LWR Fuel Cycles' Material and Isotopic Balance

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Abstract. Light water reactors are the most widely used commercial power reactors. Nowadays the nuclear fuel cycle is designed mostly to serve the operation of these reactors. One of the challenges standing before nuclear power worldwide is spent nuclear fuel management, both of historical stockpiles and of the amounts that are currently discharged or will be discharged in the future. Other issues directly influencing the expansion of the reactor fleet and nuclear energy's sustainability are the non-proliferation of nuclear materials and resource availability. The resource availability also defines partially the economics of nuclear power. These three issues should be addressed if nuclear energy's development and competitiveness are to be assured. In order to find applicable solution of these problems and to assure the sustainable development of nuclear energy, fuel cycles' material and isotopic balances should be analysed. In this paper the material feeds and savings in once-through and partially closed LWR cycles are quantified, and spent fuel isotopic composition at discharge is calculated.

Keywords: nuclear fuel cycle, plutonium, spent nuclear fuel, material balance, isotopic composition

1 Introduction

Light water reactors (LWR) are the most common commercial reactors. According to IAEA's Power Reactor Information System (PRIS) data there are 356 operational LWRs as of January 12, 2015. This number accounts for 81.5% of the total world nuclear fleet.276 of these reactors are pressurized water reactors (PWR) that represent 63.2% of all nuclear reactors. In this number the Russian design of PWR – the WWER type of power reactors, are included as well. Other 80 reactors are boiling water reactors (BWR) and they represent 18.3% of the total nuclear fleet.LWRs' total gross installed electric capacity is 331 463 MW (88.4% of the total world gross installed capacity). PWRs account for 256 110 MW of this capacity (this means 68.3% share of total gross capacity and 77.3% share of LWRs' installed capacity). Currently there are 71 power reactors under construction with total installed capacity of 68 136 MW; 64 (90.1%) of them are light water reactors with total capacity of 64 157 MW (94.2% of all reactors' under construction total capacity). 60 reactors with installed capacity of 60 232 MW are pressurized water reactors [1].

2 Nuclear Power's Development

The main forecasts for nuclear power's development generally foresee increase in nuclear power generation. OECD's International Energy Agency (IEA) expects 60% increase in gross installed nuclear capacity by 2040, thus reaching 624 000 MW [2], and British Petroleum's forecasts that 860 Mtoe of primary nuclear energy will be generated in 2035 compared with 560 Mtoe generated in 2012 (Figure 1), meaning that primary energy

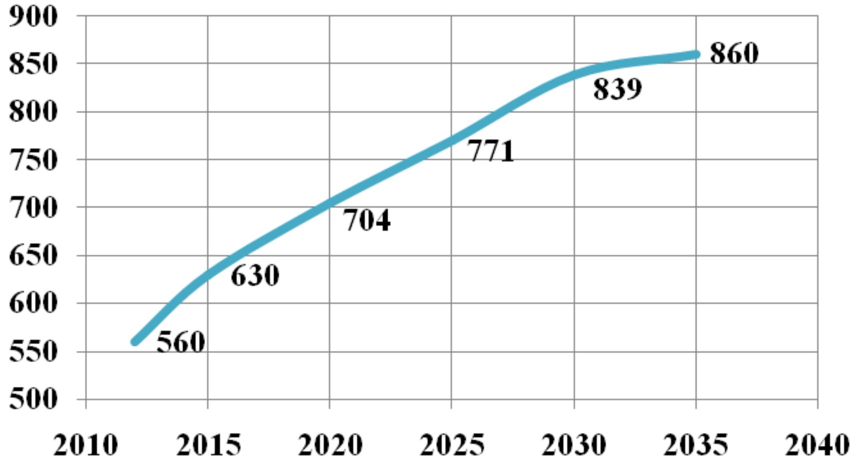

Figure 1. World primary nuclear energy production anticipated growth by 2035, Mtoe [3].

Table 1. Number, type, and electric power capacity of the operational and under construction reactors as of January 12, 2015 [1]

Reactor type	Operational		Under Construction	
	Number	Gross electric power capacity MW	Number	Gross electric power capacity MW
BWR	80	75 353	4	3 925
FBR	2	580	2	1 259
GCR	15	8 045	—	—
LWGR	15	10 219	—	—
PHWR	49	24 592	4	2 520
PWR	276	256 110	60	60 232
HTGR	—	—	1	200
TOTAL	**437**	**374 899**	**71**	**68 136**

generated by nuclear sources will increase by a rate of 1.9% annually worldwide. Nuclearprimaryenergyproduction growthinnon-OECDcountrieswilladd up to 5.9% annually [3].

