-fluid MSR is more than twice as resource-efficient as the iPWR reducing the lifetime demand for natural uranium from about 2,000 tons to 940 tons. Similarly, requirements for enrichment services are reduced by 40% from about 1.3 million SWU to 760,000 SWU over the life of the plant. As anticipated, the two-fluid design is even more fuel efficient than the single-fluid design: on average, the annual additions of make-up fuel can be reduced to 200 kg of 20%-enriched uranium compared to 500 kg required for the single-fluid design. Over the lifetime of the plant, the TF-DMSR needs only 520 tons of natural uranium and 420,000 SWU; it is

	iPWR	SF-DMSR	TF-DMSR
Initial core			
Natural uranium	200,000 kg	235,500 kg	235,500 kg
Natural thorium	—	16,000 kg	16,000 kg
Uranium enrichment	5%	20%	20%
Separative work	128,000 SWU	190,000 SWU	190,000 SWU
Enriched uranium demand	18,000 kg	5,000 kg	5,000 kg
Externally supplied services and materials, per year (average)			
Natural uranium	67,000 kg	23,500 kg	9,400 kg
Natural thorium	—	(small)	(small)
Uranium enrichment	5%	20%	20%
Separative work	42,500 SWU	19,000 SWU	7,600 SWU
Enriched make-up fuel	6,000 kg	500 kg	200 kg
Lifetime requirements: Initial core and thirty years of operation			
Natural uranium	2000 tons (reference)	940 tons (47% reduction)	520 tons (26% reduction)
Separative work	1,280,000 SWU (reference)	760,000 SWU (60% reduction)	420,000 SWU (33% reduction)

Table 1: Average resource and fuel-cycle requirements for one 200 MWe reactor.

about twice as efficient as the single-fluid design and 3–4 times more efficient than the reference iPWR.

One reason to explain the superior performance of the two-fluid design compared to the single-fluid design is the “level of denaturalization” of the fuel salt (Figure 4). Both reactors start just below the limit of 1.0 (as defined in Equation 1), but the value drops under 0.6 for the SF-DMSR due to relatively large amounts of uranium-238 added to the fuel as part of the low-enriched make-up fuel. In contrast, the TF-DMSR maintains a value over 0.8 at all times, which significantly improves the neutronics of the system.

Denatured thorium-fueled reactors eliminate the concern of having significant amounts of highly enriched uranium present in the core of the reactor. This is an important advantage vis-à-vis other proposed MSR concepts. That is not to say, however, that DMSRs present no proliferation risks. Uranium-233 is still being bred in the reactor, meaning that protactinium is present in the core and theoretically could be separated from the molten salt to decay into almost pure uranium-233. Chemical separation of the protactinium from the molten salt would be a feasible task because the plant already possesses a hydro-fluorinator to remove fission products and dissolve actinide-oxides in the molten salt. This reactor component could be modified to become a fluorinator, which could then be used to remove uranium from the fuel salt.

Figure 4: Level of denaturalization of the uranium in the fuel salt.

Moreover, single-fluid and two-fluid reactors pose characteristic design-specific proliferation challenges. In the case of a denatured single-fluid reactor, there may be an incentive to relax the fueling policy to improve the neutronics of the system. In particular, one could imagine a proposal for a denatured design using highly enriched make-up fuel (HEU) in order to avoid an "unnecessary" drop in the level of denaturalization over the life of the plant unless optimized designs can offset this effect. The situation in the case of the two-fluid design is even more complex: in the proposed design, pure uranium-233 is produced in the blanket salt, which is then immediately extracted and transferred to the denatured fuel salt; in principle, no significant quantities of uranium-233 are stockpiled, but inspecting such a system, i.e., making sure that no protracted diversion is occurring, could be a significant safeguards challenge with inherent uncertainties. Given that the production rate of uranium-233 in the blanket of a 200-MWe two-fluid design is on the order of 350 grams per day, it would take 3–4 weeks to produce one significant quantity of the material; slightly increased amounts of low-enriched make-up fuel could be used to compensate for the diverted material in a period during which a diversion is taking place. We note that "preemptively" denaturing the blanket salt does not appear a viable nonproliferation strategy and would also lead to additional neutronics challenges. Similarly, an external tank of depleted uranium (sometimes called a "poison pill") that would be used to instantaneously and automatically denature the fuel of a molten salt reactor in the case of an anticipated or unfolding breakout does not effectively address potential proliferation concerns as it must be assumed that such a mechanism could be disabled by the host.

