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Comparative Analysis of ENDF, JEF & JENDL Data Libraries by Modeling the Fusion-Fission Hybrid System for Minor Actinide Incineration

D. RIDIKAS[1)*], A. PLUKIS[2)], R. PLUKIENE[2)]

1) DSM/DAPNIA/SPhN, CEA Saclay, F-91191 Gif-sur-Yvette, France
2) Institute of Physics, Savanoriu pr. 231, LT-2053 Vilnius, Lithuania

A possibility to use fusion-fission molten salt (flibe) hybrid system to incinerate nuclear waste minor actinides (Np, Am, Cm) is considered. An optimization procedure was developed in order to determine the fission blanket composition corresponding to fast incineration rate of actinides. Performance parameters such as k_{src}, burn-up, neutron fluxes and equilibrium conditions were obtained. Monteburns code system (MCNP+ORIGEN) was used for system modeling. Approximately 1.1 tons of minor actinides, originating from the spent nuclear fuel, could be incinerated annually with an output of 3 GW_{th} fission power. Detailed burn-up calculations show that the equilibrium conditions of the fuel concentration in the flibe transmutation blanket could be achieved, if the originating tritium and fission products were removed at least in part and fresh minor actinides was added during the burn-up on line.

The optimal transmutation blanket in equilibrium is dominated by minor actinides for which the cross sections may vary depending on the nuclear data libraries used. The system performance was tested comparing different sets of data libraries. A number of major differences among ENDF, JENDL and JEF data files were identified and quantified in terms of the averaged one-group cross sections. Our study could be directly generalized for other transmutation systems with similar neutron energy spectra.

Introduction

In order to transmute efficiently the long-lived nuclear waste, the high intensity neutron source is needed. An inertial confinement fusion (ICF) device (based on D+T → ^{4}He+n nuclear reaction) could provide a powerful neutron source - 1 MW fusion power corresponds to ~ 4×10^{17} n/s [1]. A molten salt blanket (LiF-BeF_2-$(HN)F_4$ – "flibe"), surrounding this neutron source, then could serve as a medium for transuranium actinides (TRU) to be burned. Flibe also has a function of both coolant and carrier of tritium breeding material (^{6}Li in this case). A well known advantage of the molten salt is a possibility of both refueling of burned TRU and extraction of fission products (FP) on-line. The averaged neutron flux is very high (of the order of ~1.5×10^{15} n s^{-1} cm^{-2}) and corresponds to the flux typical only for high flux reactors. Fig. 1 presents a simplified geometry setup used in calculations. The diameter of cavity with Fusion Device (FD) is 400 cm, surrounded by 1 cm thickness liquid flibe (^{6}Li 0.1% in Li) wall, 0.3 cm – metallic wall (SS316, 50%), and 1 cm graphite. 60 cm thickness transmutation blanket with flibe density – 2 g/cm^3 and corresponding TRU density – 0.0074 g/cm^3 (devided into 3 zones - in yellow) is placed between the graphite and metalic wall. ^{6}Li enrichment in flibe blanket is 0.6%. All structure was surrounded by 20 cm thickness graphite-reflector and 5 cm thickness stainless steel shell [2].

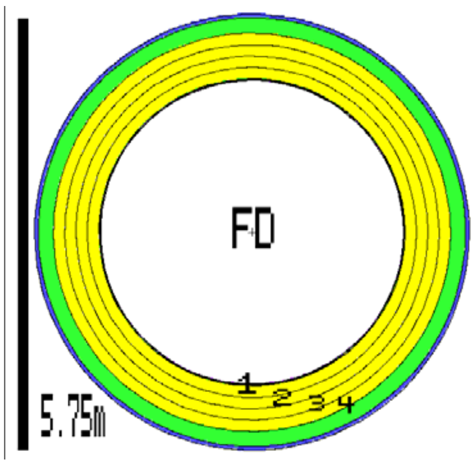


Figure 1. A simplified geometry model of the fusion-fission hybrid system.

