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Assessing the PRISM reactor as a disposition option for the British plutonium stockpile

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ABSTRACT

The United Kingdom considered using the PRISM sodium-cooled fast reactor as a disposition option for its civilian plutonium from reprocessed MAGNOX and Advanced Gas-cooled Reactor spent fuel. This article assesses the plutonium disposition capabilities of the PRISM reactor for the U.K. stockpile. The article first describes how the stockpile was created. It then provides a simulation of reactor burn-up, the resultant isotopic compositions of PRISM spent fuel are simulated and the dose rates of that fuel. Dose rates greater than 1 Sv/h at 1 meter from the fuel were assumed to establish "proliferation resistance" and would constitute a radiation barrier to proliferators. Results suggest that the U.K. stockpile could be irradiated to that proliferation resistance target in 31.3 years, using two 840 MWth PRISM cores operating at a 30 MWd/kgHM burnup rate. By the time all the U.K. plutonium has been irradiated, however a fraction of the PRISM spent fuel will have decayed below the proliferation resistance target. Thus, even though in 2019 PRISM was removed from consideration by the U.K. government because it is not expected to be available for that use for another 20 years, this paper concludes that should PRISM become available earlier it would still be a poor choice for plutonium disposition.

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Introduction

In 2011, GE Hitachi Nuclear Energy (GE Hitachi) proposed that its sodium-cooled reactor called "Power Reactor Innovative Small Module" (PRISM) could be used for plutonium disposition in the United Kingdom.[1] The reactor, in various versions and designs, has been under development since the 1970s and builds upon experience with the Experimental Breeder Reactor II (EBR-II). PRISM was listed in the U.K. as one of the Nuclear Decommissioning Authority's (NDA) credible options for plutonium disposition since 2014.[2] In March 2019 however, the U.K. government

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removed PRISM from the list of viable options, because it is not expected to be fully developed in twenty years. The government will nonetheless continue to monitor its technological progress in the future.[3] In this paper, the plutonium disposition capabilities of the PRISM reactor are analyzed in detail. This analysis provides valuable insights of the capability of PRISM to dispose separated civilian plutonium as well as for other use cases. For example, a study discussed its use for disposition of excess weapon-grade plutonium,[4] and more recently it has been proposed as a fast neutron source for a new test facility run by the U.S. Department of Energy (U.S. DOE).[5]

The U.K. owns the largest civilian plutonium stockpile in the world. At the end of 2016, the U.K. declared it owned a total of 110.3 tons[6] of separated civilian plutonium, and held an additional 23.2 tons of plutonium owned by other countries.[7] While additions to this stockpile will end when the B205 plant reprocessing plant receives its final shipments in 2020,[8] the future of this stockpile remains unclear. Storing separated plutonium is difficult because of security concerns, its hazardous nature and high cost. Most of the material is stored in oxide form which is of limited deterrence for diversion: converting the material to a metallic form for weapons purposes is a task of relatively low complexity.[9] It is also possible to a construct a weapon using plutonium oxide directly.[10] The U.K. civilian stockpiles include sufficient material to build more than 16,000 nuclear warheads.[11] In addition to the security concerns, issues with safety at the Sellafield nuclear site[12] and with the deteriorating conditions of some of the plutonium storage containers have been reported.[13] The U.K. Parliamentary Office of Science and Technology estimates storage of this plutonium costs £73 million annually.[14]

GE Hitachi has claimed that the PRISM reactor could render the full stockpile "proliferation resistant" within only 20 years, "faster than any alternatives."[15] GE Hitachi further claims that the PRISM reactor could dispose the whole plutonium stockpile within its 60-year lifetime, utilizing one reactor block with two cores.

This paper provides detailed estimates of the history and the future of the U.K. plutonium stockpile, including the isotopic composition of the stockpile over time. It then describes a simulation model of the PRISM reactor. Finally, a detailed analysis of the claims of the vendor is carried out. Burn-up calculations using the simulated PRISM reactor model provide estimates of the composition and dose rates of the fuel after irradiation. The dose rate of the spent fuel is used as the key metric of its resistance to proliferation. The vendor claims regarding the achievement of proliferation resistance and the simulation results are compared.

This analysis does not analyze claims made by the vendor that the reactor could irradiate all of the U.K. plutonium. The plutonium is stored

as plutonium oxide powder, but has different isotopic compositions and impurities depending on the source. Mixed oxide (MOX) fuel is the most common form of plutonium fuel, containing plutonium oxide mixed with oxides of natural or depleted uranium. A fraction of the U.K. separated plutonium could not be used as MOX fuel in light water reactors because it requires further purification, although PRISM would be more tolerant of impurities. However, it is out of the scope of this paper to analyze fuel fabrication options in detail. Furthermore, the vendor has proposed that the reactor could "meet all of the U.K. energy needs for the next 100 years" if all plutonium stored at Sellafield would be fissioned.[16]

In general, there are two approaches to plutonium disposition beyond continued interim storage: final storage, probably in a chemically altered form, or irradiation in nuclear reactors and storage afterwards. One of the first U.K. studies on this topic was published in 1998 by the Royal Society, when stockpiles were less than half of today's values.[17] The study noted the urgency of deciding on a policy, starting a debate on variants of the two options which continued for the coming decades.[18] In most contributions to the debate, irradiation seemed to be the preference, particularly using the plutonium as MOX fuel in thermal light water reactors. In 2011, the Department of Energy and Climate Change identified this as the U.K.'s "preliminary policy." Implementing this policy has two major obstacles: The U.K. has only one LWR operating in the U.K., and its operator EDF made clear that it is not interested in using MOX fuel.[19] Additionally, the Sellafield MOX fuel fabrication plant shut down in 2011 due to poor operation history.[20]

While the official preference for using MOX fuel continued,[21] other reactor-based options came into consideration in 2011. Two reactor vendors offered new reactor types: PRISM[22] and the CANDU EC6 reactor. Both were mentioned as "credible options" in a 2014 policy paper by the Nuclear Decommissioning Authority (NDA).[23] But as of March 2019, only the suppliers of a CANDU reactor receive continued support for certain research activities.[24] According to the 2014 policy paper, only plutonium that cannot be used in a reactor for technical reasons can go directly to final storage. For final disposition, plutonium is mixed with radioactive nuclear waste and immobilized in ceramics or vitrified in glass. The policy paper also discussed use in "low-spec" MOX which was supposed to be produced by a MOX fuel fabrication plant when capacity was not needed.

Regardless of the option chosen, the plutonium sent to storage should be in a form that is unattractive or inaccessible to diversion by the owning state or theft by other actors. When talking about a "proliferation resistance" of plutonium, the reference usually is the spent fuel standard–making the recovery of disposed plutonium as complicated as the recovery

of unseparated plutonium in spent fuel from civil nuclear power reactors.[25] A key barrier to reprocessing spent fuel is the high radiation field of the radioactive fission products in the fuel. This field forces a proliferator to use a remotely operated process and heavily shield all operations. In the original CISAC report on *"Management and Disposition of Excess Weapon Plutonium"*, a reference value of 1 Sv/h in a distance of 1 m is defined as sufficiently high to be a deterrent.[26] Three hours of exposure at that level would be lethal to 50% of its recipients.[27] This value is used both by the U.S. Nuclear Regulatory Commission (NRC) and the International Atomic Energy Agency (IAEA) as the radiation level above which the material can be considered to be self-protecting.[28] Therefore, only a lower level of safeguards is required.

