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Research Article

A new approach to the recycling of spent nuclear fuel in thermal reactors within the REMIX concept*

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Abstract

A review of simulated nuclear fuel cycles with mixed uranium-plutonium fuel (REMIX) was carried out. The concept of REMIX fuel is one of the options for closing the nuclear fuel cycle (NFC), which makes it possible to recycle uranium and plutonium in VVER-1000/1200 thermal reactors at a 100% core loading. The authors propose a new approach to the recycling of spent nuclear fuel (SNF) in thermal reactors. The approach implies a simplified fabrication of mixed fuel when plutonium is used in high concentration together with enriched natural uranium, while reprocessed uranium is supposed to be enriched and used separately. The share of standard enriched natural uranium fuel in this nuclear fuel cycle is more than 50%, the share of mixed ^{nat}U+Pu fuel is 25%, the rest is fuel obtained from enriched reprocessed uranium. It is emphasized that the new approach has the maximum economic prospect and makes it possible to organize the fabrication of this fuel and nuclear material cross-cycling at the facilities available in the Russian Federation in the short term. This NFC option eliminates the accumulation of SNF in the form of spent fuel assemblies (SFA). SNF is always reprocessed with the aim of further using the primary reprocessed uranium and plutonium. Non-recyclable in thermal reactors, burnt, reprocessed uranium, the energy potential of which is comparable to natural uranium, as well as secondary plutonium intended for further use in fast reactors, are sent as reprocessing by-products to the storage area.

Keywords

REMIX, spent nuclear fuel, nuclear fuel cycle, mixed fuel, uranium-plutonium fuel, plutonium, reprocessed uranium, recycling, cross-cycling

Introduction

The reprocessing of spent nuclear fuel is considered with the aim of using the energy potential of the remaining uranium and plutonium produced, eliminating the SNF storage and previously accumulated reprocessed products, and saving natural uranium (Fedorov et al. 2001). One of the main problems in recycling uranium recovered from SNF is the accumulation of ^{232}U radionuclide, which generates a chain of short-lived powerful gamma emitters (Smirnov et al. 2011). For a similar reason (^{236}Pu decaying into ^{232}U), the isolated plutonium needs to be deeply cleaned before being refabricated into MOX or REMIX fuel. Another reason that limits uranium recycling is the accumulation of ^{236}U, which is a neutron absorber. The presence of even isotopes in the fuel composition requires additional enrichment, which reduces the fuel cycle efficiency (Pavlovichev et al. 2006).

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There are about 24 thousand tons of SNF accumulated in the Russian Federation. Approximately 650–700 tons of SNF are unloaded from the reactors of Russian NPPs annually, while no more than 15% of this volume is reprocessed (Khaperskaya 2019). Traditionally, reprocessed nuclear materials have been used separately in Russia. Reprocessed uranium is enriched and used mainly in RBMK reactors (Kislov et al. 2012). Plutonium is accumulated in order to launch the fast reactor program; at present, the production of MOX fuel for the BN-800 has begun (Vergazov 2019).

In Russia, the concept of a two-component nuclear energy system is adopted, which includes both types of reactors (with a thermal neutron spectrum – VVER and a fast neutron spectrum – BN). Within this nuclear energy system, it is supposed to use a partial core loading with MOX fuel (~ 9% Pu mixed with depleted uranium) in thermal reactors. The transition period before the start of reactors with a fast neutron spectrum may consist in recycling reprocessed nuclear materials as a mixed fuel for thermal reactors, possibly more efficient than MOX fuel, with a partial core loading.

Simulating nuclear fuel cycles

Since 2005, the V.G. Khlopin Radium Institute in collaboration with the Kurchatov Institute has been developing REMIX fuel (REMIX – REegenerated MIXture of U-Pu oxides), which involves recycling both reprocessed uranium and plutonium to load the entire core of existing VVER-1000/1200 reactors (Fedorov et al. 2001, Pavlovichev et al. 2006, Pavlovichev et al. 2008, Zilberman et al. 2012, Dekusar et al. 2013, Postovarova et al. 2016).

