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Content

Abstract:

In this report the depletion codes ORIGEN 2.2 and ORIGEN-Jül were compared with respect to pure decay calculations. For two different isotope compositions (burned low enriched uranium fuel (9.7 wt%), burned plutonium fuel with high fraction of minor actinides) the decay of initial isotope compositions are calculated for certain time periods up to 10^6 years and the differences of the isotope masses are analysed. It is demonstrated that while the nuclear data used in the codes are slightly different the masse of the isotopes for a given decay time period are in good agreement within the limits of -0.02 % - 0.08 % for burned LEU fuel and -0.3 % – 0.7 % for burned Pu-MA fuel. For safety related assessments, e.g. final radioactivity the accuracy in determination of the isotope mass is dominating. It can be concluded that by calculating nearly equivalent isotope compositions after certain decay time periods both codes therefore will also calculate comparable values of radioactivity and radio toxicity for decay series of individual isotopes.

Analysing the relative difference of individual isotope masses significant higher differences could be observed. The largest differences were identified for isotopes with nearly negligible mass fractions. It is shown that these differences are mainly caused by differences of the decay constants and the branching ratios used in ORIGEN 2.2 and ORIGEN-Jül. The influence of additional isotopes considered in the library of ORIGEN 2.2 are negligible. The largest mass fraction in the ORIGEN 2.2 calculation is observed for Pa-234 with 2.1e-16 of the total mass. Nevertheless with respect to precise determination of very small isotope mass fractions an update of the ORIGEN-Jül library to the latest nuclear data (e.g. ENDF/B-VII, JEFF-3.1) should be realised.

1. Introduction:

Assessing safety aspects of burned nuclear fuel requires calculations to the time dependent decay of radioactivity. For thermal reactors this can be done by calculating the power history dependent burn-up with the code system VSOP /1/. While for calculations of reactor specific parameters only isotopes with significant neutronic importance (absorption, scattering) are taken into account for the determination of the radioactivity of the burned fuel all isotopes generated by nuclear reactions need to be calculated. This is realised by using the program module ORIGEN-Jül /2/ which is strongly coupled to VSOP. In this program module the calculated neutron flux and if existing the condensed cross sections of the isotopes generated in VSOP were directly used for comprehensive decay calculations. These calculations were done for every core region considered by VSOP and for the whole burn-up time. Because of the strong coupling of ORIGEN-Jül to VSOP a decay calculation for an individual isotope is very complicated compared to ORIGEN 2.2 /3/. Therefore a combination of VSOP/ORIGEN-Jül and ORIGEN 2.2 was initiated were the burn-up is calculated by VSOP/ORIGEN-Jül and subsequently the decay series for individual isotopes of this burned fuel is calculated by ORIGEN 2.2. To ensure reliable results by this combination it is essential to demonstrate that VSOP/ORIGEN-Jül and ORIGEN 2.2 will calculate equivalent amounts of isotopes after certain decay time periods for a fixed initial isotope composition.

This report comprises comparisons between the ORIGEN-Jül code and the latest ORIGEN version 2.2. The ORIGEN-Jül code is based on a previous version of ORIGEN and was adapted to the VSOP code to calculate the burn-up and the decay of isotopes in more detail. In VSOP 28 important actinides and up to 48 fission products with significant neutron importance are explicitly considered. To compensate the difference to the real absorption rate an artificial fission product is taken into account. The fraction of this artificial fission product was calculated by the version of ORIGEN also used for the ORIGEN-Jül module. Therefore burn-up and decay calculation of VSOP and ORIGEN-Jül are consistent.

The latest version of the ORIGEN code 2.2 uses updated nuclear data for burn-up and decay calculation. This report is focused on the difference of the decay calculations with respect to minor actinides and the correlated daughter nuclei with mass numbers higher than 205 (Tl-206 and above). For both codes exactly the same initial isotope composition was used. It was generated by calculating the equilibrium core condition of a thermal high temperature gas cooled graphite moderated reactor (HTR) based on the reference design of the PBMR 400 /4/. Two fuel examples are considered for the decay calculations:

1. burned fuel of plutonium (Pu) and minor actinides (MA) (1.56 g Pu + 0.44 g MA per pebble)
2. burned fuel of low enriched uranium (9 g LEU (9.7 wt% enrichment) per pebble)

While LEU is a standard fuel the Pu+Ma composition is a more theoretical composition to catch the upper limit of plutonium and minor actinide concentration in a fuel.