Cesium-137, with a half-life of about 30 years, is the main isotopic contributor to the dose rate for decades after removal from the core. Consequently, with intermediate cooling periods of up to 100 years before final storage, the radiation from the spent fuel elements decreases considerably. Spent fuel elements might well fall below a set limit during their intermediate cooling period.

In this paper, the radiation barrier is the determining factor to assess a material's proliferation resistance. While other barriers can be relevant, high dose rates emitted by the spent fuel present the greatest barriers to reprocessing.[29] This article uses a threshold value of 1 Sv/h at 1 meter after a 30 year cooling period to determine "proliferation resistance."

History and future of the U.K. Plutonium stockpile

Civilian reprocessing in the United Kingdom has existed for more than fifty years. Although the U.K. publishes its civilian plutonium stockpiles in reports according to IAEA Information Circular 549, beginning with data for the end of the year 1996, this information is not sufficient for the purpose of this article. Declarations only include absolute holdings, not the changes that occur each year. This makes it harder to track down annual changes to the stockpile due to reprocessing, ownership changes or due transfers of material to the respective country of origin. In addition, the declarations do not identify which reactor was used for plutonium production; the specific burn-up values of spent fuel prior to reprocessing; and the time of reprocessing. Such information is important to determine the isotopic composition of plutonium in the spent fuel. Over time, plutonium-241 decays into americium-241,[30] hence changing the composition after reprocessing, too.

With a possible civilian breeder program in mind, the U.K. started to develop a reprocessing program in the 1960s. At the time, uranium

resources were considered scarce relative to the anticipated global demand. Plutonium would be needed as startup fuel in fast reactors operating in a closed nuclear fuel cycle, where they would "breed" more plutonium. In the 70's forecasted uranium resource estimates increased dramatically, reprocessing plants proved uneconomic, proliferation concerns grew, and breeder programs in most countries were abandoned.

The United Kingdom's first large reprocessing plant, B205, opened in 1964. The plant was specifically designed to reprocess spent fuel from MAGNOX reactors, named after the magnesium oxide cladding for the metallic fuel. The first such reactor was Calder Hall which began operation in 1956. Its early purpose was nuclear weapons plutonium production. A total of 26 reactor blocks at 11 different sites were constructed for electricity production during the 1960s. It has been the U.K. government policy to reprocess all MAGNOX fuel. All the reactors are shut down today, the longest remaining active were those at the Wylfa site. MAGNOX spent fuel cannot be stored in pools, as the alloy corrodes quickly in water.

Consequently, the fuel continued to be reprocessed even though there was no continuing need for more plutonium. Dry storage could have been an alternative option but was not used.[31] According to recent estimates by the Nuclear Decommissioning Authority, the B205 plant will receive the last fuel discharges from reactors in 2019 and end operation in 2020, when all the fuel has been reprocessed.[32]

U.K.'s second generation of reactors, "Advanced Gas-cooled reactors" (AGR), came online starting in 1976, and several of these reactors are still in operation. The reactors use oxide fuel, which cannot be reprocessed at the B205 plant. To reprocess AGR fuel as well as open a prospective market of reprocessing foreign oxide fuel the government built its second reprocessing facility, THORP (Thermal Oxide Reprocessing Plant). When THORP started operation in 1994, the operators had secured contracts to reprocess approximately 10,000 tons of spent fuel, more than half of that from foreign sources.[33] THORP never met the operator's expectations and promises and only reprocessed a portion of the AGR fuel. In 2005, the plant suffered a massive leak leading to a nearly 3 year-long closure. THORP operation ended in November 2018.[34] With B205 closing in 2020, there should be no additions of material to the U.K. stockpile.

Figure 1 shows an estimate for the development of the U.K. civilian plutonium stockpiles in past and future. The U.K. plutonium stockpile from 2020 onwards will include 85.8 t of plutonium from MAGNOX spent fuel reprocessing, and 23.6 t of plutonium from AGR reprocessing. For this estimate, data from multiple sources have been combined to allow for separate listing of plutonium originating from MAGNOX and AGR reactors as well as foreign sources. All assumptions used for this estimate are described in

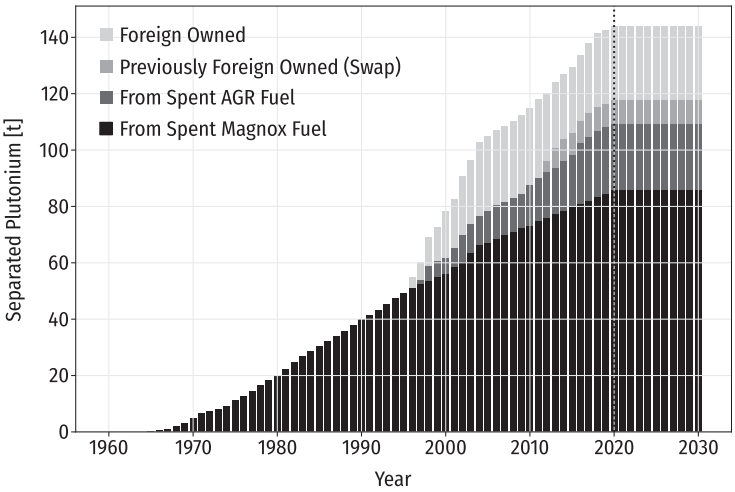

Figure 1. U.K. stockpile of civilian separated plutonium in the past and future. Different origins for parts of the stockpile are shaded differently. This figure assumes an average MAGNOX burn-up of 5 MWd/kg HM from 1996 onwards. Based on the assumptions described in the text, if the burn-up would be lower, there would be a higher fraction of plutonium from reprocessing spent AGR fuel.

Table 1. Estimated isotopic composition of the U.K. plutonium stockpile in the year 2030.

	Isotopic Composition (wt%)						
Year	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	Mass (t)
MAGNOX	0.02	71.85	22.51	1.04	0.93	3.35	85.8
AGR	0.50	53.67	30.73	3.48	5.00	6.27	23.6

the Appendix.[35] Table 1 shows the isotopic composition of the stockpile resulting from the two domestic reactor types, including the Am-241 content for the year 2030. These isotopic compositions are used for further simulations in this work.