The authors of the article carried out numerous calculations of nuclear fuel cycles using software that simulates nuclear fuel burnup based on the Monte Carlo method. The main tool in the calculations was SCALE 6.2 (SCALE 2019) – a software package that provides calculations of criticality, fuel burnup, activation of materials, characteristics of radiation sources and radiation protection. The calculations were carried out using a library of continuous dependences of the neutron interaction cross-sections on energy, based on the evaluated nuclear data files ENDF/B-VII.1. In the simulation, a three-dimensional model of the infinite core of a reactor using TVS-2M fuel assemblies was used. The neutron multiplication coefficient during fuel burnup was maintained at $k_{eff} = 1$ by controlling the boron concentration in the coolant.

Various NFC schemes were simulated using mixed uranium-plutonium fuel. The analysis considered a group of Russian-designed thermal reactors of the VVER-1000/1200 type throughout the entire period of their operation. After the fuel burnup and cooling, the SNF isotopic composition was evaluated for the possibility and expediency of its further use. With a residual energy potential of the SNF higher than that of natural uranium, the nuclear materials of this fuel are used in simulating the next recycle.

The authors investigated the basic NFC options and developed a new one based on mixed REMIX uranium-plutonium fuel.

REMIX-A

The basic option is REMIX-A (Gavrilov et al. 2019). When spent nuclear fuel is being reprocessed, recovered uranium and plutonium are separated (optionally completely) and re-mixed in solution in the initial ratio, and the necessary energy potential is provided by a 19% ^{235}U makeup within the nuclear non-proliferation limits. This fuel composition contains about 1–2% of plutonium extracted from SNF. Such fuel can be multiply recycled. When SNF is reprocessed completely, about 15% of the excess reprocessed uranium is formed on each recycle. If a highly enriched uranium makeup is used, then 55–65% recycling occurs without any excess reprocessed uranium being formed.

The REMIX-A recycling scheme is shown in Fig. 1. The recycling duration is limited to seven cycles due to the plutonium isotopic composition degradation during recycling. The first cycle (vertical sequence on the left) is the initial loading of fuel from enriched natural uranium. In this option, after seven cycles, as a result of partial reprocessing and multiple cycling of the reprocessed materials, it is possible to reduce the final accumulated amount of SNF by about four times as compared to the open fuel cycle. If reprocessing is complete (after seven cycles), it is possible to reduce the final accumulated amount of SNF up to seven times (but with the formation of excess reprocessed uranium). At the same time, natural uranium is saved at the level of 25–30%. The share of mixed fuel in the REMIX NFC is 86%. On average, 3.8 kg of minor actinides (MA) are formed from one ton of heavy metal (tHM) of the initial fuel during complete reprocessing, while 1.4 kg of MA is formed from one tHM of fuel from enriched natural uranium.

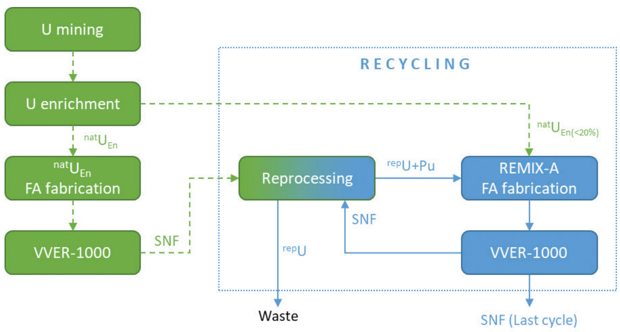

Figure 1. REMIX-A recycling scheme when ^{235}U is enriched up to 20%.

REMIX-C

REMIX-C is a development of REMIX-A that implies directing reprocessed uranium ($^{rep}U_{En}$) from SNF reprocessing to re-enrichment to about 4–5% ^{235}U, followed by its mixing with the calcined plutonium-uranium master mixture left at reprocessing plant. A natural enriched uranium makeup is used. Excess nuclear materials are excluded, but reprocessed uranium should be transported to the enrichment plant and backward.

In terms of fuel supply for reactors being built abroad under Russian projects, the REMIX-A/C concept looks optimal for the entire life cycle, since Russia intends to provide services for reprocessing SNF during recycling and returning nuclear materials to customers, but already in the form of mixed uranium-plutonium fuel and radioactive waste (Nuclear Fuel Reprocessing 2020). The decrease in the accumulation of SNF is at the same level as in the case of REMIX-A with complete reprocessing.

The REMIX-C recycling scheme is shown in Fig. 2. When seven cycles are used in this option, the amount of SNF is reduced by seven times. Natural uranium saving is 30%. The share of mixed fuel in the REMIX NFC is 86%. On average, 3.8 kg of MA is formed from one tHM of the initial fuel.

