Disposing High-level Transuranic Waste in Subcritical Reactors

Yaosong Shen

Institute of Applied Physics and Computational Mathematics, 6 Huayuan Road, 100088, Beijing, China

We propose a new method of burning high-level transuranic (TRU) waste combinedi with thorium-uranium fuel cycle in the subcritical reactors driven by the external fusion neutron sources. The thorium-based TRU fuel burns all the long-lived actinides by hard neutron spectrum while outputting power. The concept of the corresponding one dimension model is built by means of the ONESN_BURN code with new data libraries. The thorium-uranium fuel cycle produces less TRU, less radiotoxicity and fewer long-lived actinides. The thorium-uranium fuel cycle provides breeding of 233U with a long operation time (>20 years), hence significantly reducing the reactivity swing while improving safety and burnup. A detailed analysis is given in the paper.

Keywords: High-level Transuranic Waste; Thorium-Uranium Fuel Cycle; Thorium Base TRU Fuel; Actinide Burning.

1. Introduction

The nuclear electric capacity nearly 340 gigawatts-electric (GWe) produces spent fuel roughly amounting to 8000 tonnes heavy metal (tHM) per year in the world. Among various reactors, the light water reactor (LWR) is most widely deployed reactor. A modern LWR of 1 GWe capacity will typically discharge about 20-25 tonnes of irradiated spent fuel per year of operation. The spent fuel is comprised of 93-94% of the mass of uranium oxide (mostly U-238, only 0.8% U-235), about 3-5% fission products, ~1% plutonium (Pu) and about 0.1-0.2% the minor actinides (MA, including neptunium (Np), americium (Am) and curium (Cm)). The elements above uranium in the periodic table are known collectively as the transuranic (TRU). Although there are a number of fission product radionuclides of high activity (Cs-137 and Sr-90) and long half-life (Tc-99, 2.1124E+5 years; and I-129, 1.5711E+7 years) in spent nuclear fuel, actinides and their daughter products account for most of the radiotoxicity of nuclear waste after the first 100 years of disposal and most of radiotoxicity of high-level waste (HLW) is come from actinides of TRU especial to Pu (see figure 1).

There have been two types of fuel cycles: (1) open, with direct disposal of spent fuel and (2) closed, with the processing of spent fuel to recover and use the fissile and fertile fuel that remains. Traditional closed fuel cycles recycle uranium and plutonium. MA is treated as wastes. But the recently proposed technology termed as "actinide burning" [1], irradiates actinides chemically separated from spent fuel to convert them to short-lived radionuclides or non-radioactive elements. With actinide burning, all of the actinides are to be destroyed.

A fuel cycle that burns all actinides is significantly different from a traditional fuel cycle. It is these differences that lead to consideration of different reactors and fuel cycles for this mission. For example, Hyland et al., proposed MA burning in LWR or CANDU [2-3]; Forsberg et al., proposed molten salt reactors (MSRs, liquid-fueled reactors) can be used for burning actinides [4]; Hejzlar et al., proposed actinide burning in a lead-bismuth-cooled critical fast reactor [5]; Sandia National Laboratories designed a Z-pinch fusion driver to transmutation of TRU [6].

In this paper, we will propose a new concept of actinide burning with thorium-uranium (Th-U) cycle in the external fusion neutron source. Such a subcritical reactor (SR) can be driven by various external neutron sources, such as ITER, Z-Pinch, ICF or even ADS. It has a neutron spectrum harder than that of the other reactors (fast or thermal reactor); such a hard neutron spectrum is generally more effective in the fission of TRU. The fuel of SR is Th-TRU (thorium based TRU fuel). The advantages of Th-U cycle are (1) less TRU, less radiotoxicity and less long-lived actinide (see figure 2), (2) Naturally, there is only one isotope of Th-232 with stable chemical character, thorium was selected as the major fertile material to develop the metallic thorium-based TRU fuel; (3) Th-U cycle provides breeding U-233 within a long operation time (>20 years), hence significantly reducing the reactivity swing and improving safety; (4) reach a deep

burnup, (5) thorium is cheaper. The coolant of SR is liquid lead (PB) and structural material fractions of SR is austenitic stainless steel SS316 as same as that of LFR (lead cooled fast reactor).