Simulating Plutonium irradiation in the PRISM reactor

Reactor design

The PRISM reactor design as analyzed in this paper is based on several decades of combined public and private R&D efforts on liquid metal cooled reactors. After closure of the Clinch River Breeder Demonstration reactor in 1983, the U.S. DOE started a larger funding initiative focusing on the development of advanced liquid metal reactors (ALMR). As part of this funding initiative, Argonne National Laboratory (ANL) developed the integral fast reactor (IFR), a reactor concept that also included additional fuel cycle components. Technology for the IFR was in part based on experience gained with the EBR-II, an earlier breeder design also developed at ANL.[36] In parallel, General Electric (GE, now GE Hitachi) received funding from

DOE under the AMLR program to continue the development of a concept conceived in 1981, PRISM (Power Reactor Inherently Safe Module).[37] The concept for PRISM was not only a reactor core, but a full plant, able to produce fresh fuel and process spent fuel. PRISM also benefited from experience gained with EBR-II, as well as the IFR program.[38] By 1994 PRISM was progressing towards commercialization: a review by the U.S. Nuclear Regulatory Commission (NRC) stated that "no obvious impediments to licensing the PRISM design had been identified."[39] However, in the same year, DOE support for the reactor program was canceled, and most research on IFR and PRISM was stopped.[40]

In the following years, technology development continued supported by General Electric alone.[41] Government funding returned with the US Global Nuclear Energy Partnership (GNEP) in 2006.[42] One of GNEP's key elements was to design an Advanced Burner Reactor that could reuse or recycle spent nuclear fuel, for which the government sought commercial partners and announced funding for conceptual designs.[43] As part of GNEP, GE Hitachi proposed a revised PRISM model, again with a close integration of fuel production and reactors. Together with a Nuclear Fuel Recycling Center (NFRC) six of these reactor cores would form an Advanced Recycling Center. The meaning of the acronym PRISM was changed to "Power Reactor Innovative Small Module," the name currently in use.[44]

PRISM's design varies, depending on purpose. Designs include core configurations with low conversion ratios for used nuclear fuel (UNF) recycling or transuranic burning, cores that have a conversion ratio of one and cores with even higher conversion ratios allowing the reactor to operate as a plutonium breeder. These higher conversion ratios are achieved through the addition of special breeding elements, and increased plutonium content in the driving region.[45]

Since this paper's focus is plutonium disposition, the PRISM core is modeled after the descriptions for the UNF recycle configuration. The reactor model is assumed to operate at a power level of 840 MWth. The active region of the reactor has a height of 66 cm and is surrounded by reflecting and shielding elements. The core configuration is depicted in Figure 2, additional geometric details are summarized in Table 2. To achieve a more level neutron flux profile, the core has two fuel zones with differing plutonium contents. The inner fuel zone (IFZ) contains 84 fuel elements, the outer fuel zone (OFZ) 108 fuel elements. Some fuel element positions are used for reactor control and safety mechanisms, control rods, emergency shutdown rods ("ultimate shutdown rods") and gas expansion modules. The latter expel coolant from the core in case of lower coolant pressure due to coolant pump failure using a pressurized gas. This

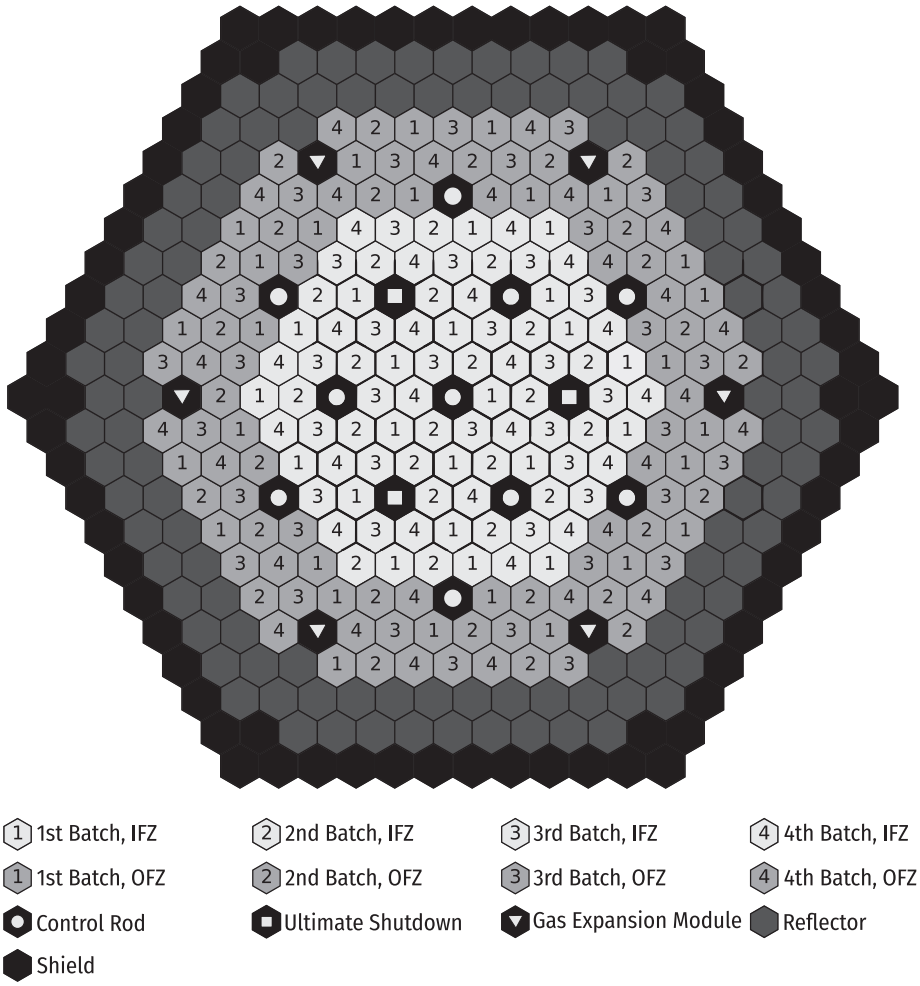

Figure 2. Core layout of the PRISM reactor model. The two different fuel zones are depicted in dark gray (OFZ) and light gray (IFZ). The different batches are assigned using different numbers. All safety elements are filled with sodium for the simulation.

Table 2. Geometric details of modeled reactor design.

Parameter	Value
Number of fuel elements[a]	84/108
Fuel element lattice pitch	16.142 cm
Duct wall thickness	0.394 cm
Duct gap	0.432 cm
Number of fuel rods per element	271
Fuel rod radius	0.27385 cm
Cladding thickness	0.0559 cm
Fuel pin pitch	0.90687 cm
Active height[a]	66 cm

Notes: Entries marked with [a] from Brian S. Triplett, Eric P. Loewen, and Brett J. Dooies, "PRISM: A Competitive Small Modular Sodium-Cooled Reactor," *Nuclear Technology* 178, (2012): 186–200, all other data from Tyler Sumner, "Effect of Fuel Type on the Safety Characteristics of a Sodium Cooled Fast Reactor" (PhD dissertation, Woodruff School of Mechanical Engineering, 2010).

mechanism reduces overall core reactivity. No control rods and safety mechanisms were included in the model. Instead, their positions were filled with sodium.[46]

The hexagonal fuel elements have a lattice pitch of 16.142 cm and contain 271 fuel rods each, which are separately modeled. The fuel is an alloy of uranium, plutonium and zirconium with a density of 15.05 g/cm^3, the zirconium content is 10% of the weight.[47] The uranium is of natural enrichment. Fuel rod cladding and fuel element structures are made from HT-9 steel.[48] To allow for fission product gas release, fuel rods contain a large gas plenum, assumed to be filled with helium gas.[49]

For plutonium disposition, the reactor would be fueled with plutonium that has been separated in the past. It is assumed that no further isotopic separation will take place before fuel manufacturing, hence plutonium decay products in particular americium-241, will be part of the new fuel. The fuel isotopic composition is based on the average composition of respective fuels in the year 2030 in the U.K. plutonium stockpile, as calculated in the previous section. Calculations of the isotopic compositions of plutonium originating from MAGNOX spent fuel will be separate from those of AGR spent fuel. Although the stockpile will contain plutonium separated over a larger timespan, the average isotopic vector will be used in each of the two sets of calculations, assuming start of reactor operations in the year 2030 (cf. Table 1).