The two main drivers of the expected worldwide nuclear power generation growth are (1) world energy demand growth and (2) the need to decrease global CO_2 emissions. It is expected that between 2012 and 2035 total world primary energy consumption will grow by a rate of 1.5% annually, thus reaching demand of 17 566 Mtoe. Total primary energy demand growth will be 41% for the whole period [3]. At the same time nuclear power generation is the second most potent technology for power generation related CO_2 emissions mitigation. In 2011 nuclear power prevented about 2.1 Gt CO_2-eq being emitted in the atmosphere, hydro power saved 2.8 Gt CO_2-eq, and all other renewable energy sources – 0.8 Gt CO_2-eq. It is estimated that if nuclear power share in world electricity generation rises from its 2005 level of 16% to 18% by 2030, the annual potential for CO_2 emissions reduction would be 1.88 Gt [4].

3 Spent Nuclear Fuel (SNF) Management

Nuclear facilities' operation has also some drawbacks, the main being radioactive waste (RAW) generation. Generally, RAW are subject to reprocessing and conditioning;some of them under certain circumstances could be exempt from regulatory supervision. Some of the generated RAW, however, are highly radioactive and their management sets high requirements for extremely long time frames. Such highly active RAW are the long-lived fission products contained in the spent fuel. One option for managing these nuclides is the final disposition of SNF, with or without preliminary reprocessing, in repositories built in geologically stable formations. Currently there is no such operational facility in the world, the reasons for this being complex and not always of technical matter [5].

According to IAEA's assessments, world SNF stockpile, generated by commercial reactors will amount at 445 000 tonnes of heavy metal by 2020. It is anticipated that only about a quarter of this spent fuel will undergo some kind of reprocessing [6]. The annual quantity of SNF discharged from commercial reactors is about 10 500 tonnes of heavy metal [7] with average annual amount generated by a 1000 MW LWR of approximately 20-30 tonnes [8]. No matter if this spent fuel will be reprocessed or not, the final stage of any nuclear fuel cycle requires final repository for either SNF or vitrified high-level radioactive waste (HLW) disposal [9]. It is necessary to estimate the amount of SNF and/or HLW that would be reprocessed and/or disposed of in order to assess the reprocessing plants' capacity needed, and to determine the requirements intermediary and final SNF and HLW repositories should comply to. Nuclear power capacity's anticipated growth determines increase in SNF quantity, which in turn makes the solution for the management strategy even more urgent. On the other hand, around 96% of SNF components could be recycled because they're either fissile or fertile materials (Figure 2), so by using radiochemical reprocessing the amount

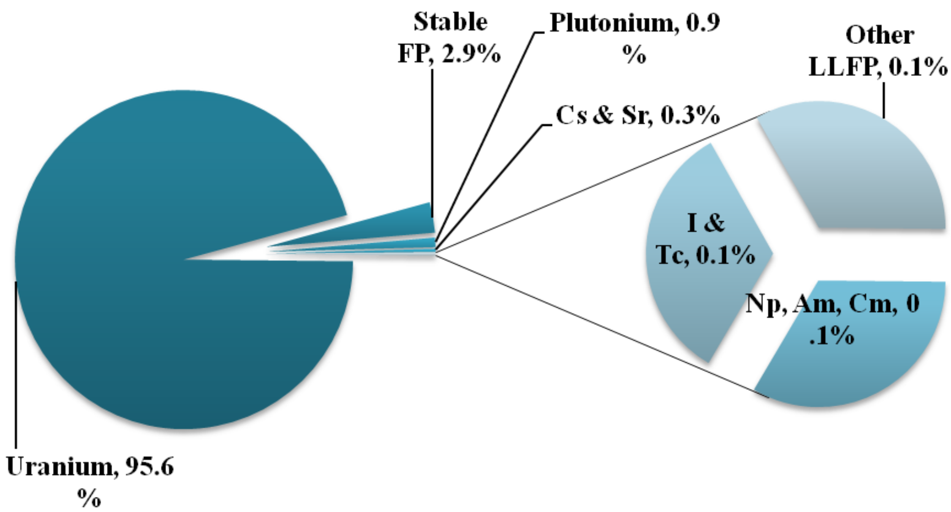

Figure 2. LWR SNF approximate composition, adapted from [9].