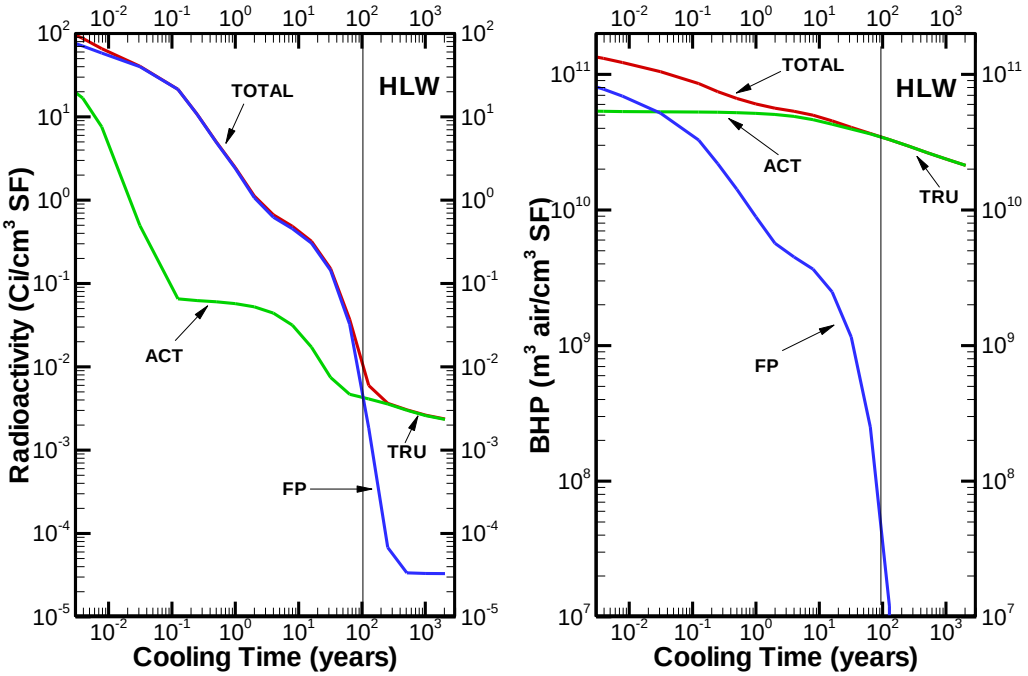

Figure 1 Radiotoxicity of the spent fuel from a modern LWR with 1GWe of 18 month operation

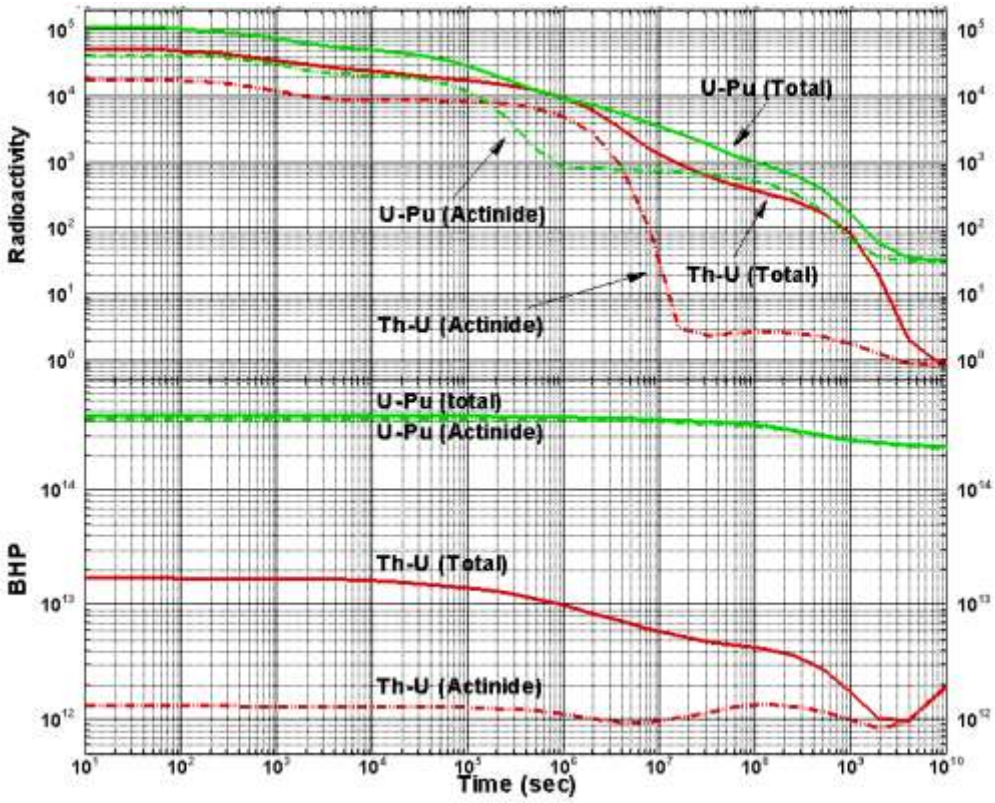

Figure 2 Radioactivity and BHP value of uranium-plutonium and thorium-uranium fuel cycle.