All calculations on the flibe-based actinide transmutation blanket were made employing the Monteburns code system [3]. MCNP [4] was used to obtain k_{eff} of the TRU blanket without an external source in the mode of a criticality eigenvalue problem, and also to estimate neutron flux as well as k_{scr} (the total neutron multiplication coefficient) in its external source

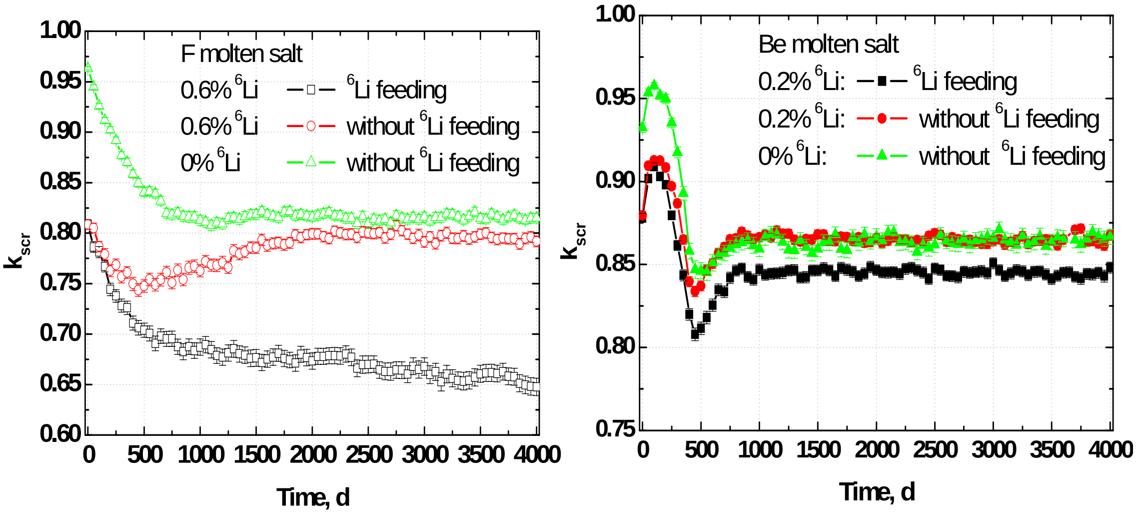
* ridikas@cea.fr

mode. k_{scr} is defined as: $k_{scr} = (M_n - 1)/(M_n - 1/\nu)$, where ν is the average number of neutrons per fission and M_n is a total neutron multiplication factor of the system [3, 4].

During simulation the fission power was kept constant ($3GW_{th}$), what corresponds to a variable fusion power of the fusion device. This implied a renormalization of the absolute neutron flux when an effective neutron multiplication coefficient of the system is changed. For all structure materials and actinides we have chosen the ENDF data files [5] being most frequently employed, while for fission products the JENDL data library [6] was taken due to the largest number of fission products available (~200).

Optimization of Minor Actinides (MA) incineration

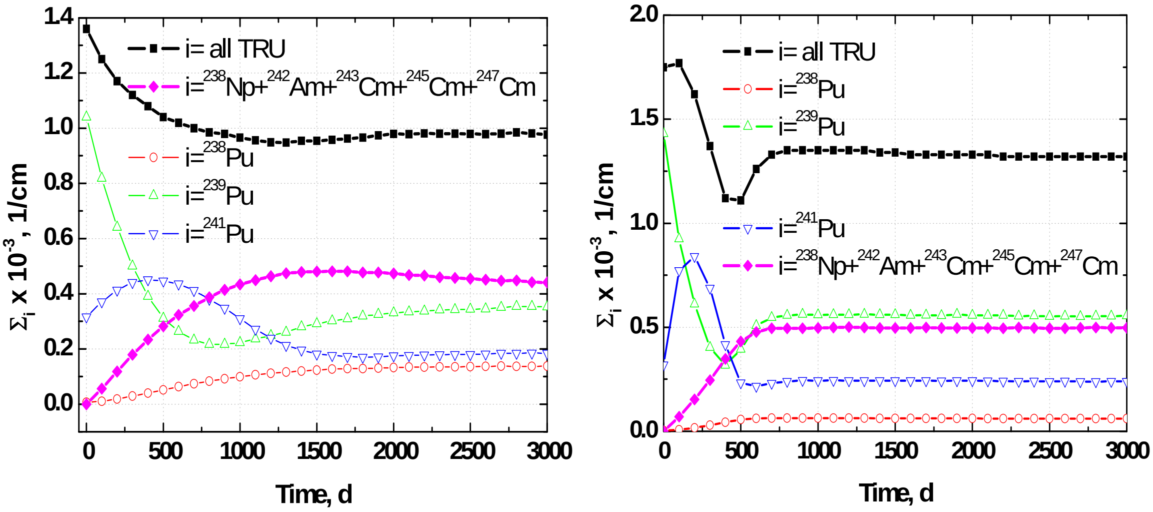
The minor actinides transmutation scenarios in the fusion-fission hybrid system molten salt blanket were investigated. To optimize the neutronic characteristics of the blanket and to improve the minor actinides transmutation process two different molten salt compositions were tested. The first molten salt blanket ($2LiF$-BeF_2-$(HN)F_4$) consists of: 28.57% - ^6Li+^7Li, 14.29% - Be, 57.13% - F (*F salt*), the second one is a modified molten salt blanket, where more than half of the blanket volume is occupied by Be: 14.29% - ^6Li+^7Li, 57.14% - Be, 28.57% - F (*Be salt*). The starting transuranium composition in the molten salt was Pu and minor actinides separated from LWR spent nuclear fuel of 30 MWd/kg burnup, the subsequent feeding - only minor actinides from the same spent fuel. Initial TRU mass is 3.04 tons in the *F - salt* blanket and 1.52 tons in the *Be salt* blanket. The fission products have been removed continuously during the irradiation.