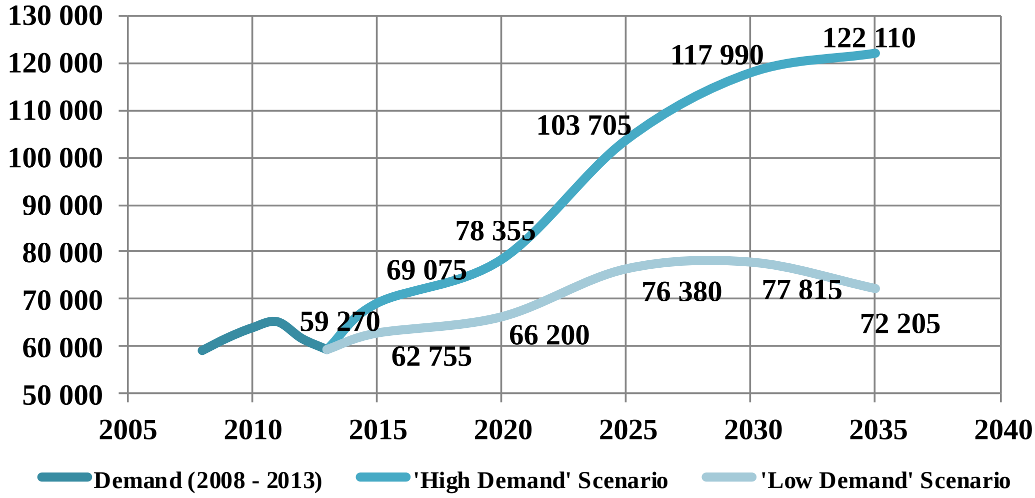

Figure 3. Natural Uranium Demand Outlook, tHM, [12-14].

of irradiated material destined for final disposition would be drastically reduced. In this way the usage of natural resources by nuclear power will become more efficient, and the volumes of irradiated material that should be disposed of will decrease as well [5].

4 Nuclear Power's Resource Base

On the other hand, nuclear energy's sustainable development depends on its resource base, i.e. the availability of energy resources that could be used as nuclear fuel. These materials could be ^{235}U (primary nuclear fuel) or other fissile isotopes that are not naturally occurring (secondary fuel), e.g. ^{233}U and ^{239}Pu among others [10]. In these terms the reactor fleet's growth would put strain to the production capabilities of the plants in different stages of the front end of the fuel cycle – mines, milling, conversion and enrichment plants, in spite of the increasing efficiency of nuclear fuel use (increased burn-up) [11]. Because of this reason it is necessary to estimate natural uranium consumption and uranium ore reserves needed to back the projected nuclear power generation growth. Widening nuclear power's resource base could be done not only by new mining development, but by closing the fuel cycle as well. Recycling the plutonium and reprocessing the uranium would not only diminish the natural uranium and separation work requirements, but would help in resolving the problems standing before SNF management – namely volume and activity reduction.

The reprocessed uranium use partially eliminates the need of mining uranium ore for natural uranium production, thus reducing the waste and tailings from mining and milling.Currently stockpiles of reprocessed uranium are small but it is possible that in the future the annual production could reach 4 000 tonnes of uranium metal with ^{235}U concentration between 0.4% and 0.8%. That's equivalent to between 5% and 10% of current world uranium demand and is equal to the annual production of a large mine [5]. On the other hand, natural uranium demand will grow even in low nuclear power development cases, only with slower rate (Figure 3).

5 Nuclear Fuel Cycles' Balances

Determining material and isotopic balances of the fuel cycle gives the opportunity to assess different fuel cycle plants' needed production capacities, and the availability of primary and secondary nuclear fuel. Defining the amount of generated plutonium is important not only for assessing nuclear materials' availability for fuel production, but is crucial in determining and managing proliferation risks.

The investments in the fuel cycle and SNF and RAW management, which participate in nuclear electricity total cost formation, are defined by the production capacities of the different stages of the fuel cycle, the needed storage capacity for different materials, e.g. uranium hexafluoride, depleted uranium, and SNF and RAW repositories needed capacity [15].

SNF's isotopic composition determines the activity of the material that would be temporarily stored or disposed of. The activity and the quantity of these materials define the requirements that storage facilities and repositories should meet [5].

6 Input Data

Fuel cycle's material balance and spent fuel isotopic compositions are calculated using IAEA's software product Nuclear Fuel Cycle Simulation System (VISTA) [16]. The calculations are made for once-through and partially closed fuel cycles for the three types of light water reactors – PWR, BWR, and WWER. In the partially closed fuel cycle simulation the fuel used is mixed uranium-plutonium oxide fuel (MOX) that's been fabricated by blending depleted uranium (^{235}U concentration – 0.3%) and reactor-