2. Decay of burned plutonium-minor actinides fuel

Starting with a initial fuel of 1.56 g plutonium and 0.44 g minor actinides per fuel element (pebble) for a final average discharge burn-up of 198.7 MWd/kgHM a wide range of isotopes above mass number 205 are generated. The kernel size of the coated particle used in this calculations is 0.2 mm in diameter. Additional information to this fuel can be found elsewhere /5/. Based on this fuel equilibrium core condition can be achieved for a residence time of 449 days of the fuel elements in the core assuming a six fold reload of the pebbles and then finally discharged. After discharge of the pebbles (1006 pebbles per day) the mass of the total discharged volume is used as the initial isotope composition for both codes (isotope composition see Appendix A).

2.1 Detailed analysis of radioactive decay for selected actinides

The radioactive decay was calculated for several time periods. The time dependent difference of the isotope masses calculated by ORIGEN-Jül and ORIGEN 2.2 are normalized to the isotope mass calculated by ORIGEN-Jül (Fig. 1).

A faster decay for Pu-238 immediately after discharge can be observed up to a final difference of 17.6 % after 1000 years ($\approx 11\ \tau_{1/2}$) Because the lack of long living mother nuclei ($\tau_{1/2}$(Cm-242): 1.63 d, $\tau_{1/2}$(Am-238): 1.63 h, $\tau_{1/2}$(Am-242): 16 h, $\tau_{1/2}$(Am-242m): 141 a, $\tau_{1/2}$(Bk-246): 1.8 d) the mass fraction of Pu-238 vanishs after this time period.

For the isotope Am-241 almost no difference occurs (< 1 %) even for a time period up to 10^4 years ($\approx 23\ \tau_{1/2}$). The difference only is observed for a longer time period of 10^5 years ($\approx 231\ \tau_{1/2}$), where ORIGEN 2.2 calculates 20 % more mass. But for this time period only a very small fraction of the initial isotope is left (reduction by factor 10^6) and the difference is still existing for equivalent decay constants in ORIGEN-Jül and ORIGEN 2.2 (see chapter 5).

For Pu-241 a similar difference can be observed for a time period of 10^5 years. But this isotope has an even shorter half live than Am-241 and the initial mass is reduced by a factor of 10^9. Also this difference is not affected when using equivalent decay constants for the ORIGEN-Jül and the ORIGEN 2.2 calculation. The lower Pu-241 mass calculated by ORIGEN 2.2 for a time period of 10 to 10^4 years is not observable for Am-241. Therefore this difference is caused mainly by a larger decay constant and a smaller branching ratio for β decay (higher fraction of α decay) of Pu-241.

Considering the mother nuclei Cm-245 (α decay, $\tau_{1/2}$ = 8500 years) which dominates the generation of new Am-241 via Pu-241 (Fig. 2) in this time frame no significant differences between ORIGEN 2.2. and ORIGEN-Jül are observable. The very small higher residual mass of Cm-245 calculated by ORIGEN 2.2 would be correlated with a negligible lower production of Am-241. The branching ratios (fraction of the specific type of decay of the isotope as α decay or spontaneous fission) also could induce a varying mass of the daughter nuclei for a given total decay constant. But they are equivalent for ORIGEN 2.2 and ORIGEN-Jül (see chapter 5). Nevertheless the small difference of the decay constant of Cm-245 affects the differences of Am-241 and Pu-241 for longer time periods.

For Am-243 and Pu-240 a very similar difference behaviour can be observed. Because of the longer half life up to a time period of 10^3 years ($\approx 0.2\ \tau_{1/2}$) almost no difference occurs (< 1 %). Then the differences increase up to a time period of 10^5 years ($\approx 14\ \tau_{1/2}$) which is equivalent to a lower isotope mass calculated by ORIGEN 2.2. Later the difference decreases again and nearly vanish for a time period of 10^6 years ($\approx 140\ \tau_{1/2}$). The decay chains of both isotopes are separated and doesn't share a certain mother nuclei. But both isotopes have a similar decay constant. A possible reason for this behaviour is the larger decay constant used

in ORIGEN 2.2. After 10^6 years the initial Pu-240 and Am-243 is decayed and residual differences are induced by the decay of long living mother nuclei (Cm-247 ($\tau_{1/2}$ = 1.56e7 a) → Am-243, Pu-244 ($\tau_{1/2}$ = 8.e7 a) → Pu-240).

The mass difference for Pu-239 is nearly negligible for a time period up to 10^4 years because of their small decay constant. For the following time period up 10^5 years the difference increase up to 5 % which is equivalent to a smaller mass calculated by ORIGEN 2.2. The mother nuclei of Pu-239 is Am-243 which also is calculated with a smaller mass by ORIGEN 2.2. Therefore the reduced mass of Pu-239 is not induced by a slower decay of Am-243. Due to its large decay constant also the production via α decay of Cm-243 is not a probable reason. Again this can be explained by differences in the decay constant of Pu-239 itself.