All calculations in the paper are performed using the One-dimension Sn neutron-Photon Transport and Burn-up calculation code (OneSn_Burn) developed at Institute of Applied Physics and Computational Mathematics (IAPCM). OneSn_Burn has several data libraries (1) 172 group neutron cross section for 501 nuclei; (2) 32 group photon cross section for all 1-100 elements; (3) 3468 isotopes of decay library; (4) 33 actinides fission product yields library; (5) misc data library for Biological Hazard Potential (BHP) calculation and material neutron and photon radiation damage (DPA) calculation.

2. Calculation and Analysis

Table 1 shows the TRU properties of regular LWR spent fuel; the atomic fraction of TRU is taken from the Ref.[6]. Here, Pu is more than 80%, Am is about 11%, Np is about 7% and Cm is less than 1%. Firstly, we calculate the effectiveness of burning by different neutron spectrum. Figure 3 gives the neutron spectrum, the blue line is the typical pressure water reactor (PWR) represented by thermal reactor and the red one is what we use, fusion fission subcritical reactor (FFSR) driven by 14.1MeV external fusion neutron source. It is very clear that the PWR spectrum reaches peak at 0.01-0.1eV, The FFSR spectrum seems to be more complicated and it has several peaks at 14.1MeV, fission energy zone and 10-100eV, respectively. The FFSR spectrum is similar to critical fast reactor LFR except at 14.1MeV.

Table 1 TRU properties of LWR spent fuel. The data of atomic fraction is taken from Ref.[6].

Actinide Isotope	Atomic fraction	Half life (y)	Decay mode
Np-236	7.3147E-08	1.5310E+05	β
Np-237	7.3435E-02	2.1454E+06	α
Np-239	1.7966E-08	6.4547E-03	β
Pu-238	2.6630E-02	8.7760E+01	α
Pu-239	4.5452E-01	2.4127E+04	α
Pu-240	2.6288E-01	6.5656E+03	α
Pu-241	9.2059E-03	1.4299E+01	β
Pu-242	5.7352E-02	3.7376E+05	α
Pu-244	1.1055E-05	8.1156E+07	α
Am-241	9.2075E-02	4.3290E+02	α
Am-242	1.9452E-09	1.8287E-03	β
Am-242m	1.6305E-04	1.4109E+02	γ
Am-243	2.0530E-02	7.3749E+03	α
Cm-242	3.9434E-07	4.4641E-01	α
Cm-243	2.7573E-05	2.9120E+01	α
Cm-244	2.1100E-03	1.8123E+01	α
Cm-245	8.6423E-04	8.5057E+03	α
Cm-246	1.9336E-04	4.7633E+03	α
Cm-247	3.7225E-06	1.5610E+07	α
Cm-248	4.2804E-07	3.4824E+05	α