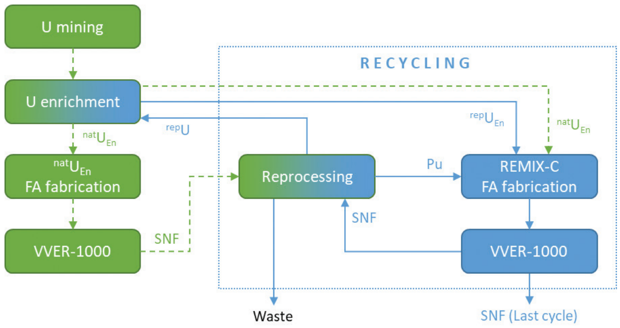

Figure 2. REMIX-C recycling scheme.

REMIX-B

In the REMIX-B option, it is not intended to use a makeup during recycling; accordingly, the amount of mixed fuel and its final SNF are reduced. The Pu content in this fuel can exceed 4%. After the fuel burnup of ~ 47 GW day/tHM, approximately 1.25% of plutonium is formed from enriched natural uranium. Accordingly, for the production of mixed fuel with a plutonium content of 4%, it is necessary to reprocess three to four times more SNF. To achieve the necessary effective fuel enrichment, the reprocessed uranium enrichment method is used. The fabrication of REMIX-B fuel requires great precision in mixing the components; however, the amount of secondary fuel is several times less than the original SNF from natural uranium.

The REMIX-B recycling scheme is shown in Fig. 3. Since there is no makeup, the fuel is "compressed" and the plutonium concentration is increased on each recycle. This option is limited in the number of recycles due to the high plutonium content. The plutonium content starts from 4% in the first recycle and from 8% in the second recycle. With one uranium loading and one REMIX-B cycle, the amount of SNF is reduced, as in the case of seven REMIX-A cycles (with partial reprocessing), by four times. When two recycles are used, the amount of SNF is reduced by more than 10 times as compared to the open NFC. Natural uranium is saved at the level of 25–30%. The share of mixed fuel in the REMIX NFC is 20–30%. On average, 2.4 kg of MA is formed from one tHM of the initial fuel. This fuel composition was patented in the Russian Federation (Zilberman et al. 2014).

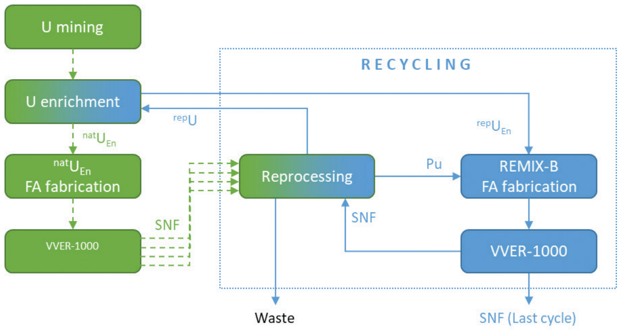

Figure 3. REMIX-B recycling scheme.

New approach: REMIX-E

An estimated economic analysis of NFCs based on prices from open sources (INL/EXT-17-43826 2017) showed that the main contribution to the cost of the nuclear fuel cycle involving recycling nuclear materials is due to the fabrication of mixed uranium-plutonium fuel. The most economically attractive NFC option is the one, in which the share of mixed fuel is lower (NFC, where uranium-plutonium fuel is used in fewer reactors) with comparable savings in natural uranium and reduced amounts of SNF. The high-plutonium REMIX-B option "with compression" during recycling is well suited for these criteria. However, this approach is currently technologically complicated, since it does not use a makeup, and seems unlikely in the near future.

The authors propose a new approach – REMIX-E – that implies a simplified fabrication of mixed fuel when plutonium is used in high concentration together with enriched natural uranium, while reprocessed uranium is supposed to be enriched and used separately. This NFC option seems to be optimal in terms of quick start and makes it possible to reduce the accumulation of SNF at the level of other options. This fuel composition was patented in Russia (Zilberman et al. 2019) and is now being patented abroad.