For nearly the total decay time period the difference of Np-237 is relative small (< 0.3%) except at the time of 10^3 years. The higher mass calculated by ORIGEN 2.2 for this time period is maybe caused by the larger (Am-241) and/or the smaller (Cm-245) decay constant of their mother nuclei.

A significant difference for Pu-242 can only be observed for long time periods > 10^5 years. For this time period ORIGEN 2.2 calculates a higher isotope mass of Pu-242 which result in a deviation of about 4 %. Because all mother nuclei have a significant higher decay constant the possible reason for the observed differences are differences of the decay constants of Pu-242 itself.

The half live of the considered isotopes are given in Tab. 1

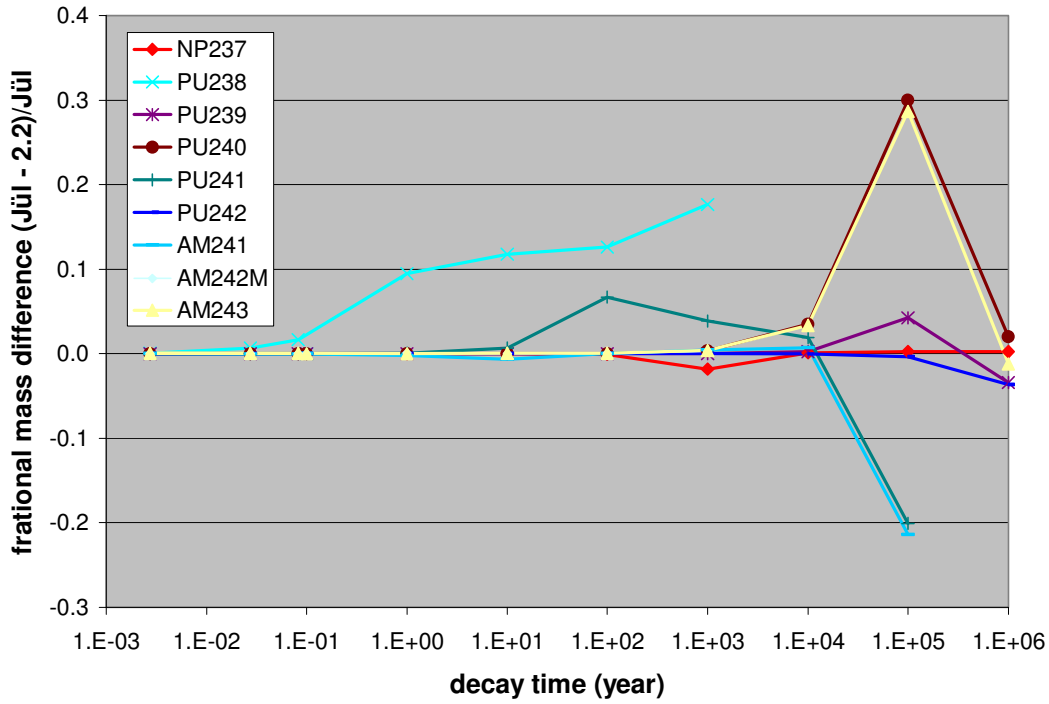

Fig. 1: Mass differences after decay for selected isotopes, normalized to the specific isotope mass calculated by ORIGEN-Jül, for burned Pu-MA fuel