The UNF core model described in the literature specifies that the fraction of the fissile plutonium isotopes (plutonium-239 and plutonium-241) is 11.3% in the IFZ and 13.5% in the OFZ, with a total transuranic content of 18.9% and 22%, respectively.[50] The model described here uses fuel with a different isotopic composition. The total transuranic content for the model used here has been slightly adjusted to achieve an initial core multiplication factor of 1.08.[51] For fuel based on plutonium from MAGNOX reprocessing, this yields a total transuranic content of 18.3% in the IFZ and 21.9% in the OFZ. For fuel based on AGR plutonium, the content is 21.8% and 26.0%, respectively. The reactor model described here was validated intensively in other works.[52]

Simulating reactor fuel depletion

To simulate the change of the fuel composition over time, depletion calculations were performed using VESTA 2.1 which couples MCNPX with its own depletion module PHENIX.[53] For this step, MCNPX in version 2.7 has been used.[54] Rings of fuel elements have been defined as different burn-up cells, geometric regions where the material composition and the

Table 3. Different cycle lengths derived from different burn-ups, annual throughput (per core), and time needed to process entire U.K. stockpile. It is assumed that the reactor operates 300 days per year. The processing time is calculated assuming two cores operating simultaneously.

Burn-up	Cycle Length	Throughput MAGNOX	Throughput AGR	Time to Process Entire Plutonium Stockpile
MWd/kgHM	days	t/year	t/year	years
15	196	3.41	4.06	15.67
20	261	2.56	3.05	20.87
25	327	2.05	2.43	26.14
30	392	1.71	2.03	31.34
40	522	1.28	1.52	41.73
50	653	1.02	1.22	52.20
87.5	1142	0.59	0.70	91.30

neutron flux is considered to be constant during one-time step. The total number of burn-up cells is nine, six in the IFZ and three in the OFZ.

GE Hitachi claims that 20 years would be enough to render the U.K. civil plutonium stockpile proliferation resistant, using short irradiation times.[55] Consequently, besides the targeted burn-up of 87.5 MWd/kgHM for the UNF core,[56] this analysis simulated other short cycle lengths including 15, 20, 25, 30, 40, and 50 MWd/kgHM for both a PRISM model fueled with plutonium from MAGNOX reprocessing and a PRISM model fueled with plutonium from AGR reprocessing, despite the fact that these might not be optimum modes of operation for electricity production.

As each fuel assembly contains 57.1 kg heavy metal, the reactor contains a total fuel load of 10.96 tons heavy metal. From this, necessary fuel element irradiation times (cycle lengths) can be calculated, and are summarized in Table 3. A single full cycle was divided into 8 burn-up time steps. At the beginning of each of these time steps, Monte Carlo simulations to estimate a new neutron flux distribution were carried out. If the reactor is fueled with plutonium from MAGNOX origin, a full, fresh reactor core contains 2.23 tons of plutonium. For a reactor fueled with AGR plutonium, the plutonium fraction is higher, hence a full core contains 2.65 tons of plutonium. The table also lists the annual throughput for the different fuels using one reactor core. It also includes the time needed to irradiate the complete U.K. plutonium stockpile, 85.8 tons fuel originating from MAGNOX reprocessing and 23.6 tons fuel originating from AGR reprocessing (cf. previous chapter). In this case, two simultaneously operating cores operating are assumed. The values in the table are calculated assuming operation of the reactor at 840 MWth with a capacity factor of 82.2% (300 days per year). GE Hitachi has claimed that the U.K. plutonium could be disposed in 60 years using two reactor blocks.[57] For the model and operation as described here, the maximal burn-up to allow for full stockpile irradiation would be 57.5 MWd/kgHM.

To avoid large variations in core multiplication and the spatial neutron flux distribution, after one-fourth of a full cycle length one-fourth of the fuel assemblies are dischargs and replaced by fresh fuel assemblies. This frequent fuel element replacement results in a lower capacity factor than might otherwise be achieved. Figure 2 shows the distribution of different batches in fuel zones. To estimate the change in composition of fuel at an equilibrium level, i.e., when the core is loaded with four different sets of fuel elements, each of a different burn-up, three full reactor cycles were simulated, starting with a core filled with only fresh fuel elements. For further analysis, the composition of fuel elements from the third cycle of the first batch was used.[58]

As the isotopic composition will vary in different fuel elements placed in the core, multiple regions with different compositions have been calculated. Eight different geometric areas have been defined, based on rings of fuel elements. All fuel elements in such a ring are assumed to have the same isotopic composition. The innermost ring, Ring 1, surrounds the central control rod. The outermost ring, Ring 8, is the ring directly adjacent to the reflector elements. Rings 1 to 5 form the IFZ, Rings 6 to 8 the OFZ. To account for cooling periods after discharge from the core, the decay is directly simulated as periods of zero neutron flux. The analysis considered cooling periods up to 200 years after the end of reactor operation for decay calculations.

Resulting change of fuel isotopic composition

At the start of simulations fresh metallic fuel consists of 10 wt% zirconium, 18.3 wt%/21.9 wt% plutonium of MAGNOX origin and the remaining part uranium (21.8 wt% and 26.0 wt% for plutonium of AGR origin). Figure 3 shows the relative reduction of plutonium at the end of the different simulated cycle lengths. Overall, the initial plutonium is only reduced by 15 to 26% of its original content–while initially available plutonium is fissioned, new plutonium is bred through neutron captures in the uranium in the fuel. Plutonium from AGR spent fuel reprocessing sees a higher reduction because it contains less plutonium-239 and plutonium-241 in exchange for other plutonium isotopes, which have significantly lower fission cross sections below 1 MeV but can absorb neutrons in other reactions. To achieve the same power output, a higher neutron flux is needed to achieve the same total number of fissions.[59]

As shown in Table 4, the plutonium isotopic composition for both fuel types changes only slightly over burn-up. The isotopic composition starts as reactor-grade plutonium, and plutonium-239 fractions are further reduced during the time in the reactor. For short irradiation times,