(a) (b)

Figure 2. The behavior of k_{scr} in F-salt (a) and in Be-salt (b) with different intial ^6Li quantity and feeding modes.

In addition to the different flibe composition, the influence of ^6Li enrichment in Li on molten salt blanket characteristics was tested. The $^6Li(n,\alpha)T$ reaction is very important for the tritium breeding, but at the same time has a strong influence on k_{eff} and k_{scr} of the transmutation blanket. The hybrid system performance parameters with the *F-salt* blanket containing 0% and 0.6% of ^6Li in lithium with different ^6Li feeding modes, and with the Be-salt blanket containing 0% and 0.2% of ^6Li in lithium with different ^6Li feeding modes were analyzed. k_{scr} behavior in the different molten salt blanket cases with different ^6Li treating options is presented in Figs. 2a and 2b. The best system performance results in terms of k_{scr} and, accordingly, in terms of the fusion power stability were obtained in the case of the *F-salt* blanket with 0.6% ^6Li and without ^6Li feeding (k_{scr} max. fluctuation during irradiation 0.74÷0.81, P_{fus} - 188÷301 MW, at equilibrium stage k_{scr}=0.79±0.004, P_{fus}~230 MW) and in the case of the *Be-salt* blanket with 0.2% ^{6}Li and without ^{6}Li feeding (k_{scr} max. fluctuation 0.83÷0.91, P_{fus} 100÷210 MW, at equilibrium stage k_{scr} 0.86±0.003, P_{fus}~164 MW).



(a) (b)

Figure 3. Averaged macroscopic fission cross sections for fissile isotopes (a) in F based molten salt with 0.6% ^{6}Li without Li feeding and (b) in Be based molten salt with 0.2% ^{6}Li without Li feeding.

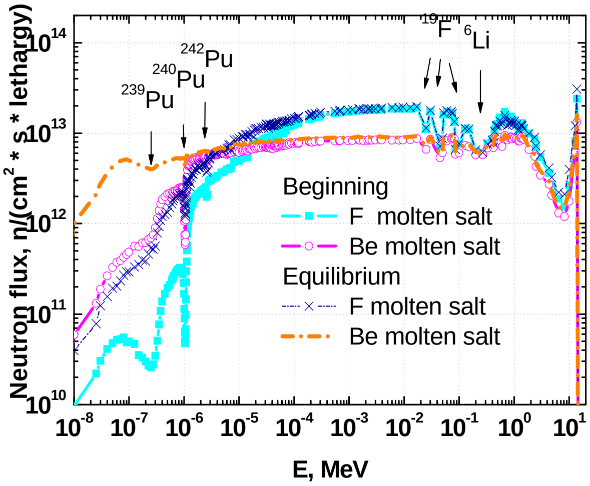


Figure 4. Averaged neutron energy spectra at the beginning of irradiation and at equilibrium stage for F and Be molten salt blankets.