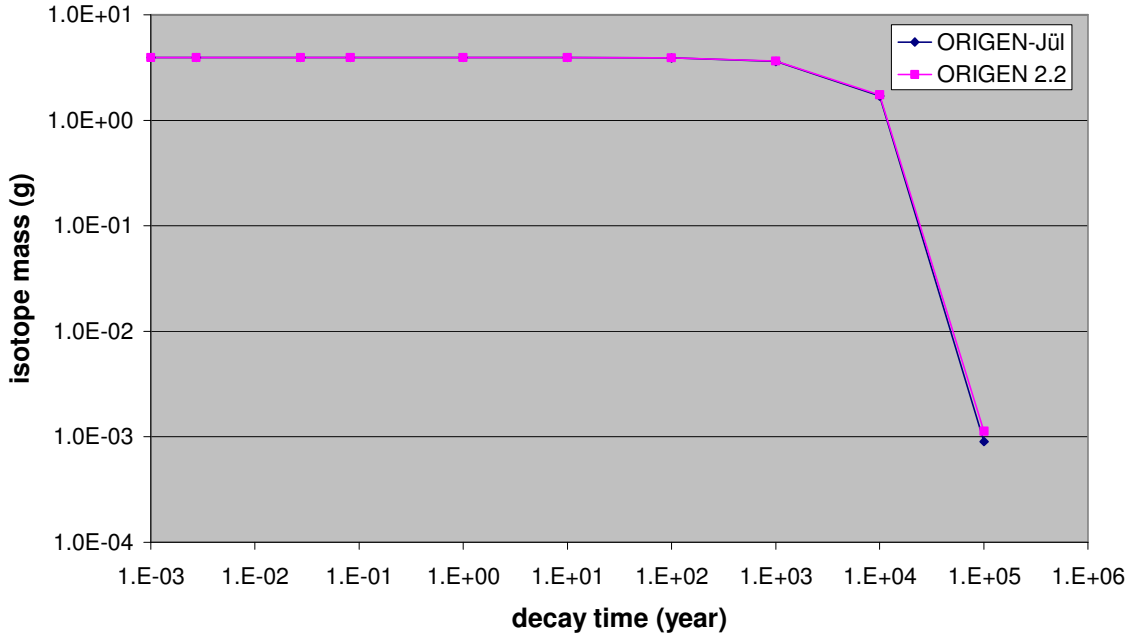

Fig. 2: Total mass of Cm-245 after certain decay time periods (α decay, $\tau_{1/2}$ = 8500 years)

isotopes	$\tau_{1/2}$ (year)	isotopes	$\tau_{1/2}$ (year)	isotopes	$\tau_{1/2}$ (year)
Np-237	2.14 e+6	Pu-240	6563	Am-241	432.2
Pu-238	87.74	Pu-241	14.35	Am-242M	141
Pu-239	2.41 e+4	Pu-242	3.75 e+5	Am-243	7370

Tab. 1: Half life of selected isotopes based on Karlsruher Nuclide Chart 2006 (may differ from the data used in ORIGEN-Jül or ORIGEN 2.2)

2.2 General analysis of decay for heavy metal isotopes (mass number greater 205)

The decay of all actinides and their correlated daughter nuclei are calculated by the two codes ORIGEN-Jül and ORIGEN 2.2 and analysed with respect to the mass difference for the selected time periods 1, 100, 10^4 and 10^6 years. The differences are normalized in the first step to the specific isotope mass calculated by ORIGEN-Jül to see the individual relative differences (Figs. 3-6). In the second step the isotope mass differences are normalized to the total heavy metal mass for the specific time period (Figs. 7-10). This clarify the importance of the individual isotope mass differences to the total isotope composition (some of the isotopes with relative large relative difference have mass fractions several orders of magnitude lower then the total mass).

2.2.1 Isotope mass differences normalized to isotope mass

After 1 year of decay only small differences (< ± 5 %) are observed for the majority of the isotopes (Fig. 3). Only for Po-214 (14 %) and Es-253 (11 %) a lower mass is calculated by ORIGEN 2.2. The differences for Am-244 and Bk-250 probably caused by numerical effects because these isotopes are almost completely decayed ($\tau_{1/2}$ (Am-244) = 10.1 h, $\tau_{1/2}$ (Bk-250) = 3.217 h) after 1 year. Another reason could be small differences of the decay constant which will be evident only for large time periods far beyond the half live.