Conclusion

Molten salt reactors operated on a thorium fuel cycle continue to attract the attention of several research groups, private companies, and independent developers as a radical alternative to technologies deployed today. In particular, as the analysis in this paper confirms, MSRs could offer significant advantages with regard to resource efficiency compared to conventional thermal reactors based on light-water reactor technology. Depending on specific design choices, even fully denatured reactors could reduce uranium and enrichment requirements by a factor of 2–4. While significant, these improvements may or may not be sufficient to gather support for the near-term demonstration of a modern prototype reactor as many technical challenges have to be resolved and licensing would prove difficult.

Specific design choices for molten salt reactors have important implications for associated proliferation risks. Some aspects of these risks can be quantified by examining fissile inventories, but other nonproliferation aspects have to be assessed based on the nature of the reactor fuel, the core design, and the proposed chemistry. It is critical to keep these design options in mind from the outset when narrowing technology choices for MSRs. This paper highlights some of the nonproliferation challenges that ought to be addressed and resolved for MSR designs. Overall, fully denatured single-fluid reactors using low-enriched make-up fuel appear as the most promising candidate technology minimizing overall proliferation risks and should therefore receive particular attention even if their neutronics performance is inferior to some competing designs.

Endnotes

[1]This paper is partially based on Edward B. McClamrock, *Molten Salt Nuclear Reactors: A Comparative Assessment of the Resource Requirements and Proliferation-Risk Attributes of Single-Fluid and Dual-Fluid Denatured Designs,* Senior Thesis, Department of Mechanical and Aerospace Engineering, Princeton University, May 2013.

[2]The Aircraft Reactor Experiment (ARE, 2 MWt) was conducted at Oak Ridge from 1953–1954 and evolved to the 60 MWt Aircraft Reactor Test. The program was cancelled in 1961, just before the second reactor was completed. See: *Review of the Manned Aircraft Nuclear Propulsion Program, Atomic Energy Commission and Department of Defense,* Report to the Congress of the United States, Comptroller General of the United States, U.S. General Accounting Office, Washington, DC, February 1963.

[3]For more information on these particular concepts, see: www.terrestrialenergyinc.com, www.flibe-energy.com, and www.transatomicpower.com. In addition, there are several websites dedicated to thorium as an alternative reactor fuel that directly or indirectly advocate specific types of molten salt reactors.

[4]Since 2005, the Generation IV International Forum has prioritized fast-spectrum MSRs concepts (MSFRs), which envision graphite-free cores, see www.gen-4.org for details.

[5]On average, uranium-233 emits 2.3 neutrons after absorption of a thermal neutron, which is higher than the respective values for uranium-235 (2.0) and plutonium-239 (2.2).

[6]An exception is the excellent review of MSR concepts by David LeBlanc, "Molten salt reactors: A New Beginning For an Old Idea," *Nuclear Engineering and Design,* 240 (6), June 2010, pp. 1644–1656. For a remarkable and extensive online libraries of historic documents and reports, see www.moltensalt.org/references/static/downloads/pdf.

[7]Zhiwen Xu et al., "An Improved MCNP-ORIGEN Depletion Program (MCODE) and its Verification for High Burnup Applications," *PHYSOR,* Seoul, Korea, 2002; A. G. Croff, *A User's Manual for the ORIGEN2 Computer Code,* Oak Ridge National Laboratory, July 1980; S. Ludwig, *Revision to ORIGEN2 — Version 2.2,* Oak Ridge National Laboratory, 2002; *MCNP5-1.40 RSICC Release Notes,* Los Alamos National Laboratory, November 2005.

[8]J. R. Engel et al., *Conceptual Design Characteristics of a Denatured Molten-Salt Reactor with Once-Through Fueling*, ORNL/TM-7207, Oak Ridge National Laboratory, July 1980.

[9]The design of the SF-DMSR discussed here is slightly simplified compared to the original design proposed by Oak Ridge; these changes should only marginally affect the neutronics performance.

[10]Fission products are undesirable in the blanket salt since they absorb neutrons and are difficult to separate in the presence of thorium. An increasing rate of fission events in the reactor blanket also affects the power distribution in the core.

[11]A. Glaser, L. Berzak Hopkins, and M. V. Ramana, "Resource Requirements and Proliferation Risks Associated with Small Modular Reactors," *Nuclear Technology*, 184, October 2013.