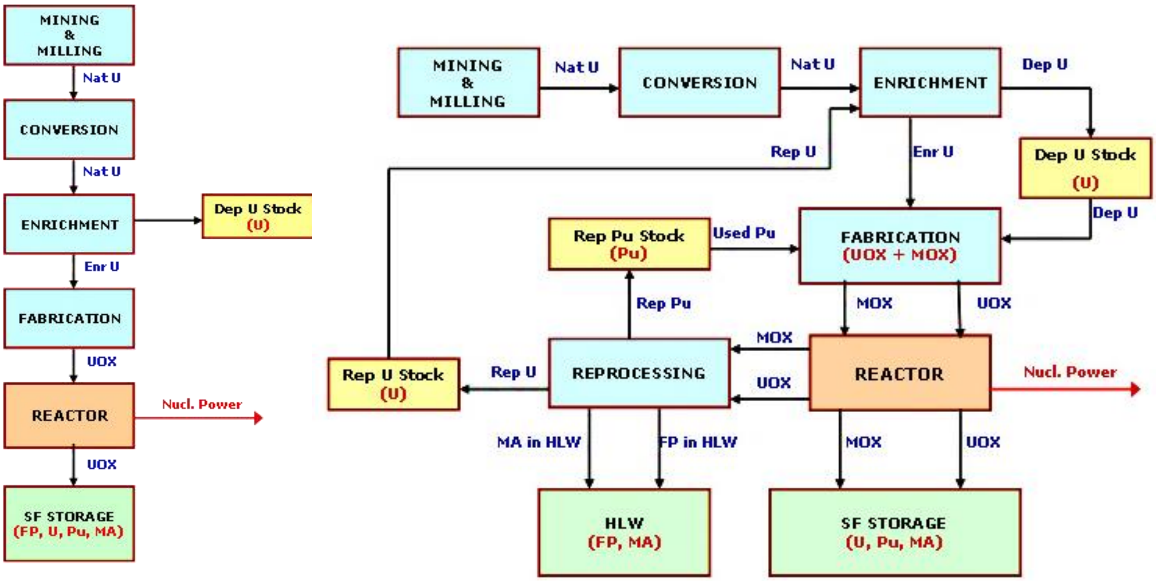

Figure 4. Fuel cycle diagrams, illustrating VISTA's material balance and flow calculation patterns – LWR once-through (left) and LWR closed cycle (right) [16].

grade plutonium (^{239}Pu concentration – around 60 w%). Two possibilities are considered – 5% and 30% share of the core loading is MOX.It is assumed that the plutonium used in the MOX fuel is extracted from LWR spent fuel, using PUREX process, and the SNF has been cooled down for 7 years before reprocessing. It is assumed that reprocessed uranium has not been used in the closed cycle.

Figure 4 representsa diagram of the fuel cycle models used to calculate the material balances. The main goal of this article is to preliminarily assess fuel cycles' material balances that would be used in later fuel cycle analyses. That's why the model input parameters assume the mean values that are typical for each power reactor type. These values are shown in Table 2. The designations are as follows: N – generating unit's gross installed capacity, MW; φ – capacity factor, %; η – unit's gross thermodynamic efficiency, %; y – tails assay from enrichment, %; c_5 – nuclear fuel (UO_2) enrichment, %; B – burn-up, GWd/tU; c_{Pu} – plutonium mass fraction in MOX-fuel ($(U;Pu)O_2$), %; B_{MOX} – MOX-fuel burn-up, GWd/tTM. Uranium concentration in the ore is assumed as 1%. The calculations are done for a time-frame of 1 year.

Table 2. Input data

	N MW	φ %	η %	y %	c_5 %	B GWd/t	c_{Pu} %	B_{MOX} GWd/t
PWR	1000	85	32.6	0.3	3.968	45	7.23	45
BWR	1000	85	32.6	0.3	3.968	45	7.23	45
WWER	1000	85	32.6	0.3	3.685	40	6.5	40

7 Results

The main results from the calculations for once-through and partially closed fuel cycles for the three types (PWR, BWR, and WWER) of LWR are summarized in the tables and charts below.

7.1 Once-through Fuel Cycle

From the results shown in Table 3 becomes evident that the WWER reactor type uses slightly more natural uranium (about 4% more) and requires a bit more (less than 1% however) separation work than its LWR counterparts. That means that the amounts of generated depleted uranium and discharged spent fuel are higher as well (about

Table 3. Once-through fuel cycle material flows

	WWER	PWR	BWR
Uranium ore, [t]	19 595.30	18 874.20	18 874.20
Natural Uranium, [t]	196.00	188.70	188.70
Separation work, [MTSWU]	111.40	110.30	110.30
Enriched Uranium requirements, [t]	23.80	21.10	21.10
Depleted Uranium to stock, [t]	172.20	167.60	167.60
Gross installed capacity, [MW]	1000.00	1000.00	1000.00
Generated electricity, [GWh]	7446.00	7446.00	7446.00
Discharged SNF, [t]	23.80	21.10	21.10