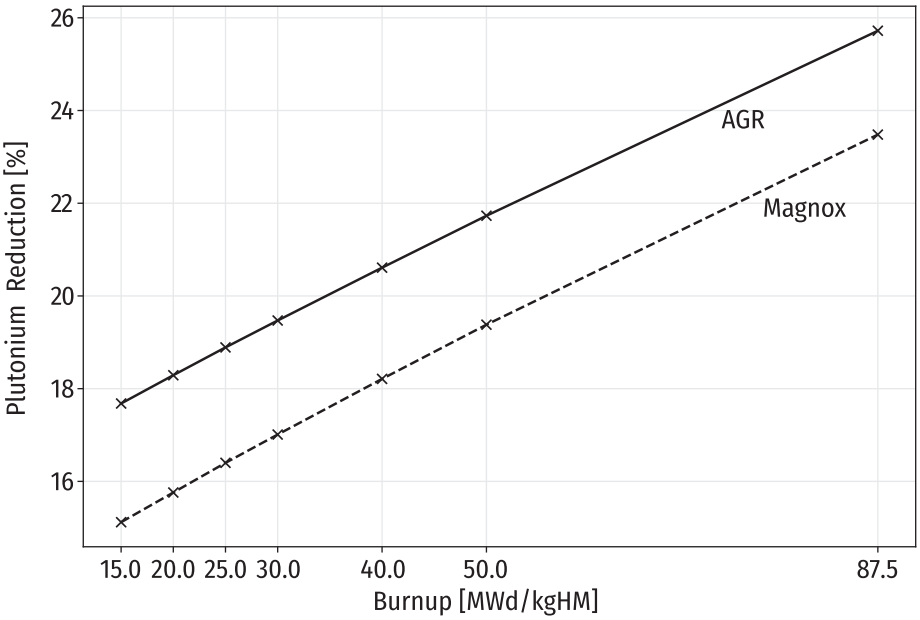

Figure 3. Reduction of plutonium inventory for different burn-up levels. The figure shows the relative change of the mass of the major five plutonium isotopes.

Table 4. Isotopic composition of fresh fuel and after irradiation. Listed are values for Ring 1 (innermost ring) and Ring 8 (outermost ring), values are fractions of total sum of listed isotopes.

Burn-up MWd/kgHM	Ring Nr.	Pu-238 wt%	Pu-239 wt%	Pu-240 wt%	Pu-241 wt%	Pu-242 wt%	Am-241 wt%
AGR							
Fresh fuel		0.50	53.86	30.84	3.49	5.02	6.29
30	1	0.81	52.8	31.91	3.81	5.22	5.44
30	8	0.67	52.85	31.7	3.61	5.17	6.02
87.5	1	1.67	51.2	33.29	4.29	5.5	4.05
87.5	8	1.24	50.94	33.09	3.85	5.42	5.45
MAGNOX							
Fresh fuel		0.02	72.07	22.58	1.04	0.93	3.36
30	1	0.23	69.29	24.77	1.79	1.06	2.85
30	8	0.13	70.32	23.96	1.42	1.0	3.17
87.5	1	0.78	64.75	28.12	2.9	1.35	2.09
87.5	8	0.49	66.97	26.46	2.1	1.16	2.83

e.g., 30 MWd/kgHM (as would be used to minimize the throughput time) the plutonium-239 content is reduced by 3 percent points for MAGNOX fuel and about 1% for AGR fuel. Even after the proposed possible irradiation time of 87.5 MWd/kgHM, plutonium-239 contents are at maximum reduced by 8 percent points in Ring 1 for MAGNOX-based fuel. The small changes occur because fact that relevant amounts of plutonium are bred from uranium-238, so the isotopic vector is "replenished" starting with plutonium-239, and higher mass isotopes are created through neutron captures.

Evaluation of the reactor-based disposition option

Spent fuel dose rate estimates

Using the isotopic compositions generated with the calculations presented in the previous section, the dose rate from gamma radiation of a single fuel element was calculated. Separate calculations were carried out for elements originating from each of the different burn-up cells–the rings of fuel elements in the reactor. Only the dominant gamma emissions from the spent fuel elements were considered in calculating the resultant dose rate. For these calculations, MCNP6 was used.[60] MCNP6 allows a particle source to be defined based on a material mixture. The source emits gamma particles with energies of the different decays of radioactive isotopes in this mixture, using the $par = sp$ parameter in the source definition code. A volume source was implemented to properly account for the distribution of the spent fuel in the fuel rods.[61]

The activity of the source material was estimated based on the activity output MCNP6 calculated for the material. The gamma particle flux was tallied at 1 meter from the surface of the fuel element with a F5 ring tally. Energy bins were defined using the same energy bins used for fluence-to-dose-rate-conversion-coefficients, as evaluated and tabled by the International Commission on Radiological Protection (ICRP).[62] Conversion coefficients depend on the analyzed situation. Here, the coefficients for the ambient dose rate were used, providing a conservative (typically lower) estimation of the actual dose rate used in radiation protection. Dose rates were calculated based on the binned particle flux tally, the respective coefficients, and the calculated activity.

Proliferation resistance of spent fuel

One of the key promises of the use of PRISM for the disposition of U.K. plutonium is that it could render the full stockpile "proliferation resistant" within 20 years. One indication for a material's proliferation resistance is the dose rate due to the material's radioactive decay. Often called the Spent Fuel Standard, a common definition describes a material emitting a dose of 1 Sv/h after 30 years of decay as very unattractive to a possible proliferator. This metric was used to evaluate GE Hitachi's claim. Dose rates shown in the following are dose rates from a single fuel element in air at 1 meter.

Figure 4 shows the development of a fuel element's dose rate over time, for fuel from different PRISM cycle lengths. The outermost fuel elements in Ring 8 receive the lowest irradiation, and are the fuel elements for which dose rates will first drop below the limits described above. Ring 1, the

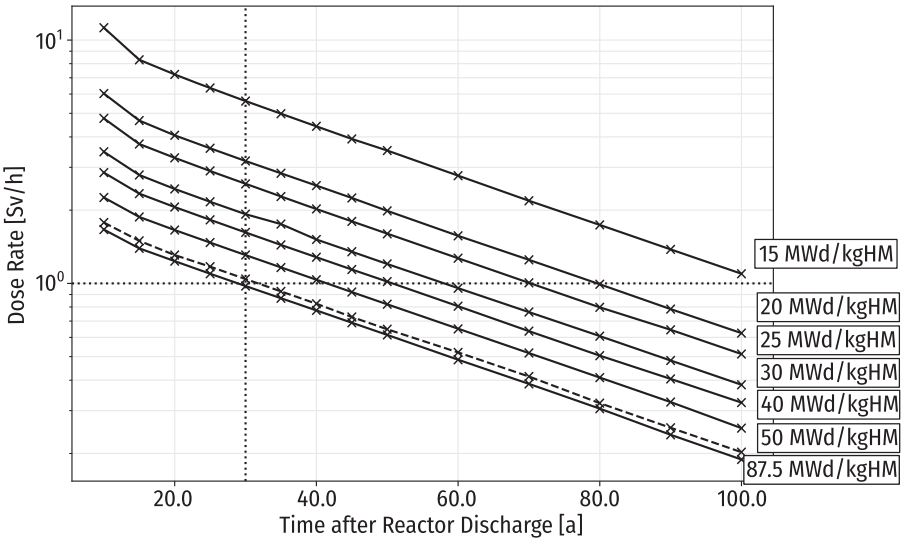

Figure 4. Development of ambient dose rate in 1 m distance of a single fuel element for PRISM fueled with plutonium originating from MAGNOX origin. The solid lines shown are for Ring 8, the outermost ring of the reactor, the dashed line shows Ring 1 for a burn-up of 30 MWd/kgHM.