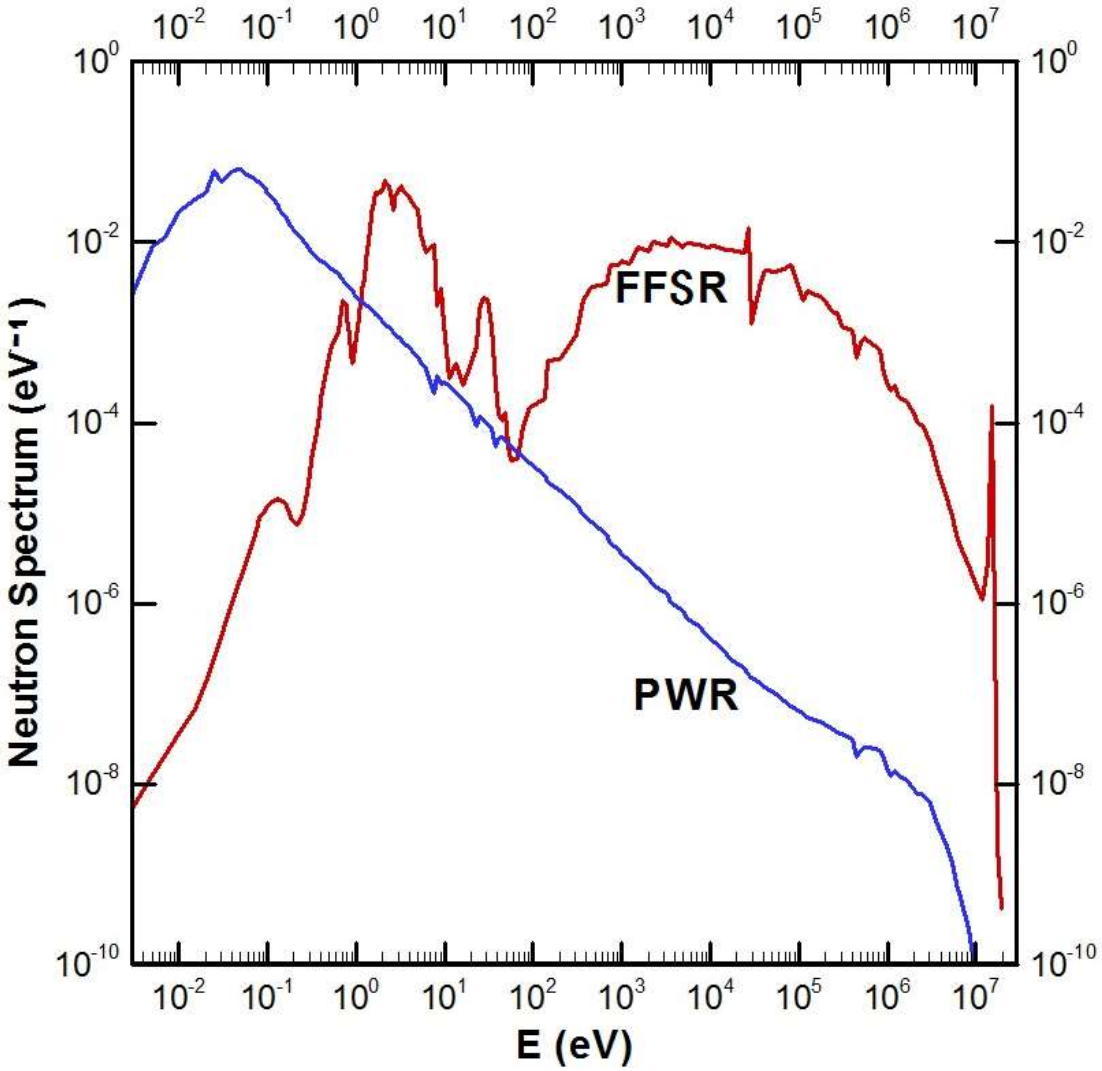

Figure 3 The neutron spectrum, the blue line is the PWR spectrum and the red line is FFSR spectrum. The PWR spectrum is calculated by PWR fuel and the FFSR spectrum is calculated by TRU fuel with 14.1MeV neutron source. All calculation is done by OneSn_Burn code.

Figure 4 shows the actinide radioactivity of TRU fuel changes by different neutron spectrum (PWR, FFSR and single energy 14.1Mev Spectrum) with a constant neutron flux (1.0E+14 n/s/cm3, regular neutron flux in fuel zone at the thermal reactor). Under 18 month operation (PWR typical operation time) at the constant neutron flux, the radioactivity shows the different change. The original TRU result (nothing to do with TRU fuel) is given for comparison. When the beginning of burning, the radioactivity will increase rapidly because of production of many short-lived actinides from neutron reaction, such as (n, γ), (n,xn) and (n,p/t/α). Harder the neutron spectrum, less short-lived actinides. During the 18 month burning, the radioactivity keeps stable and changes very small. After shutdown, the radioactivity quickly decreases because of short-lived actinides decay to stable nuclides. It is very obviously that FFSR and single energy 14.1MeV lines will decrease below the original line after several years of cooling, but PWR line cannot below to the original line until to 500 years of cooling almost. The reason is that the thermal neutron spectrum in PWR is very soft and is less suitable for burning TRUs as this causes a build-up of TRU isotopes with low fission probability and large (n, γ) cross section of Pu-239 and Pu-240, it brings to lots of Pu-241 (14.3 years half life, see Table 1). The number densities of Pu-241 after 18 month operation at PWR, FFSR and single energy 14.1MeV spectrum are 4.0682E+21/cm3, 2.3570E+20/cm3 and 2.2966E+20/cm3, respectively. The fatal disadvantage of actinide burning at a thermal reactor is that it needs a long cooling time (>100 years) after the reactor shutdown because of Pu-241.

The cyan line in figure 4 shows the actinide radioactivity of thorium (Th-232) by FFSR spectrum. Th-232 is a stable element and no radioactivity if no neutron irradiation. After one (n, γ) reaction and two β decay, Th-232 brings to the easy fissile element U-233 that similar to U-235 and this fuel cycle called Th-U cycle. The actinide radioactivity of Th-U is much less than the regular U-Pu cycle in thermal reactor. As shown at left of figure 4, the actinide radioactivity of Th-232 is closer to that of TRU fuel because of the short-lived actinide at the beginning of the burning, but after reactor shutdown, the actinide radioactivity of Th-232 deceases very rapidly during 1-6 month compare to TRU fuel. Th-U cycle can not bring to TRU element and the long-term actinide radioactivity mainly comes from Pa-231 (half life 4.73E+04 years) and its decay daughter nuclide. In fact, the actinide radioactivity of Th-232 is only about 0.01-0.1% of the TRU at same mass after 6 month cooling. So we ignore the actinide radioactivity of Th-232 at following calculation.