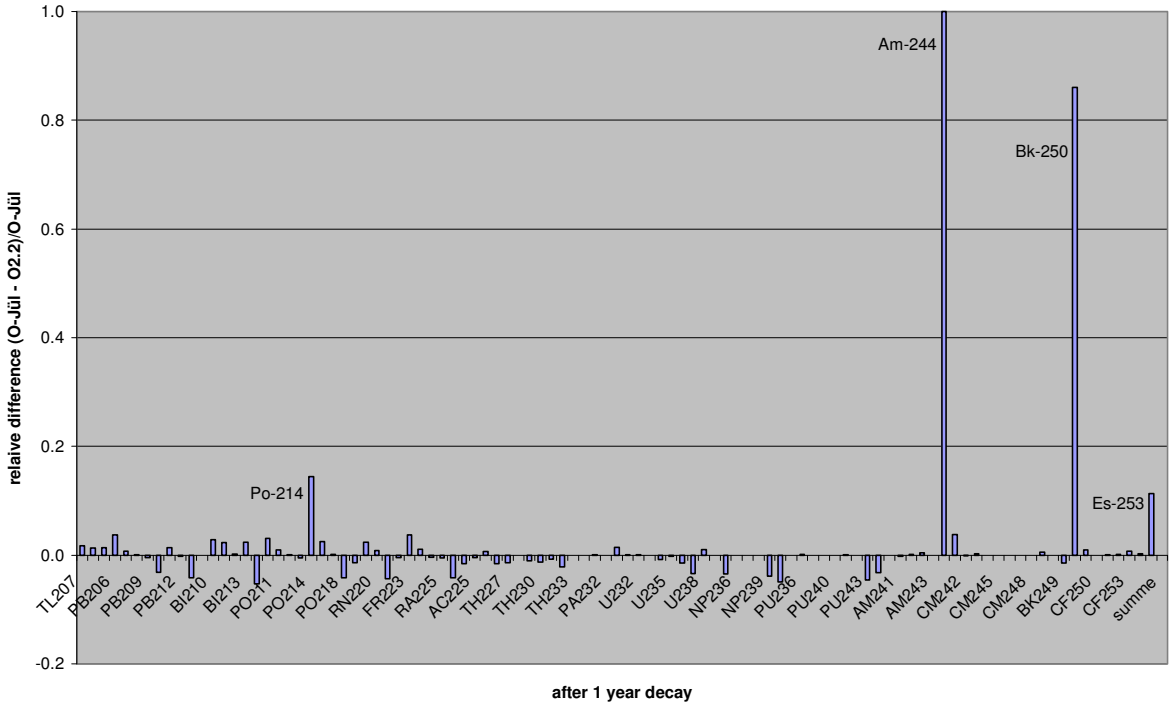

Fig. 3: Relative isotope mass difference after 1 year decay

The decay in a period of 100 years results in higher differences for Cm-243 (24 %) and Cf-252 (9 %) while the difference for Pu-236 results from numerical effects ($\tau_{1/2}$ = 2.858 a) or small differences of the decay constant (Fig. 4). Again Po-214 shows a slightly higher difference (13 %) while Es-253 is completely decayed. A general trend seems to be establish that ORIGEN 2.2 calculates slightly higher isotope masses after 100 years than ORIGEN-Jül which is equivalent to a negative relative difference.