The observed k_{scr} behavior can be explained by fissile isotope quantity and interaction with neutron properties in the blanket. In Fig. 3 averaged macroscopic fission cross sections are presented for ^{238}Pu, ^{239}Pu,^{241}Pu isotopes, MA and all TRU in F- and Be-salt blankets. At the beginning ^{239}Pu is a dominant fissile isotope in F-salt. Due to incineration of ^{239}Pu k_{scr} is decreasing till ^{241}Pu and some other fissile nuclei start dominate fission process in the blanket. In Be-salt the sharp variation in k_{scr} at the early stage of operation is explained by fast plutonium isotope mass equilibration process. At the beginning k_{scr} increases due to ^{240}Pu conversion to ^{241}Pu, and later decreases due to intensive burning of plutonium isotopes (^{239}Pu, ^{240}Pu ^{241}Pu). k_{scr} increases again before equilibrium is reached due to ^{238}Pu accumulation from different chains (^{237}Np(n,γ)→^{238}Np(β-)→^{238}Pu, and (^{241}Am(n,γ)→^{242}Am(β-)→^{242}Cm) ^{242}Cm(α)→^{238}Pu. ^{238}Pu converts to ^{239}Pu by (n,γ) reaction compensating ^{239}Pu disappearance. In both salt cases the same incineration rate (1.1 tons/year) of actinides was obtainded, but in the case of Be-salt the equilibrium stage is reached after 2 years and in F-salt case – only after 5 years. The reason for different plutonium incineration in F and Be molten salt blankets is different neutron spectra. The neutron spectra for both transmutation blankets are presented in Fig. 4. The thermal neutron contribution in the neutron spectrum of Be-salt medium is ~10 times larger at the beginning of irradiation and at equilibrium stage as compared with F-salt. By comparing two molten salt transmutation media, better hybrid system performance parameters were obtained in the Be-salt case: a lower fusion power to sustain 3 GW_{th} is needed, an equilibrium is reached faster, the total mass of transuranium elements in the blanket is smaller, so the criticality safety and radiation protection concerns are of somewhat smaller scale. In addition, TRU are incinerated more effectively.

Sensitivity to different nuclear data libraries

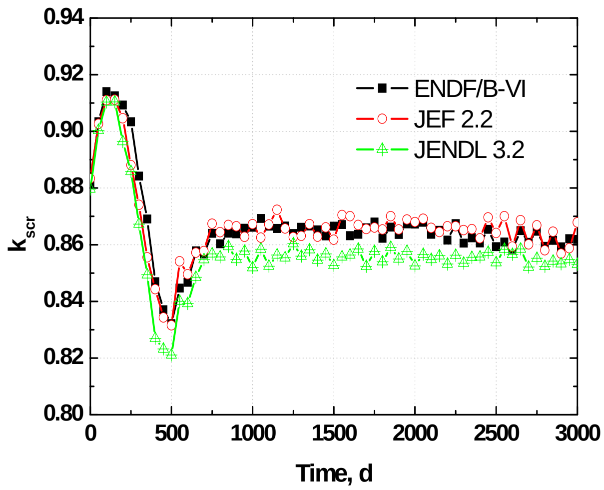


Figure 5. Evolution of k_{scr} in the transmutation blanket for different actinide data files used for calculation (1σ=0.004).

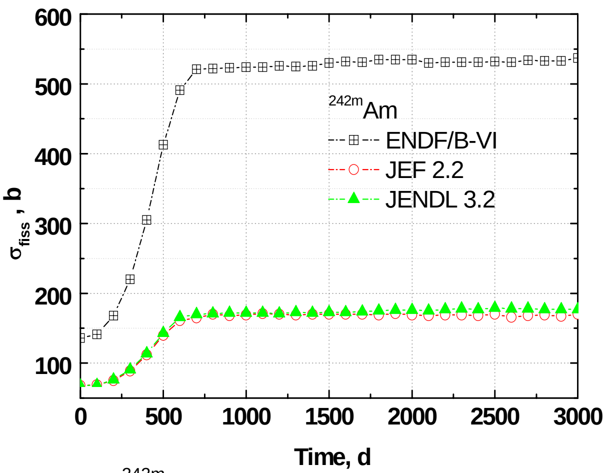


Figure 6. ^{242m}Am one group fission cross sections in molten salt blanket for different data files.

Investigated above optimal Be transmutation blanket in equilibrium is dominated by MAs for which the cross sections may vary depending on the data libraries used. Therefore, we decided to test the performance of the hybrid system by comparing different sets of data libraries in terms of k_{src}, equilibrium conditions, neutronics characteristics and the evolution of the fuel composition in particular. In all cases the same Monteburns code system was used making our results dependent only on the evaluated data tables [3, 4].

First the different data files (ENDF/B-VI [5], JEF 2.2 [7] and JENDL 3.2 [6]) for actinides were tested. ENDF data files were taken for all structure materials, while JENDL was chosen for fission products as previously.