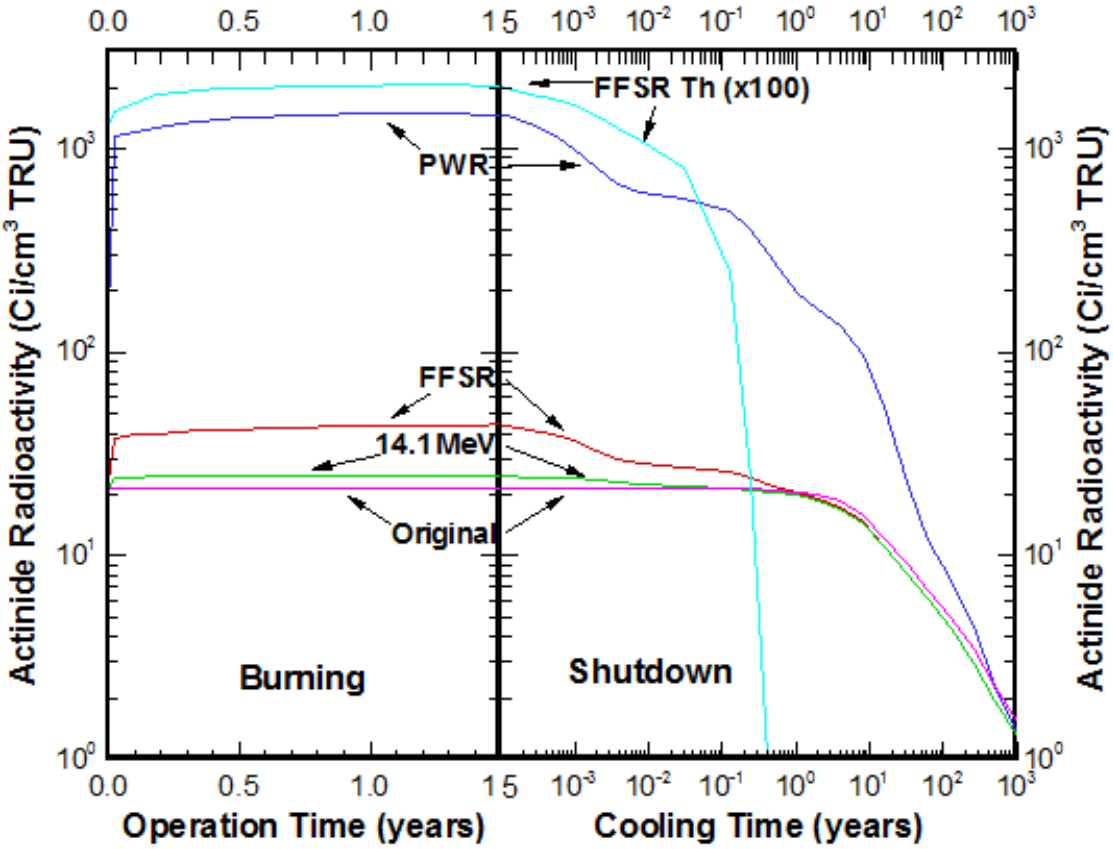

Figure 4 Actinide radioactivity of TRU fuel during burning and shutdown. Left figure is burning with a constant neutron flux (1.0E+14 n/s/cm3) for 1.5 years, and then, shutdown (no neutron flux) for cooling at right figure. The blue, red and green lines denote the results calculated by different neutron spectrum of PWR, FFSR and single energy 14.1MeV, respectively. The purple line is the original value line with TRU unirradiated (no neutron flux). The cyan line which multiply a factor of 100 is Th-232 result by FFSR spectrum (the original value of Th-232 is almost zero).

We propose a concept of TRU burning with thorium-uranium (Th-U) cycle by FFSR with k-effective (Keff) 0.8-0.85. The thorium based TRU fuel is designed closer to 14.1MeV fusion source (core) and pure thorium fuel surrounding with liquid lead coolant. The neutron flux at FFSR will reach to 1.0-3.0E+15 n/s/cm^3 with 200MW 14.1MeV fusion neutron source. When FFSR operation, the fission reactivity of TRU will decrease and the fission reactivity of Th-U cycle (from U-233) will increase slowly to compensate it and keep the total power of the reactor (3000MWe heat) to balance for more than 30 years. In the calculation, we adjust a little of the source strength to keep the total power of reactor at 3000MWe heat with about 10 tons of TRU fuel.