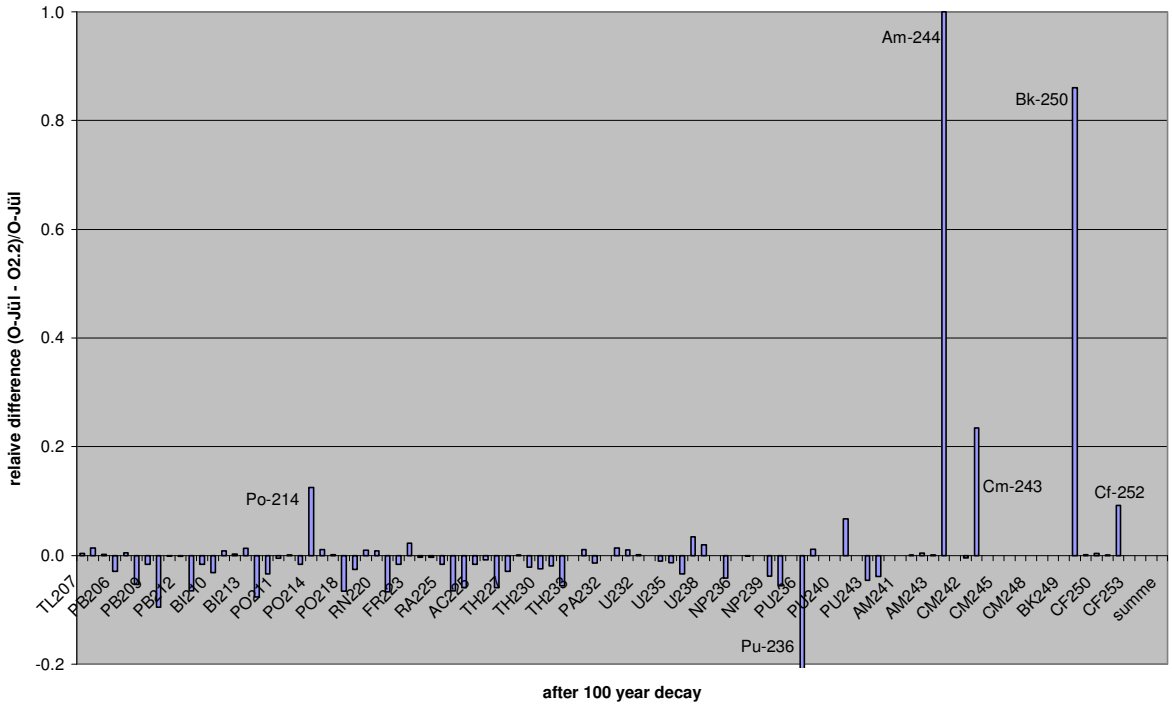

Fig. 4: Relative isotope mass difference after 100 years of decay

After 10^4 years this trend seems to be intensified, only Po-214 has a opposite trend and a positive relative difference (Fig. 5). This continuously growing difference of Po-214 for longer time periods is a strong indication for different decay constants used in ORIGEN-Jül and ORIGEN 2.2. The differences of Am-244, Cm-244 ($\tau_{1/2}$ = 18.1 a), Bk-250 and Cf-250 ($\tau_{1/2}$ = 13.08 a) again reflect numerical effects for isotopes almost completely decayed or small differences of the decay constant.