In the REMIX-E NFC, three types of fuel are used: enriched natural uranium ($^{nat}U_{En}$): 56%, enriched reprocessed uranium ($^{rep}U_{En}$): 18% and mixed fuel ($^{nat}U_{En}$-Pu): 25%. The low share of mixed fuel results from the increased initial plutonium concentration of 5%; the fuel is “compressed”. It is important to note that this ${}^{nat}U_{En}$-Pu fuel uses enriched natural rather than reprocessed uranium. Such production can now be organized at existing MOX production facilities. Once-reprocessed uranium is supposed to be used, as it is done now, at existing facilities in the form of ERU (enriched reprocessed uranium) fuel. Most of the fuel in this NFC is standard enriched natural uranium. Natural uranium is saved at the level of 25–30%. On average, 2.2 kg of MA is formed from one tHM of the initial fuel.

The REMIX-E recycling scheme is shown in Fig. 4. The nuclear material utilization scheme is shown in Fig. 5. The rectangles indicate the fuel used: natural uranium (${}^{nat}U_{En}$), mixed uranium-plutonium (${}^{nat}U_{En}$-Pu) and enriched reprocessed uranium (${}^{rep}U_{En}$), where the index “1” indicates that the reprocessed uranium is primary, i.e., obtained from spent natural uranium fuel, and the index “2” indicates that the reprocessed uranium is secondary. The arrows indicate the material recovered after reprocessing and sent to fabricate new fuel or for storage.

In this NFC, there is no restriction on the cross-cycling duration. By way of illustration (see Fig. 5), we considered several cycles: two of them were with mixed fuel (${}^{nat}U_{En}$-Pu) with “compression”. “Compression” was up to 5% of Pu in the first cycle and up to 10% of Pu in the second cycle. The second (optional) Pu-cycle is used only for re-burning plutonium, after which the number of odd Pu isotopes is almost equal to the number of its even isotopes. If necessary, such plutonium is sent to the storage area for further use in fast reactors. Due to the fact that experimental studies of fuels for VVER-type thermal reactors with Pu> 5% were not carried out, and the calculations predict the impossibility of using this concentration at existing VVER-1000, the cycle marked by the sign “?” in Fig. 5 is considered only theoretically (as a possible option).

The REMIX-E approach makes it possible not to accumulate SNF at all. A distinctive feature of this cycling option is that it generates not SNF but burnt reprocessed uranium, non-recyclable in thermal reactors, which energy potential is comparable with that of natural uranium, and secondary plutonium.

Figures 6, 7 schematically show the stationary loading of 42 power units with REMIX-E fuel in comparison with the low-plutonium REMIX-A fuel. From such a number of VVER reactors, including foreign ones, we can expect SNF to be delivered for reprocessing. This is 800 tHM/year, which is equal to the total optimal productivity of the RT-1 plant (FSUE «PA «Mayak») and PDC (FSUE «MCC») after the planned expansion.

At present, VVER-1000 reactors operate on an 18-month fuel cycle. Every 18 months, 67 fuel assemblies out of 163 are loaded and unloaded (for VVER-1200 – 72 fuel assemblies). Some fuel assemblies have two 18-month fuel cycles in the reactor, and some have three. Each fuel assembly contains 0.465 tHM; therefore, over a period of 3×18 months, 93.5 tHM are used. For calculating material balances, the period of 3×18 months (taking into account overloads and maintenance of about 4.5 years) is taken for a full cycle of a fuel batch, for which all fuel reaches an average burnup of 47 MW×day/kg. While this fuel batch is cooled and reprocessed, and new fuel is fabricated from it, it is necessary for the reactor to operate on exactly the same fuel batch. As a result, the concept of *pair cycles* of fuel batches emerges. Over the period of the reactor operation for about 63 years, approximately seven pair cycles are obtained. Since batches of fresh fuel assemblies are constantly uploaded, the cycles do not go sequentially, but layering on top of each other. The result is an excess of the seven pair cycles. In the concept of reprocessed fuel, enlarged cycles are composed of a zero (uranium) cycle and cycles with fuel recovered from reprocessed nuclear materials. For the REMIX-E concept, the number of recycles does not matter, i.e., it can be indefinite. Even in the second or third cycle, the composition of the REMIX-E mixed fuel stabilizes and does not change further. Note that for REMIX-A/C low-plutonium fuel, which implies multi-recycling, the recycling duration is limited to six or seven recycles due to the plutonium isotopic composition degradation during recycling.