innermost ring, is subject to the highest neutron flux and throughout the calculations has the highest dose rate, it is only shown for one burn-up value in the figure. Fuel from both origins shows very similar behavior, the dose rate is relatively independent of the initial plutonium composition. As can be seen in the figure, shortly after irradiation, spent fuel of every cycle lengths emits a very high dose rate, leading to lethal dose levels in minutes.

Figure 5 shows the time after which spent fuel dose rates fall below the level of 1 Sv/h as a function of burnup rate for different fuel types from different locations in the reactor block. The figure also shows an average dose rate calculated based on the dose rates per fuel element weighted by the number of these fuel elements in a full reactor core. It becomes apparent that the source of the plutonium in the fuel (AGR or MAGNOX) has only a small influence on the resulting dose rates, as the total number of required fissions is set by the total energy produced in both cases, and the fission yields for different actinides are very similar. Hence to achieve the standard of 1 Sv/h after a cooling period of 30 years for all fuel elements in a reactor, burn-up levels of 30 MWd/kgHM and above are required. A 2014 U.S. DOE report calculating dose rates of excess weapon-grade plutonium disposition in a PRISM-type reactor found similar results: In their model, a burn-up averaged overall fuel elements of 2.7 atom% (equivalent

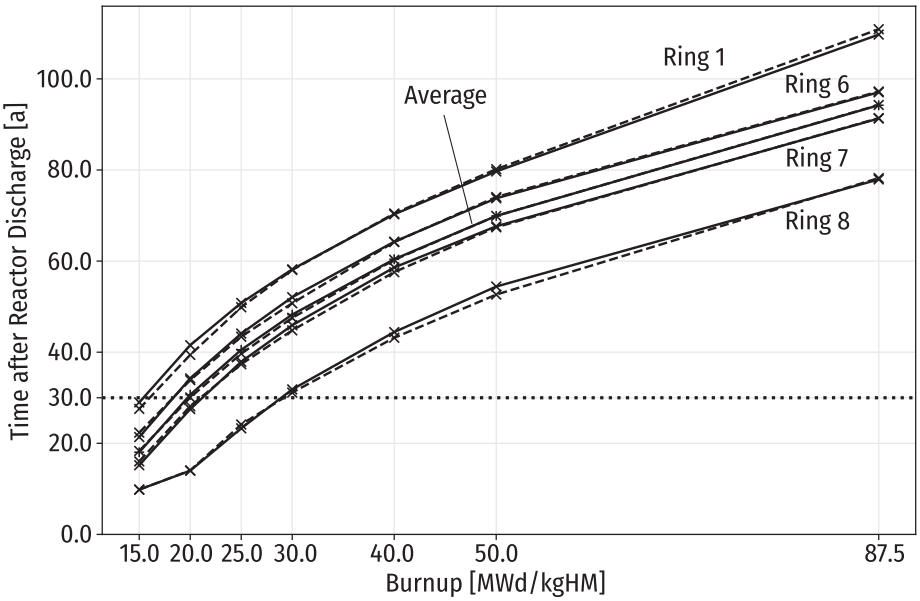

Figure 5. Time for which the emitted dose rate is higher than 1 Sv/h for different reactor areas. Values connected by solid lines are for MAGNOX-based fuel, values connected by dashed lines are for AGR-based fuel.

to 24.6 MWd/kgHM) would emit a dose rate of more than 1 Sv/h after 30 years of cooling.[63] A burn-up of 30 MWd/kgHM is equivalent to an irradiation time of 392 days (cf. Table 3). Operating the reactor 300 days per year, it would take two reactor blocks 31.3 years to transform the separated plutonium into spent fuel that has a radiation barrier of more than 1 Sv/h after 30 years, a period significantly longer than the 20 years claimed by GE Hitachi.[64] However, the calculation presented here did not consider the most optimal case. Additional fuel element exchanges, adding more maintenance time, could lead to a more evenly distributed radiation barrier among the discharged elements. Alternatively, load fraction of the reactor could be increased above 300 days in a year, resulting in higher throughput of fuel.

In addition, as visible in Figure 5, Ring 7, the second outermost ring of fuel elements, achieves the radiation level goal much earlier, after a burn-up slightly higher than 20 MWd/kgHM. Such a burn-up corresponds to a total irradiation time of 20.87 years, closer to GE Hitachi's claim. However, Ring 8, which accounts for 21.9% of the total fuel and an even higher fraction of plutonium because of the higher fraction in the OFZ, does not achieve the standard at that burn-up. Hence, overall, it seems implausible that all plutonium could be rendered proliferation resistant in 20 years using two PRISM reactor cores.

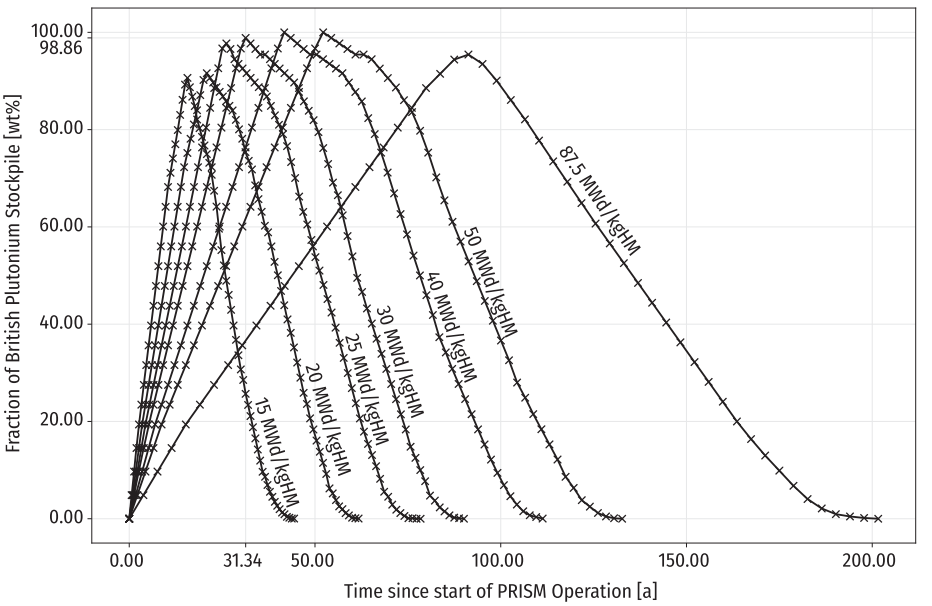

Figure 6. Fraction of the U.K. plutonium stockpile that at the given time emits gamma radiation resulting in a dose rate of more than 1 Sv/h.