Table 4. SNF isotopic composition at discharge in once-through fuel cycle

	PWR		BWR		WWER	
	[t]	[w%]	[t]	[w%]	[t]	[w%]
Uranium-235	0.1412	0.668	0.1300	0.615	0.2321	0.976
Uranium-236	0.1101	0.521	0.1108	0.524	0.1079	0.453
Uranium-238	19.6811	93.061	19.6976	93.139	22.1773	93.212
Plutonium	0.2151	1.017	0.2080	0.983	0.2750	1.156
Plutonium-239	0.1082	0.512	0.1027	0.486	0.1562	0.656
Other Pu isotopes	0.1069	0.505	0.1053	0.498	0.1188	0.500
Minor Actinides	0.0056	0.027	0.0064	0.030	0.0065	0.027
Fission products	0.9811	4.639	0.9811	4.639	0.9810	4.123

2.8% and 12.8 % respectively). In terms of mass, the considered WWER annually discharges 2.7 tHM of spent fuel more than the other LWRs. That could be explained with the slightly lower initial enrichment and the lower burn-up achieved (see Table 2).

The considered WWER also produces more plutonium (about 60 kg more than PWR and 65 kg more than BWR). The obtained quantity of ^{239}Pu is also greater (about 50 kg more than in PWR); both in terms of mass and as a relative share of the plutonium mixture (see Table 4). Fission products' mass is generally equal in all types of LWR but their relative share is lower in WWER because of the lower burn-up resulting in higher heavy metal requirements. Regarding ^{235}U contents in SNF, it is about 1.7–1.8 times greater in WWER's spent fuel, again as a consequence of the lower burn-up. This results in more inefficient use of primary energy resources, but on the other hand the production of secondary nuclear fuel (namely ^{239}Pu) is also higher in WWER.

7.2 Closed Nuclear Fuel Cycle

Table 5. Closed fuel cycle material flows

	5% MOX	30% MOX
Uranium ore, [t]	17930.50	13212.00
Natural Uranium, [t]	179.30	132.10
Separation work, [MTSWU]	104.80	77.20
Enriched Uranium, [t]	20.10	14.80
Depleted Uranium to stock, [t]	159.20	117.30
Nuclear fuel requirements, [t]	21.10	21.10
UOX, [t]	20.10	14.80
MOX, [t]	1.10	6.30
Used reprocessed Pu, [t]	0.08	0.46
Used depleted Uranium, [t]	1.00	5.90
Plutonium balance, [t]	**-0.01**	**-0.41**

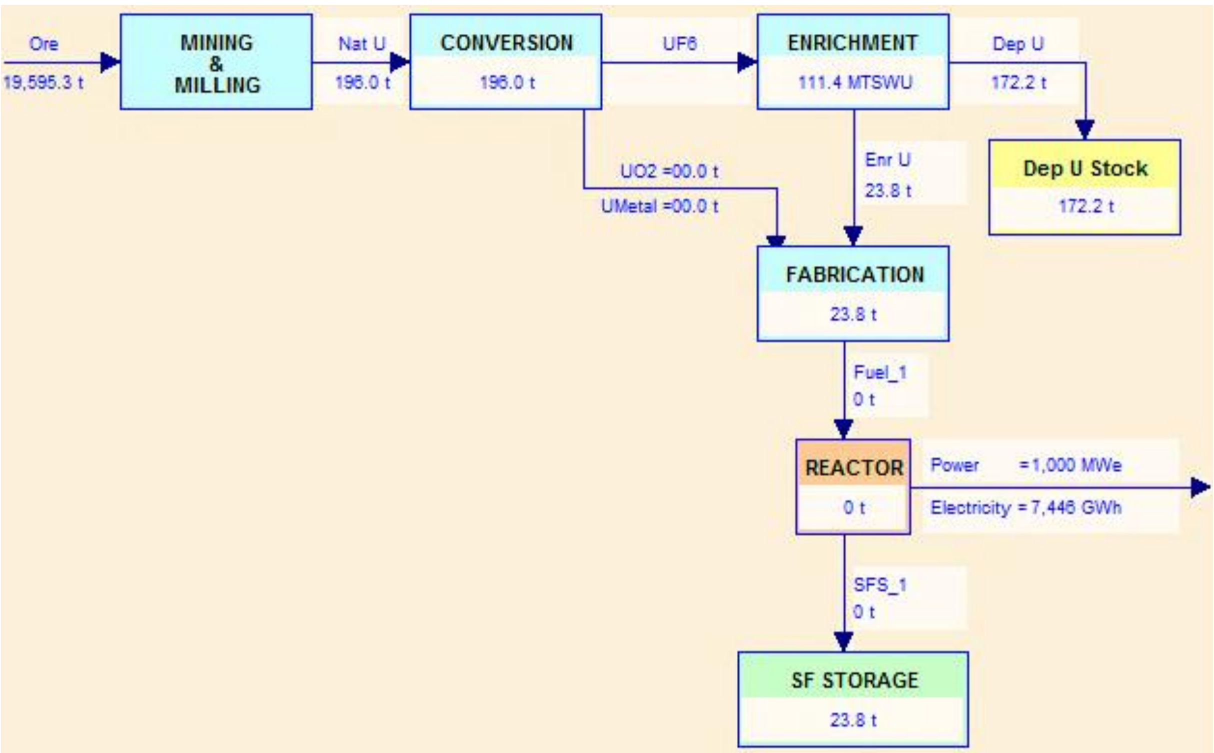

Figure 5. WWER once-through cycle material flows.