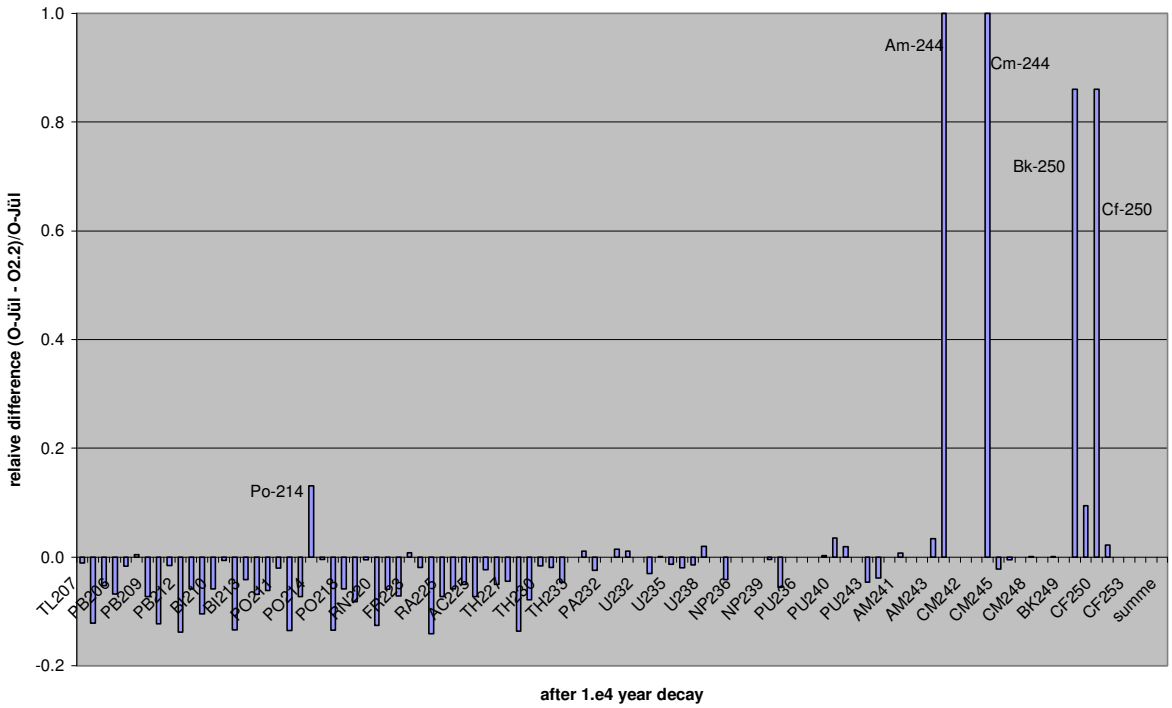

Fig. 5: Relative isotope mass difference after 10^4 years of decay

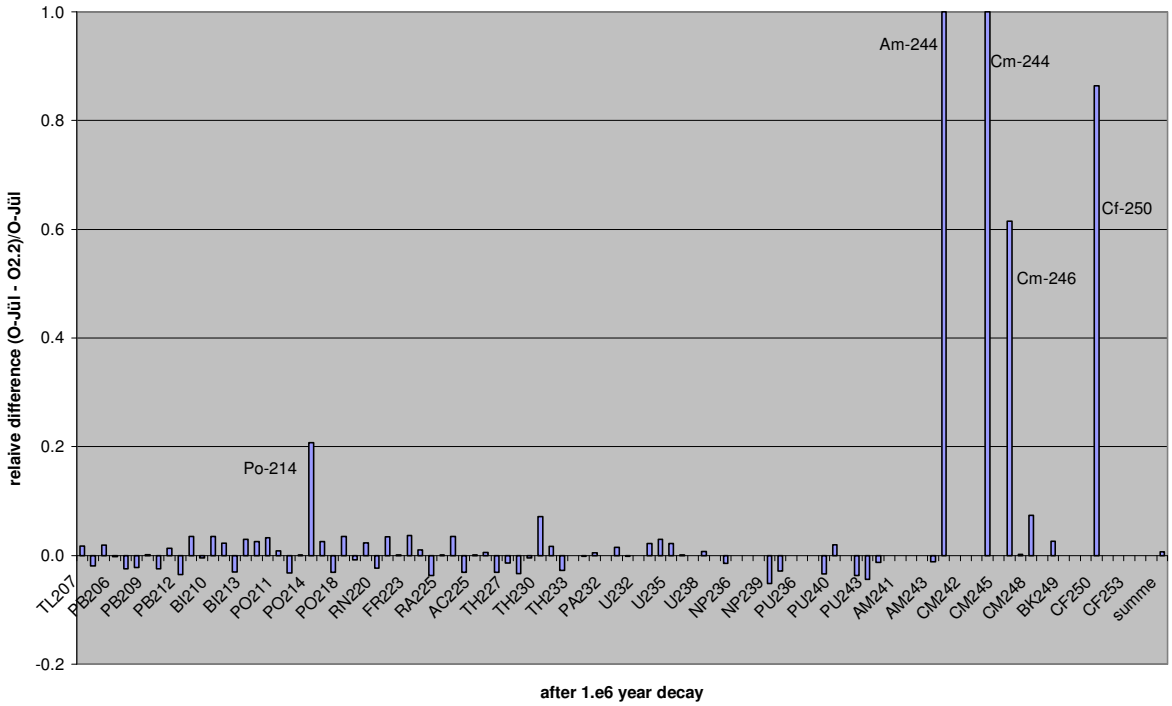

Fig. 6: Relative isotope mass difference after 10^6 years of decay

After 10^6 years the trend of the previous time period has vanished and the differences of the isotopes are again small (< ± 5 %) except for Th-230 (7 %) and Po-214 (20 %). For Cm-246 the big difference ($\tau_{1/2}$ = 4730 a) is induced by numerical effects or small differences of the decay constant (Fig. 6). Beside the short half life of Po-214 some of its mother nuclei are long living as U-234 and Th-230. This results also for long decay time periods in a significant amount of Po-214 in the material composition.

2.2.2 Isotope mass differences normalized to total heavy metal mass

When normalizing the mass differences to the total heavy metal mass for a specific decay time period the dominant isotopes with respect to the total mass of the material composition can be identified. While this comparison doesn't take into account the varying neutron absorption cross section of the different isotopes it nevertheless is an indication which isotopes are dominant in the considered material composition. Additionally this normalized difference allow a quantitative comparison of decayed masses of mother nuclei and generated masses of daughter nuclei. If this values vary constantly between the both codes it is an indication for a changed decay constant. But if only the mass of the mother or the daughter nuclei varies this indicates a change of the fractions of the different decay channels and if existing the spontaneous fission.

After a decay time of 1 year the mass differences are nearly negligible (< ± 0.02 %) (Fig. 7). The similar difference of Pu-241 (1.1e-4) and Am-241 (-1.1e-4) indicates a larger decay constant of Pu-241 used in ORIGEN 2.2.