The neutron spectrum obtained in the blanket and the performance of the system was quite similar for all actinide data files considered. On the other hand the differences in k_{scr} and actinides mass evolution were obtained due to non negligible differences in MA cross sections. Differences in k_{scr} in the case of JENDL 3.2 were observed (see Fig. 5). are due to higher fission cross section of ^{238}Np. Consequently less of ^{238}Np by β^- decay is converted to ^{238}Pu (by ~30 kg) and less ^{239}Pu (by ~4 kg) is produced by (n, γ) reaction. At equilibrium less of ^{242m}Am and ^{243}Am were obtained with ENDF due to higher fission cross section of ^{242m}Am as it is presented in Fig. 6.

Large differences were also observed for Cm isotopes both for capture and fission cross sections (see Fig. 7 for details). In the case of ^{243}Cm the capture and fission cross sections differ in 10% at the beginning (epithermal neutron flux) and 20% in equilibrium stage (more thermalized neutron spectrum). 8% difference in one group capture cross sections is observed for ^{244}Cm (its accumulation in molten salt is substantial ~240 kg). 10% deviation in fission cross section is observed for ^{245}Cm at equilibrium.

The sensitivity analysis was also performed using different data libraries for molten salt structure materials. Now the ENDF data file was used for all actinides. As it is shown in Fig. 8, JEF data files used for structure materials give very different level of sub-criticality in the system. The large differences of the entire system behavior are caused by discrepancies in ^{9}Be elastic scattering and (n,2n) reaction cross section.

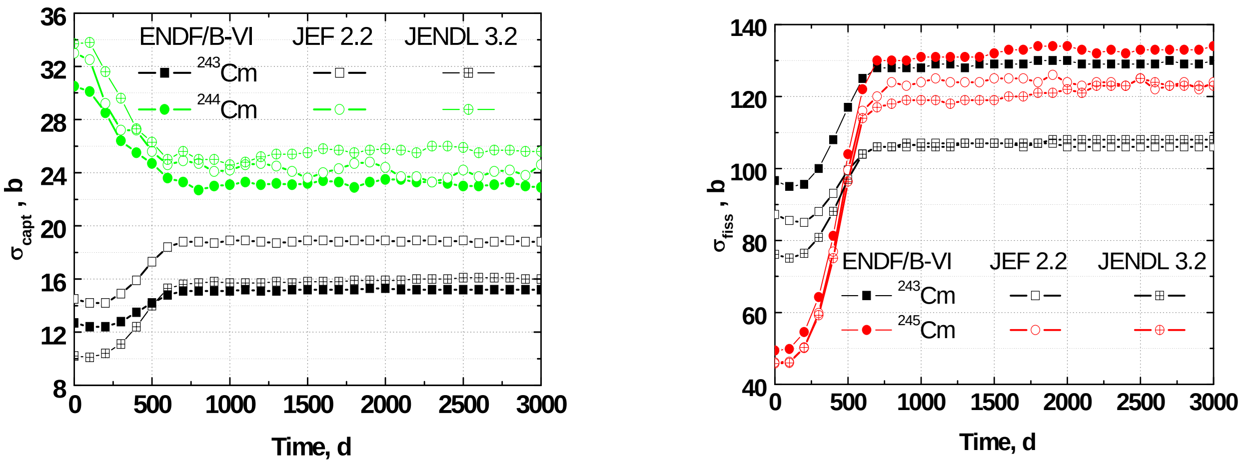


Figure 7. One group capture and fission cross sections for Cm isotopes in the molten salt blanket with ENDF, JEF and JENDL data libraries.

Indeed, molten salt transmutation blanket enriched in ^{9}Be (57.14%) is extremely sensitive to its cross sections. The absence of ^{9}Be (n,2n) reaction (in the case of JEF) gives different neutron multiplication value in the system. With JEF the amount of ^{9}Be remains constant, but with ENDF about 300 kg ^{9}Be and in JENDL case about 100 kg ^{9}Be are transmuted during the irradiation. Accordingly the bigger contribution to neutron propagation from ^{9}Be(n,2n) reaction is with ENDF and JENDL data files.

Additionally the elastic scattering cross section for ^{9}Be in JEF 2.2 data file is less by 0.15 b (2.5%) comparing with ENDF/B-VI and JENDL 3.2. This difference results in slightly different level of neutron flux thermalisation in the blanket – about 1% less thermal neutrons, and about 1% more epithermal neutrons were obtained with JEF. Consequently, different one group cross sections for actinides are calculated with JEF as presented in Fig 9.