Figures 5-7 shows the numerical results of TRU burning by FFSR. It is clearly that the different TRU element contributes to the different cooling time. Figure 5 shows the actinide radioactivity, before 50 years, the primary

radionuclides of TRU will be Pu-238, Cm-244 and Pu-241 and the contribution of Am-241 decreases obviously after 30 years burning; Cm-244 and Pu-241 will completely decay after 100 years. The long-term actinide radioactivity after 100 years has a significant decrease because all the long-lived TRU elements, such as Pu-239, Am-241, Pu-240 and Am-243 (Np-239 is the decay daughter nuclide of Am-241 with a short half life of 2.356 days), are more or less reduced.

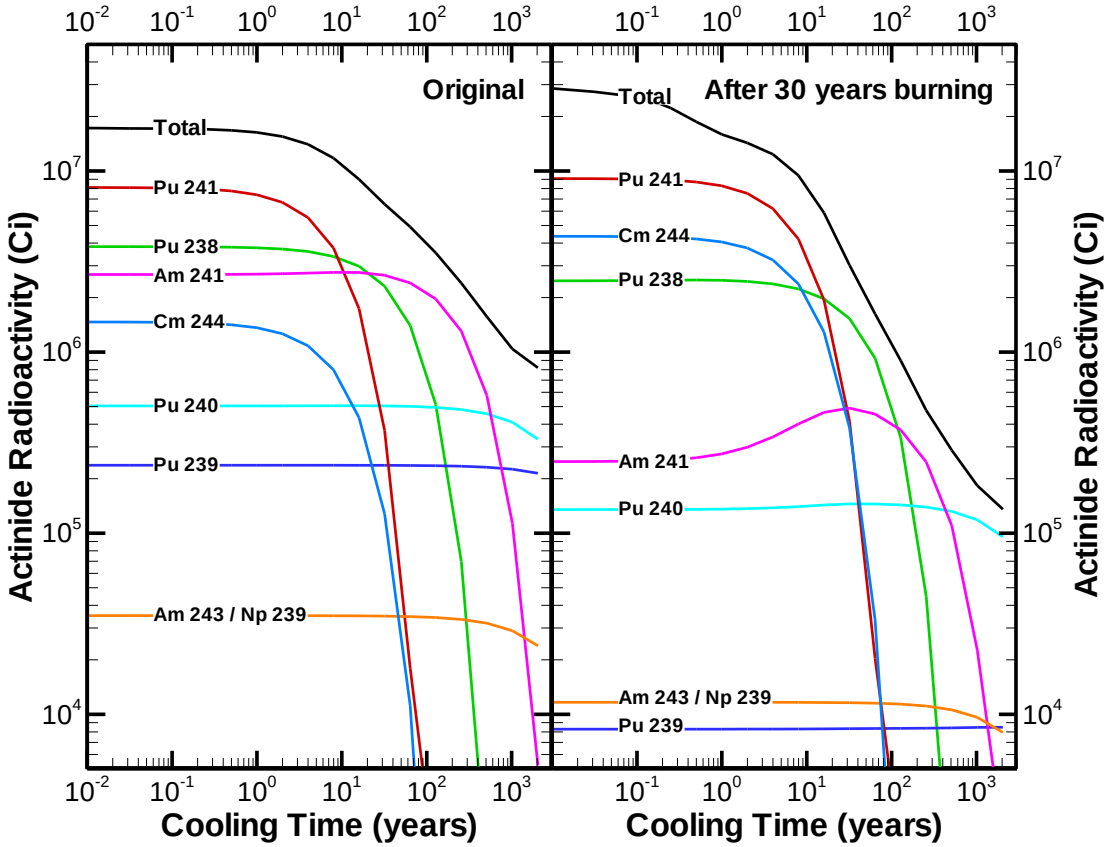

Figure 5 Actinide radioactivity of TRU fuel vs. cooling time, left figure is the original TRU fuel and right figure is TRU fuel after 30 years burning at FFSR with the constant power 3000Mwe heat.