Long-term effects on the U.K. Plutonium stockpile

As has been shown above, it would take a significant amount of time to irradiate the full U.K. plutonium stockpile to a level that exhibits a significant radiation barrier. Even very short irradiation cycles, such as 15 MWd/kgHM would lead to a processing time of decades and would not provide a sufficient radiation barrier. Using the shortest cycle length achieving the goal of 1 Sv/h for all fuel elements, irradiation of the total U.K. stockpile would need more than 30 years to complete. It is important to note that while some plutonium is being irradiated, the original radiation barrier of the previously irradiated spent fuel decreases over time due to radioactive decay. A more detailed analysis of this effect is shown in Figure 6. The figure shows the fraction of the total stockpile which has a radiation level of more than 1 Sv/h as a function of time. The figure considers the varying times radiation levels are retained based on the location of the fuel elements in the reactor.

Remarkably, the figure shows that independent of the irradiation time in the reactor, there will never be an extended time period when the full U.K. stockpile has a radiation barrier as defined above. Instead, the fraction of spent fuel having a radiation barrier peaks when the last batch of fuel is unloaded from the reactor. After that the barrier decays away. Although longer irradiation times in the reactor lead to higher radiation levels in individual fuel elements, increasing irradiation times does not improve the

overall situation. As it takes longer to irradiate remaining fuel, radiation barrier in already irradiated fuel has a longer time to decay. The cycle length of 30 MWd/kgHM selected above reaches a maximum value of 98.86% after 31.3 years. However, ninety years after the first plutonium was loaded in a reactor, none of the spent fuel has a significant radiation barrier left because of decay. For the 50 MWd/kgHM case, it takes about 30 years to generate a radiation barrier in 50% of the stockpile; in 100 years after first irradiation the fraction again drops below that level. For a target burn-up of 20 MWd/kgHM–which could be enough for fuel elements except for the outermost ring–the maximum fraction is slightly higher than 90%, and after 63 years, all spent fuel elements emit less than 1 Sv/h.

These findings show that, assuming the radiation barrier is a sufficient proliferation barrier, irradiation in a PRISM type fast reactor would never provide such a barrier to the full stockpile. Irradiation in reactors could be used as a temporary measure to eliminate illegitimate access to plutonium–with a final, sealed repository as the long-term option–but would only give a few decades of additional protection.

Conclusion

The paper first presented detailed estimates of the history and the future of the U.K. plutonium stockpile. When the operations of the U.K.'s two plutonium reprocessing plants end, the country will have a total of 109.4 tons of separated plutonium produced from domestic spent fuel. It is estimated that approximately one-fifth of this plutonium originated from AGR reactor spent fuel, the rest from MAGNOX reactor spent fuel. Realistic estimates for the stockpile's isotopic content have been presented, which include the fraction of americium-241, which is produced through the decay of plutonium-241. In 2030, plutonium separated from MAGNOX spent fuel on average contains more than 70% plutonium-239 and approximately 3.35% americium-241. Plutonium separated from spent MAGNOX fuel contains more than 50% plutonium-239 and approximately 6.27% americium-241.

Using publicly available literature, a simulation model of the PRISM reactor was prepared to simulate the irradiation of spent nuclear fuel. According to the description, the model has a breeding ratio smaller than one, and the simulations show that depending on the target burn-up level between 15% and 24% of the original plutonium is fissioned when irradiating MAGNOX origin plutonium, and 17% to 26% of the plutonium of AGR reactor origin. In all cases, the resulting plutonium vector remains reactor-grade plutonium. With a burn-up of 50 MWd/kgHM, using two

PRISM cores, operating 300 days per year, the 109.4 tons could be irradiated in approximately 52 years.

Estimates of the radiation barriers of the different spent fuel elements were made by simulating their photon emissions. Dose rates were calculated at 1 m from the center of the fuel element. The Spent Fuel Standard of 1 Sv/h at 1 m was used as the metric to determine a sufficient radiation barrier to potential proliferators. The model determined that achieving a dose rate higher than that standard for fuel elements at all reactor positions is only possible for burn-up levels of 30 MWd/kgHM and more, and that irradiating all plutonium to that that level will take 31.3 years.

This finding contradicts one of the key claims of PRISM's vendor, GE Hitachi, that the reactor would be capable of rendering the U.K. stockpile proliferation resistant in 20 years. Even a span of 30 years, as was mentioned in earlier publications, might not be enough. The time to achieve the radiation barrier is dominated by the irradiation time required for the outermost ring of fuel elements, which account for more than one-fifth of all reactor elements in the core. Reactor operations could be optimized to expose these fuel elements to higher neutron flux levels. However, the impact on reactor operation of additional fuel element changes to achieve higher radiation barriers would extend necessary reactor outages.

An additional analysis showed that irradiation of fuel using a PRISM-based reactor option would at no point in time provide adequately high radiation barriers to proliferation for 100% of the stockpile. Once irradiation is complete, the fraction of the stockpile meeting the standard quickly decreases because of decay. Even with a burn-up as high as 50 MWd/kgHM, only half of the spent fuel has a radiation barrier for more than four decades after all fuel has been consumed. In the 30 MWd/kgHM case, 90 years after irradiation has started, none of the resulting spent fuel would be protected anymore. These findings show that irradiating separated plutonium in a reactor could, at best, serve as an interim solution until a final repository has been found. The benefit of a reactor-based deposition option to achieve proliferation resistance of spent fuel is marginal.

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total U.K.'s energy consumption would be approx. 586 EJ (5.86×10^{20} *J*). Assuming that every single fission releases 200 MeV of energy or 3.2×10^{-11} *J*, 1.83×10^{31} plutonium atoms would need to be fissioned to produce 100 years of U.K.'s energy demand. Given that there are about 2.5×10^{27} atoms per ton of plutonium, such an energy production would require the fission of 7,286 tons of plutonium.

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Appendix

This appendix describes the assumptions used for the estimate of the U.K. plutonium stockpile. From 1996 onwards, the main source of the stockpile size are the statements by the government as part of its INFCIRC 549 declarations. The total amount of U.K. plutonium was separated by reactor type. The amount originating from ownership changes of

formerly foreign plutonium was identified first. In recent years, the U.K. has conducted title swaps for plutonium on its soil that was produced by reprocessing foreign spent fuel (cf. Table 5). Secondly, the plutonium resulting from spent MAGNOX fuel reprocessing was calculated based on throughput figures for the B205 plant. Any remaining plutonium was assumed to be plutonium resulting from reprocessing of AGR spent fuel.