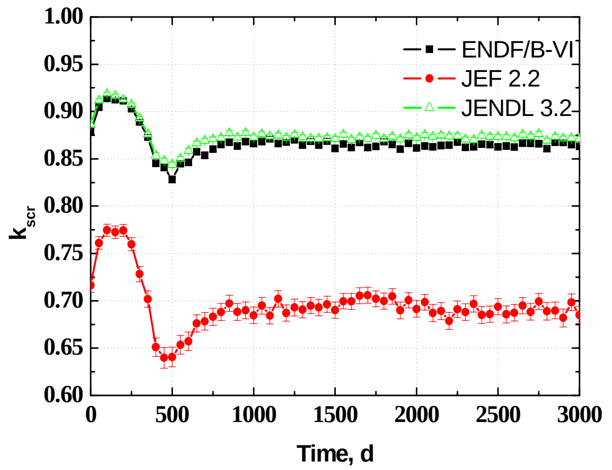


Figure 8. k_{scr} evolution in the transmutation blanket in case of different data files for structure materials (ENDF, JENDL 1σ=0.002, JEF 1σ=0.008).

Conclusions

The MA transmutation scenarios in the fusion-fission hybrid system with two different molten salt compositions were investigated. In both F-salt and Be-salt cases the same incineration rate (1.1 tons/year) of actinides was obtained, but Be-salt shows better hybrid system performance in terms of k_{src} behavior, requested fusion power, equilibration period and actinide mass in the blanket.

A number of major differences among ENDF, JENDL and JEF data files were identified and quantified in terms of the averaged one-group cross sections. One should note that performance of the system is very similar despite non negligible differences in the cross-sections comparing calculations with different nuclear data files for actinides. The worst situation is for Am and Cm isotopes in the entire energy range. ^{238}Np(n,f) cross-section should be measured as the major parameter for the Np-Pu chain. New evaluations should be done for the $^{241\text{-}242m}$Am capture and fission.

Our analysis with different construction material data files showed significant differences in elastic scattering and (n,2n) reaction of ^{9}Be from JEF, what strongly influences the entire system behavior. The present study could be directly generalized for other transmutation systems, characterized by a similar neutron energy spectra.

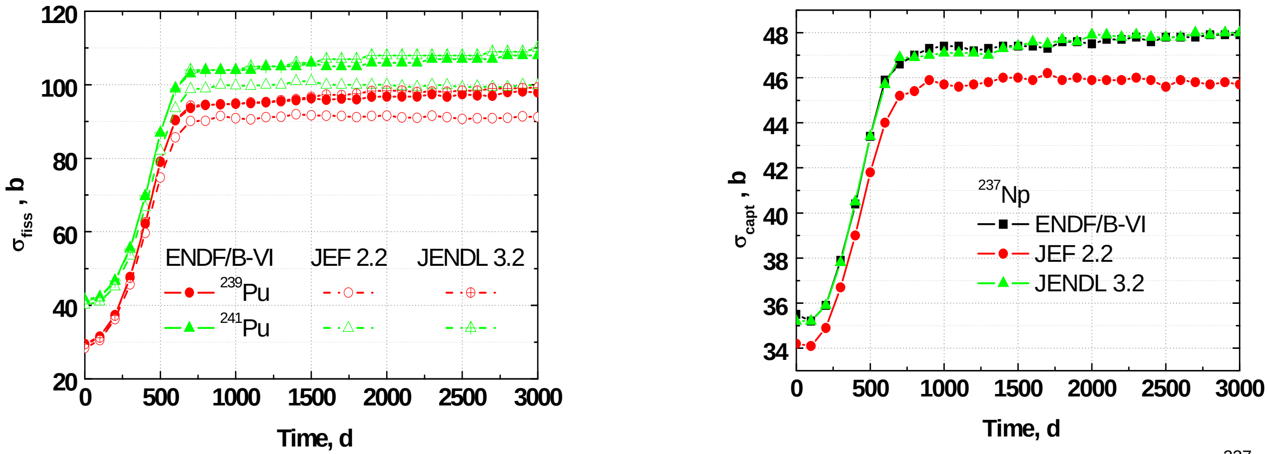


Figure 9. One group fission cross sections for fissile Pu isotopes and capture cross section for ^{237}Np obtained in the molten salt blanket with different data libraries for structure materials.

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