Regarding REMIX-A, it can be said that only 14% of this fleet (i.e., six reactors) should run on natural fuel, while even now the corresponding Russian capacities are supplying such fuel for 30–35 reactors and efforts are under way to expand this production. Suppose that fabricating mixed oxide fuel, we can rely on its supply only for 10–15 reactors, i.e., about 30% of the total capacity or 250 tons/year with a calculated increase in the reactor fleet up to 50–55. Then, it makes sense to consider low-plutonium fuel mixed with reprocessed uranium (REMIX) only for a limited group of 3–5 reactors, including the experimental

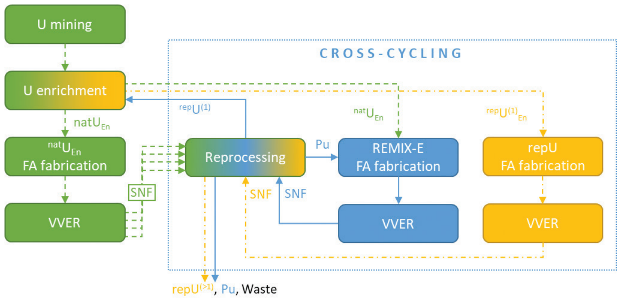

Figure 4. REMIX-E recycling scheme.

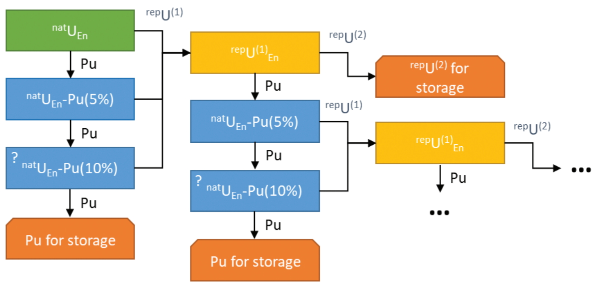

Figure 5. Nuclear material cross-unilization scheme.

Pair cycles	Units																																								
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42
1.1	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U
1.2	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U
2.1	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U
2.2	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U
3.1	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U
3.2	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U
4.1	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U
4.2	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U
5.1	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U
5.2	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O	O	O	O	O	U	U
6.1	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O
6.2	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1	E2	E2	O	O	O	O
7.1	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1
7.2	E2	E2	O	O	O	O	O	O	O	O	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	U	E1	E1	E1	E1	E1	E1

Figure 6. REMIX-E NFC. Stationary loading of 42 power units. U is $^{nat}U_{En}$ fuel; E1 is mixed $^{nat}U_{En}$-Pu fuel (5%) with energy-grade plutonium; E2 is mixed $^{nat}U_{En}$-Pu fuel (10%) with secondary plutonium; O is $^{rep}U_{En}$ fuel.

Pair cycles	Units																																								
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42
1.1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1
1.2	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1
2.1	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2
2.2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2
3.1	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3
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4.2	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4
5.1	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5
5.2	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5
6.1	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6
6.2	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6
7.1	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U
7.2	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U	R6	R5	R4	R3	R2	R1	U

Figure 7. REMIX-A NFC. Stationary loading of 42 power units. U is $^{nat}U_{En}$ fuel; Ri is $^{rep}U_{En}$-Pu fuel (i is the recycle number).

group for the phased development of the closed nuclear fuel cycle or commercially for foreign small groups of reactors provided that this nuclear fuel cycle is economically justified.

If the REMIX-E scenario is used, $^{nat}U_{En}$-Pu fuel accounts for only 15% of the capacities in one Pu-cycle and also 6% in the second Pu-cycle; the main share falls on standard $^{nat}U_{En}$-fuel and partly on enriched reprocessed uranium ($^{rep}U_{En}$).

Conclusion

The authors propose a new approach to recycling SNF in thermal reactors within the REMIX concept. The REMIX-E NFC, as compared to other REMIX concepts, according to our estimates, seems to be optimal in terms of quick start and can completely eliminate the accumulation of SNF in the form of SFA.

A specific feature of the REMIX-E approach is the endless cycling of the same fuel, $^{nat}U_{En}$-Pu and $^{rep}U_{En}$, produced from standard components, i.e., primary reprocessed uranium and primary energy-grade plutonium from standard enriched natural uranium fuel. As a result, the production of such fuel eliminates the problems of mixing SNF from another fuel in one reprocessing chain, as in the case of multi-recycling low-plutonium REMIX fuel; $^{rep}U_{En}$ transporting to the facility for reprocessing SNF and fabricating uranium-plutonium fuel is also excluded.

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