Table 6. Different isotopes' masses in discharged SHF in closed fuel cycle

	PWR		BWR		WWER	
	MOX 5%	MOX 30%	MOX 5%	MOX 30%	MOX 5%	MOX 30%
Uranium-235, [t]	0.0913	0.0748	0.0841	0.0690	0.1495	0.1195
Uranium-236, [t]	0.0704	0.0534	0.0708	0.0537	0.0690	0.0525
Uranium-238, [t]	13.4815	14.9572	13.4973	14.9969	15.1955	16.8790
Plutonium, [t]	0.1853	0.3912	0.1757	0.3575	0.2310	0.4646
Plutonium-239, [t]	*0.0862*	*0.1549*	*0.0779*	*0.1231*	*0.1235*	*0.2176*
Other Pu isotopes, [t]	*0.0991*	*0.2363*	*0.0978*	*0.2344*	*0.1075*	*0.2470*
Minor Actinides, [t]	0.0166	0.0322	0.0171	0.0317	0.0149	0.0261
Fission Products, [t]	0.6735	0.7545	0.6735	0.7545	0.6735	0.7544

Table 7. Different isotopes' relative shares in discharged SNF in closed fuel cycle

	PWR		BWR		WWER	
	MOX 5%	MOX 30%	MOX 5%	MOX 30%	MOX 5%	MOX 30%
Uranium-235, [w%]	0.629	0.460	0.579	0.424	0.915	0.653
Uranium-236, [w%]	0.485	0.328	0.488	0.330	0.422	0.287
Uranium-238, [w%]	92.857	91.969	92.966	92.213	93.034	92.254
Plutonium, [w%]	1.276	2.406	1.210	2.198	1.414	2.540
Plutonium-239, [w%]	*0.594*	*0.952*	*0.536*	*0.757*	*0.756*	*1.190*
Other Pu isotopes, [w%]	*0.682*	*1.453*	*0.674*	*1.441*	*0.658*	*1.350*
Minor Actinides, [w%]	0.114	0.198	0.118	0.195	0.091	0.143
Fission products, [w%]	4.639	4.639	4.639	4.639	4.123	4.123

When calculating the material flows in partially closed fuel cycle with MOX fuel, the output flows are the same for the three types of light water reactors. That's why table 5 represents the material flows only as a function of the core fraction loaded with MOX.

Tables 6 and 7 summarize the SNF isotopic composition at discharge for each reactor type. The values are obtained by summing up the content of each isotope in the uranium fuel and in the mixed oxide fuel because the aim is to determine the total annual production rate of each nuclide.

Increased MOX-fuel share in core loading leads decreased ^{235}U and ^{236}U concentrations in the discharged fuel because of the partial replacement of primary nuclear fuel with secondary fuel, mostly ^{239}Pu. There is an increase in ^{238}U content that can be explained by the fact that MOX fuel is a mixture of plutonium and depleted uranium, with