Since the radioactivity in above figures does not include a measure of the radiotoxicity of a radionuclide, a quantity called "biological hazard potential" has been introduced (BHP, unit m^3 of air contaminated by entire reactor). The metabolism of radionuclides in the so-called reference man has been formulated in models based on which the maximum permissible concentrations (MPC) of radionuclides in air (or water) for occupationally exposed workers have been calculated. The definition of the potential biological hazard (BHP) is

$$BHP = \sum BHP_i = \sum_i Q_i / MPC_i$$

here, Qi, MPCi and BHPi are the ith radionuclide's radioactivity (unit is curie Ci), maximum permissible concentrations (unit is Ci/m^3), and biological hazard potential (unit m^3 of air), respectively. Figure 6 shows the BHP of TRU fuel, before 100 years, the primary radionuclides of TRU will only be Pu-238; after 500 years, the primary radionuclides of TRU will only be Pu-240 after 30 years burning. Here the BHP of Am-241 and Pu-239 decrease significantly and Pu-240 and Am-243 decrease a little. Because the actinide elements contribute to more than 90% of total long-term BHP, the key of BHP reduction is decreasing the density of Pu-238 and Pu-240.

Figure 7 shows the density of some long-term TRU elements changes by the 30 years operation time. It is clearly that Pu-239, Np-237 and Am-241 drop down quickly, Pu-240 and Pu242 decrease a little slower than Am-241. Pu238 and

Pu241 increase at first 3-5 years and decrease slowly, reach to the original level at the end of 30 years. Cm-242 increases at first 2 year rapidly and decrease to original level near 30 years. Only Cm-244 has a significant increase at the end of 30 years because Am-241 has a (n,γ) reaction to Am-244 (half life 14.57 hours), which decay to Cm-244 quickly. The amounts of density of TRU will decrease to 17% of the original value after 30 years burning by FFSR and the reactor has a capacity of burning about 280kg TRU per year and output 3000Mwe heat.

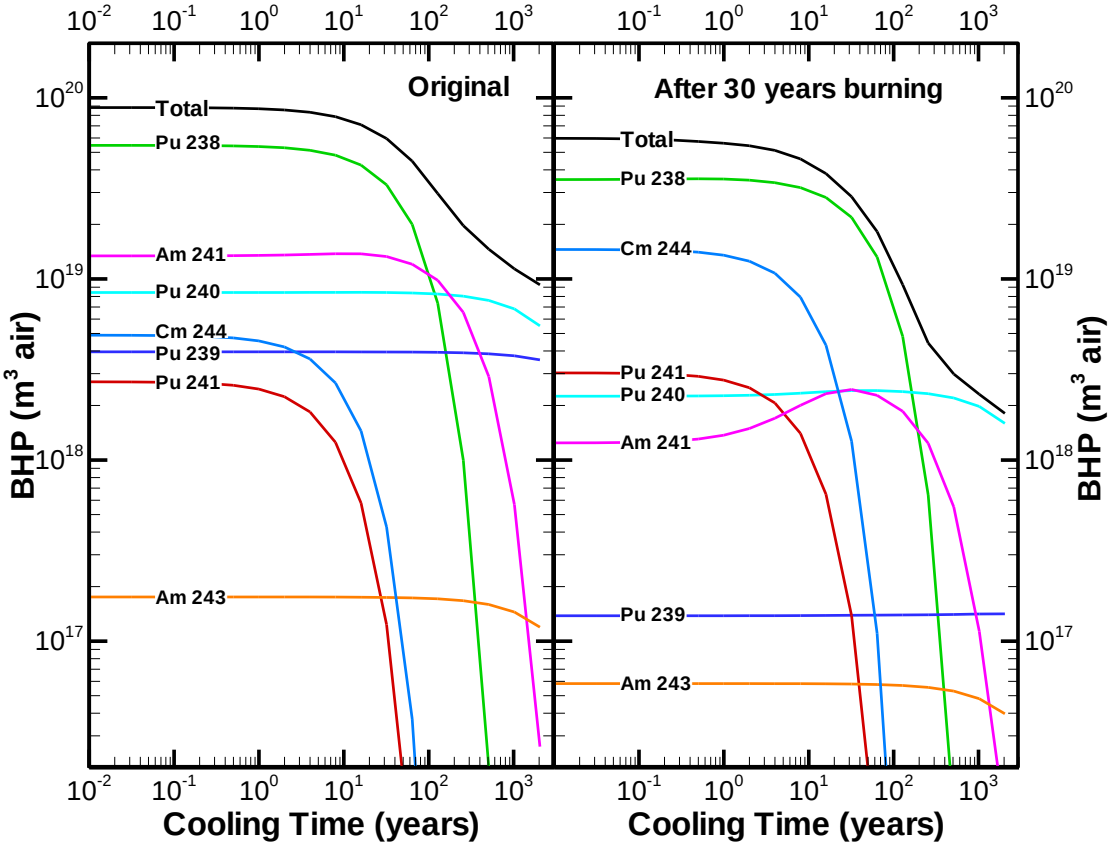

Figure 6 Same for figure 5 but for BHP.