Throughput figures for the B205 reprocessing plant were taken from a report by the non-governmental organization "Cumbrians Opposed to a Radioactive Environment."[65] The amount of plutonium in spent fuel is dependent on the burn-up in the fuel, however publicly available data on recent fuel burn-up is limited. The analysis therefore assumed a burn-up of 5,000 MWd/tHM. Reported production estimates for the years 1985–1995 achieved an average of 4,359 MWd/tHM.[66] Choosing this higher value takes into account possible improvements in reactor operations (as were visible over the operation history) and serves as an upper bound. To estimate the amount of plutonium per tU of spent fuel, an estimation that was used in other references is applied:[67]

$$pu_{tot} = 0.9235B^{0.6946}$$

According to this equation, a ton of spent fuel with a burn-up of 5,000 MWd/tHM contains 2.8 kg of plutonium. Calculating plutonium production based on the B205 throughput values, there are four years for which the calculated plutonium production from MAGNOX reactors is higher than the total declared increase in U.K. plutonium stockpiles. This is probably caused by processing times in the reprocessing plant. To correct for this, it is assumed that THORP plutonium production from AGR fuel in that year is zero. The missing plutonium is taken equally from the respecting two following years (equivalent to assuming that the declarations are delayed).

For the period prior 1996, it is assumed that only plutonium from reprocessing MAGNOX fuel is added to the U.K.-owned stockpile. Plutonium arising from U.K. MAGNOX fuel until 1995 are listed in the book from Albright et al. (1997, Table C.1).[68] As the values do not exactly match the official INFCIRC 549 statement from 1996, they have been adjusted by a factor of 0.910.[69]

Table 5. Title swaps for separated plutonium in the U.K. References are given in the endnotes listed for the respective years. In 2013, a swap of 1,850 kg Plutonium "originally allocated to repay plutonium loans" was announced, without specifying the previous owner of that plutonium.

Year	Original Owner	Amount transferred (kg)
2012	Germany	4,000
2013	Germany	650
	Netherlands	350
		1,850
2014	Sweden	800
	Germany	140
2017	Spain	600

Notes: The 2017 swap included 5 kg from Germany, which are omitted here. Sources: World Nuclear News, "German plutonium stays in U.K.," 13 July 2012, http: //www.world-nuclear-news.org/WR-German_plutonium_to_stay_in_UK-1307127.html; Michael Fallon, Management of overseas owned plutonium in the U.K., Written Statement (Government of the United Kingdom, 23 April 2013), https://www.gov.uk/government/speeches/written-ministerial-statement-by-michael-fallon-management-of-overseas-owned-plutonium-in-the-uk; David Lowry and Mycle Schneider, "U.K. decision to take over foreign plutonium raises safeguards questions," 14 July 2014, http://fissilematerials.org/blog/2014/07/uk_decision_to_take_over_html; Parliamentary Under Secretary of State, Minister for Energy and Industry Jesse Norman, Management of Overseas Owned Plutonium in the U.K.: Written statement, Written Statement HCWS422 (Government of the United Kingdom, 19 January 2017), http://www.parliament.uk/business/publications/written-questions-answers-statements/written-statement/Commons/2017-01-19/HCWS422/.

Table 6. Plutonium isotopic vectors for Magnox and AGR spent fuel. All values are given in wt%.

	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
MAGNOX 3 MWd/kgHM	0.1	80	16.9	2.7	0.3
MAGNOX 5 MWd/kgHM		68.5	25	5.3	1.2
AGR 18 MWd/kgHM	0.6	53.7	30.8	9.9	5.0

Notes: Isotopic composition according to M Beauvy, "Solid State Chemistry and Thermophysical Properties of Actinides," in Actinides and the Environment, ed. P. A. Sterne, A. Gonis, and A. A. Borovoi, NATO Advanced Science Institute (1996).

Table 7. Isotopic composition of the British plutonium stockpile based on estimates presented in this chapter.

	Isotopic Composition (wt%)						
Year	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	Mass (tons)
Plutonium from spent MAGNOX fuel							
2000	0.04	73.69	21.29	2.25	0.79	1.84	56.2
2010	0.03	72.47	22.13	2.02	0.89	2.32	73.1
2020	0.02	71.87	22.53	1.69	0.93	2.75	85.8
2030	0.02	71.85	22.51	1.04	0.93	3.35	85.8
2050	0.02	71.81	22.46	0.40	0.93	3.88	85.8
2100	0.01	71.71	22.34	0.04	0.93	3.92	85.8
Plutonium from spent AGR fuel							
2000	0.59	53.70	30.79	8.90	5.00	1.00	5.5
2010	0.56	53.69	30.78	6.98	5.00	2.89	14.7
2020	0.54	53.68	30.76	5.64	5.00	4.20	23.6
2030	0.50	53.67	30.73	3.48	5.00	6.27	23.6
2050	0.43	53.64	30.66	1.32	5.00	8.19	23.6
2100	0.29	53.57	30.51	0.12	5.00	8.71	23.6

After 2014, no reprocessing throughput figures are known, and no plutonium declarations have been made after 2016. The Nuclear Decommissioning Agency stated that in April 2012 3,800 tU of MAGNOX fuel remained to be reprocessed.[70] As the B205 reprocessing plant is supposed to reprocess all MAGNOX fuel produced in the U.K., throughput numbers for B205 were estimated to be evenly distributed to reprocess the given fuel value until 2020. Plutonium separation from MAGNOX fuel is then calculated as described above.

For plutonium produced from AGR fuel after 2016, THORP reprocessing figures[71] are extended, assuming the average annual throughput of the time from 1995–2014 to be the annual throughput for the years 2015–2018. Plutonium production from AGR in 2017 and 2018 is based on these throughputs and the average plutonium production per ton of fuel processed from the earlier years of THORP operation. For these two years, the same calculation generates values for foreign plutonium.

If one assumes a lower burn-up of MAGNOX fuels for the years 1996 onwards, the method as described would yield a higher fraction of AGR based fuel. For example for a burn-up of 4,000 MWd/tHM, the stockpile in the year 2020 and later would include 80.8 tons plutonium from MAGNOX spent fuel reprocessing and 28.3 tons of plutonium from AGR reprocessing. Since the total amount through 2016 is based on the INFCIRC 549 declarations, the final difference is small. For all calculations in this article, the estimate based on 5,000 MWd/tHM is used.

Based on the data collection above, isotopic compositions of the U.K. plutonium stockpile were calculated. This was done separately for plutonium coming from MAGNOX fuel

reprocessing and for plutonium coming from AGR reprocessing. Spent fuel immediately after reprocessing was assumed to have isotopic vectors for spent fuel as shown in Table 6. Before adding new plutonium for a given year, the existing plutonium stockpile was modified to reflect the decay of radioactive isotopes over one year, mostly leading to americium-241 build-up. The results for the stockpile composition for selected years are listed in Table 7. The decay products were calculated using the Transmuter function of PyNE, a python toolkit for nuclear engineering.[72] It can be seen that over a century, most plutonium-241 decays into americium-241. The other isotope fractions remain relatively constant.