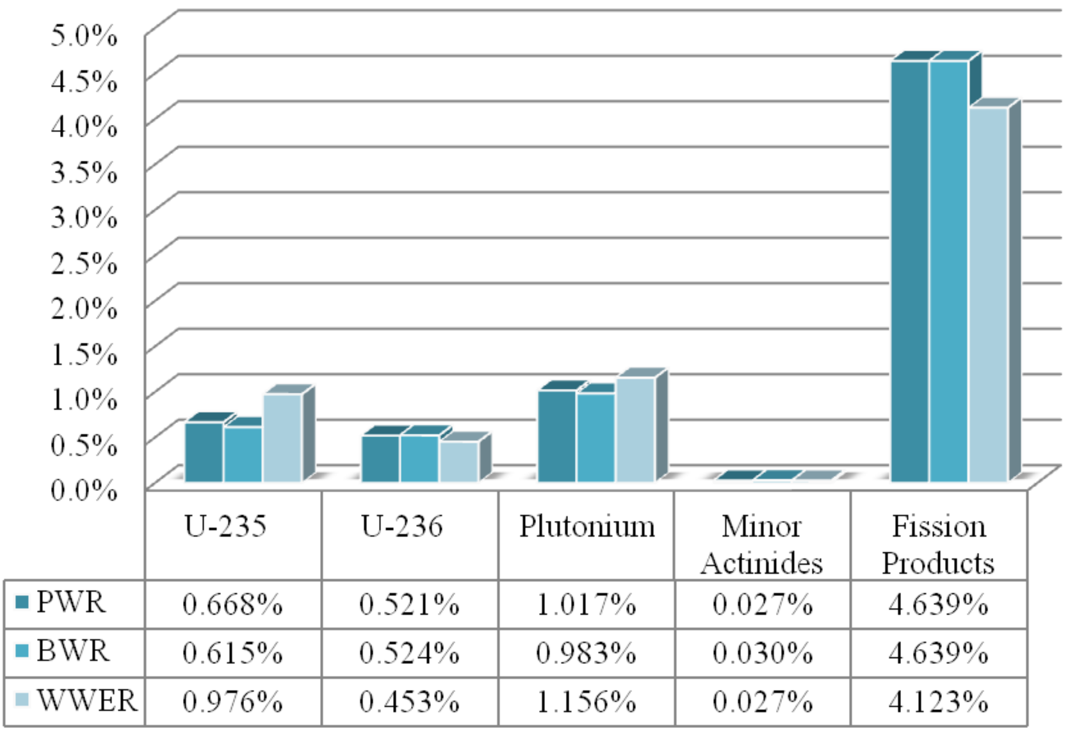

Figure 6. Relative shares of spent uranium fuel components at discharge in once-through cycle.

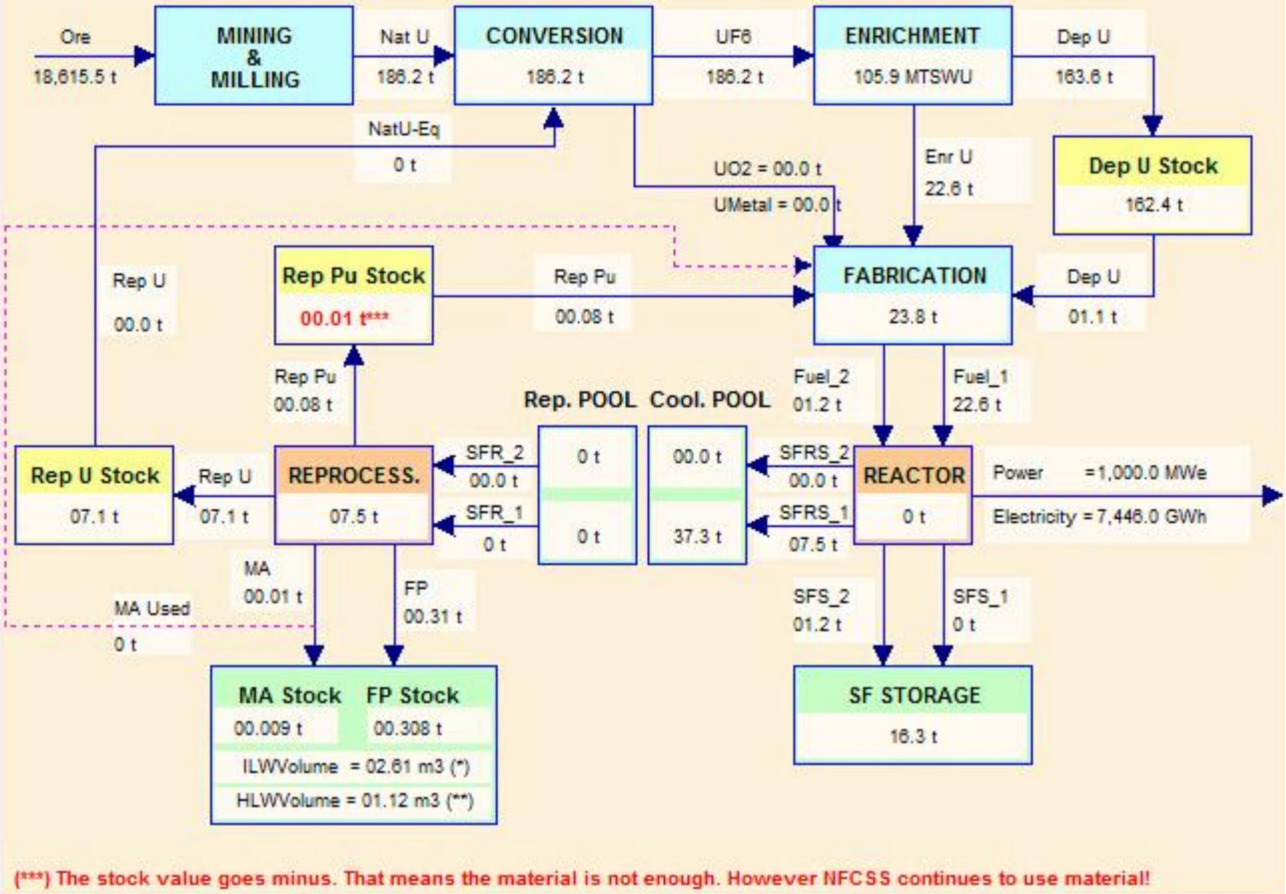

Figure 7. WWER partially closed fuel cycle material flows.

about 93% depleted uranium share.