3. Conclusion

Above 3-5Mev, (n,γ) cross section of most TRU elements is more smaller than that of (n,f) (n,xn) (n,p/t/α) cross section, Above 0.1MeV, (n,γ) cross section of most TRU elements is more smaller than that of fission (n,f) cross section. But below 10eV, (n,γ) reaction is dominant and it is the only reaction of some TRU elements. The key of 'Actinide Burning' is TRU fission, so the essential condition is the hard neutron spectrum. Such a hard spectrum, comes from fusion, accelerator or fast reactor, seems to more efficient than the soft one. Actinide burning needs a very long time (>30 years) of reactor operation, Th-U cyele can extend the operation time and get deep burnup, so actinide burning withTh-U cycle in the external fusion neutron source is the better way which both burning TRU and output energy.

In the calculation, FFSR is very good to burning Np-237, Pu-239 and Am-241, a little burning to Pu-240, but fail to Pu-238, Pu-241 and Cm-244. Because about 90% of TRU fuel mass consist of Pu-239, Pu-240, Am-241 and Np-237, FFSR seems to successful to actinide burning. Pu-238 seems to very important to BHP (relative to human hazard) until to 100 cooling years and how to reduce Pu-238 is worth to further study.

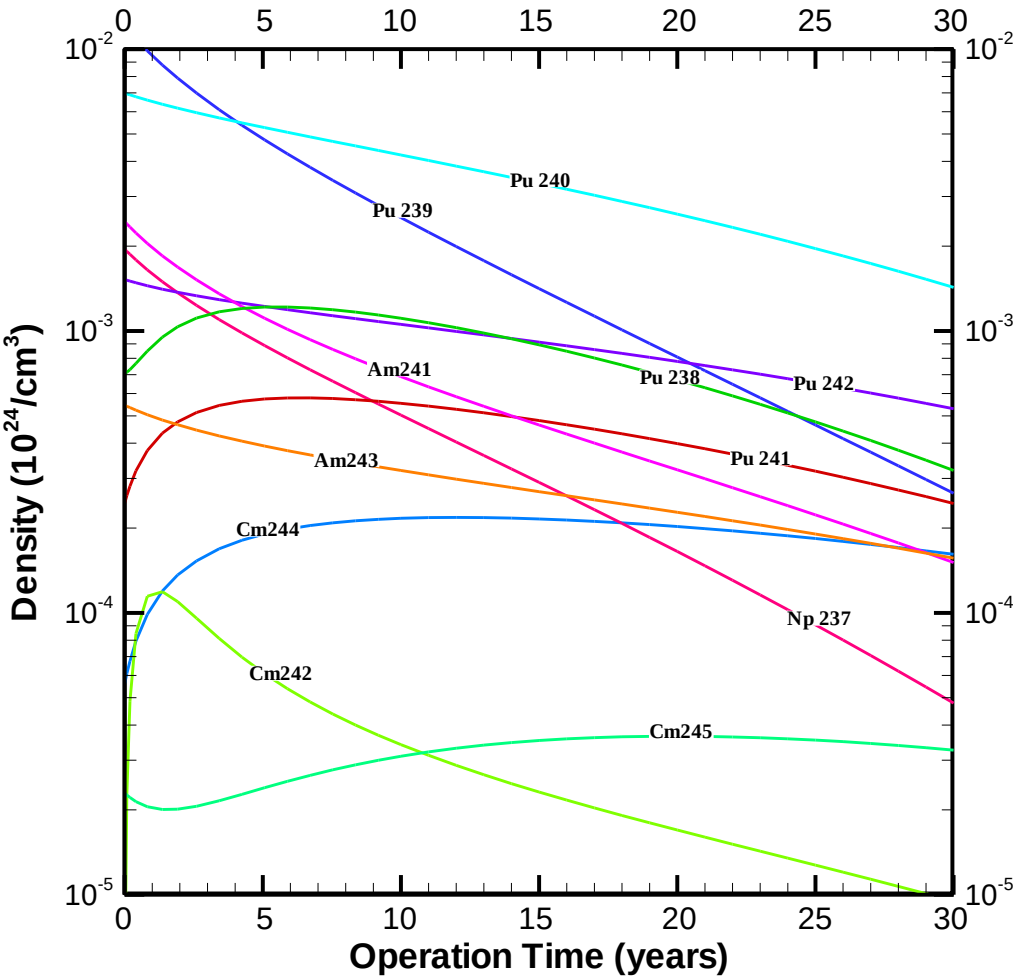

Figure 7 Density of some long term TRU elements vs. the operation time.

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