7.3 Savings in Partially Closed Fuel Cycle

Table 8 summarizes the potential savings that could be achieved by applying partially closed nuclear fuel cycle. When 5% of the WWER core is loaded with MOX the achieved savings of uranium ore and natural uranium are around 8.5%, and the amount of depleted uranium from enrichment is reduced by 8%. If 30% of the core are loaded with MOX, the savings are about 33%. In the case of 5% MOX loading of PWR/BWR core, the savings of ore and natural uranium are around 5%, and increase to 30% in the case of 30% MOX core loading.

8 Conclusion

From the results presented above, we could draw the following conclusions:

1. Recycling even a small fraction of the spent fuel could significantly decrease uranium ore mining and leads to natural uranium and separation work savings, and decreases the expenditures for conversion;
2. WWER-1000's fuel cycle offers the greatest potential for savings – its material requirements are the greatest amongst light water reactors, and the relative share of ^{239}Pu in the spent fuel is the highest;
3. From non-proliferation view point plutonium recycling in PWR and BWR is more efficient because ^{239}Pu relative share decreases nearly by a factor of two;
4. The relatively higher content of fissile materials in WWER's spent fuel make this type of power reactors suitable for use in fuel cycles comprising plutonium recycling in fast breeder reactors (FBR);
5. MOX use contributes to decreasing plutonium and

Table 8. Savings achieved by using MOX fuel

	WWER		PWR/BWR	
	5% MOX	30% MOX	5% MOX	30% MOX
Uranium ore, [t]	1664.80	6383.30	943.70	5662.20
Natural Uranium, [t]	16.70	63.90	9.40	56.60
Separation work, [MTSWU]	6.60	34.20	5.50	33.10
Enriched Uranium, [t]	3.70	9.00	1.00	6.30
Unproduced depleted Uranium, [t]	13.00	54.90	8.40	50.30

depleted uranium stockpiles. At 5% core loading with MOX, and reprocessing 1/3 of uranium spent fuel, virtually no additional plutonium is generated. At 30% core loading with MOX, the plutonium stockpile is reduced at a rate of 410 kg per reactor-year;

6. Increased plutonium and minor actinides content in spent MOX fuel complicates its management;
7. Spent MOX fuel could be reprocessed in an appropriate moment in the future when advanced commercially applicable process is available in order to recycle the plutonium, and transmute the minor actinides.

References

[1] IAEA: Power Reactor Information System (PRIS), http://www.iaea.org/PRIS/

[2] OECD IEA: World Energy Outlook 2014, OECD, Paris, 2014

[3] British Petroleum: Energy Outlook 2035, 2014

[4] IAEA: Climate Change and Nuclear Power 2014, Vienna, 2014

[5] К. Филипов, И. Найденов: Намаляване на количеството радиоактивни отпадъци чрез прилагане на затворени ядрени горивни цикли, Сборник доклади XIX научна конференция с международно участие ЕМФ 2014, том I, стр. 112-118, Созопол, 2014 (in Bulgarian)

[6] IAEA: Costing of Spent Nuclear Fuel Storage, NF-T-3.5, Vienna, 2009

[7] International Panel on Fissile Materials: Spent Fuel from Nuclear Power Reactors, 2011

[8] IAEA: Spent Fuel Reprocessing Options, IAEA-TECDOC-1587, Vienna, 2008

[9] OECD NEA: Advanced Nuclear Fuel Cycles and Radioactive Waste Management, Paris, 2006

[10] В. Велев, К. Филипов: Ядрени горива, ИфоДизайн, С., 2008 (in Bulgarian)

[11] Nuclear Fuel Cycle Science and Engineering, edited by I. Crossland, Woodhead Publishing, Cambridge, 2012

[12] OECD NEA, Uranium 2009: Resources, Production and Demand, Paris, 2010

[13] OECD NEA, Uranium 2011: Resources, Production and Demand, Paris, 2012

[14] OECD NEA, Uranium 2014: Resources, Production and Demand, Paris, 2014

[15] OECD NEA: The Economics of the Back End of the Nuclear Fuel Cycle, Paris, 2013

[16] IAEA: Nuclear Fuel Cycle Simulation System (VISTA), IAEA-TECDOC-1535, Vienna, 2007