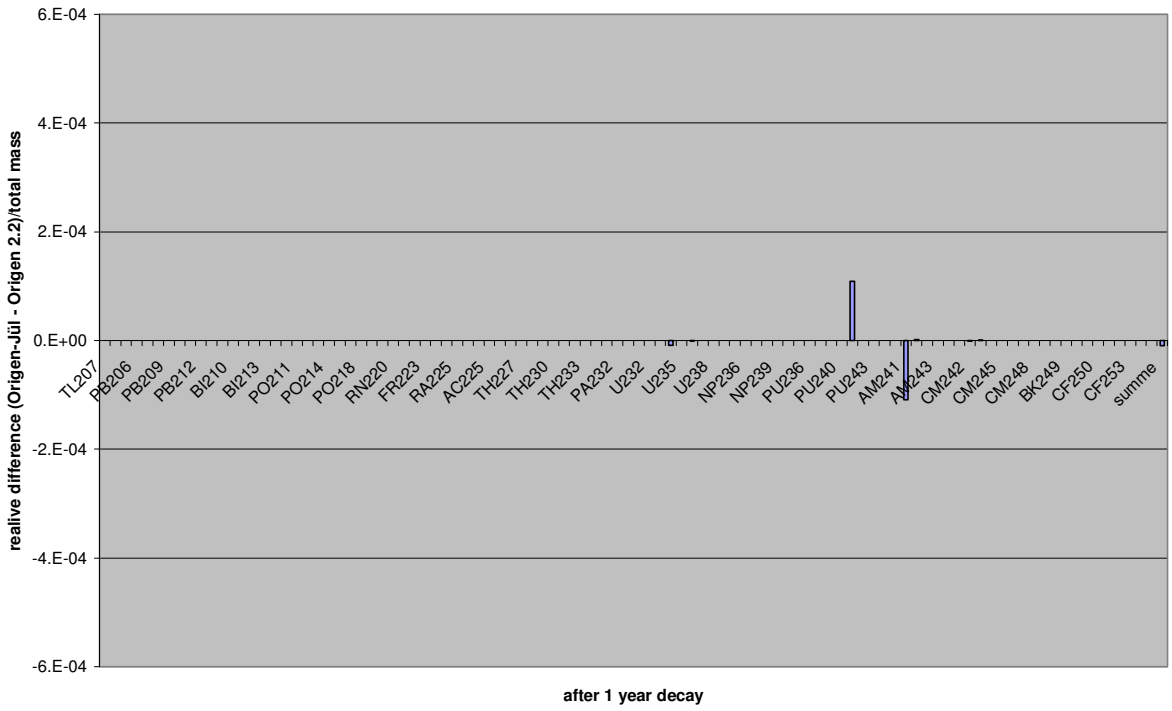

Fig. 7: Isotope mass difference after 1 year decay, normalized to total heavy metal mass

After 100 years of decay slightly larger differences can be observed for U-234 (- 0.05 %) and Pu-238 (0.05 %) (Fig. 8). The similar value of the differences and the fact that Pu-238 is a mother nuclei of U-234 indicate a larger decay constant in ORIGEN 2.2 for Pu-238.

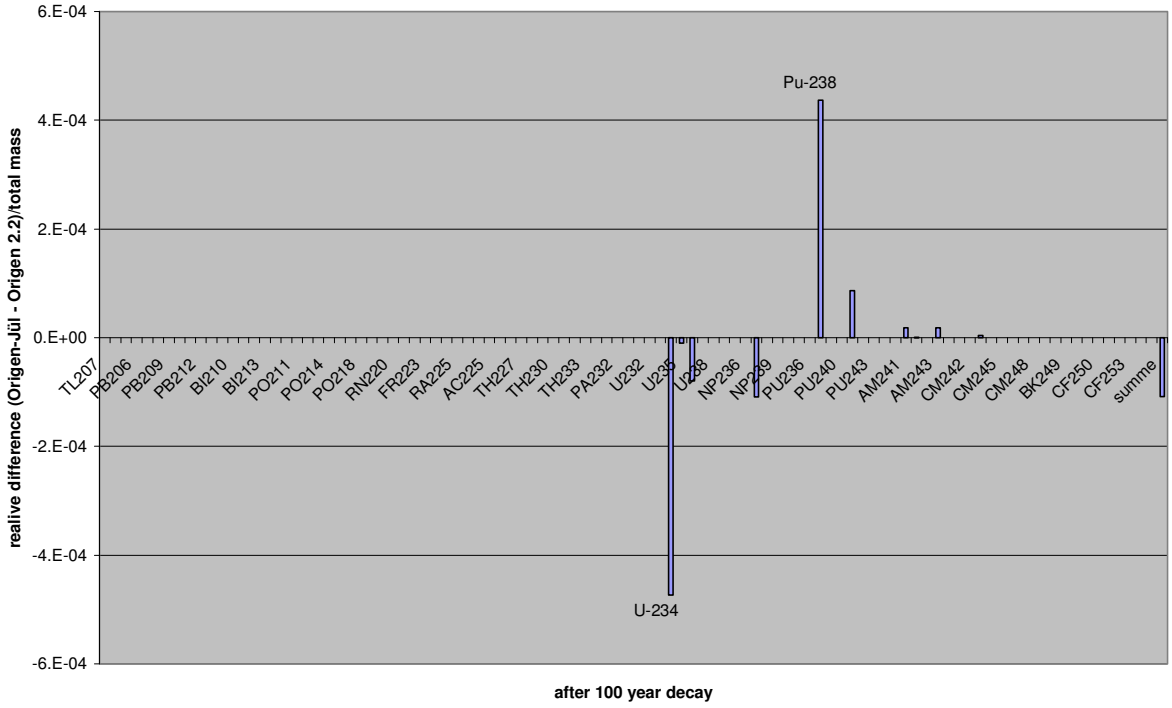

Fig. 8: Isotope mass difference after 100 year decay, normalized to total heavy metal mass

After 10^4 years of decay negative differences can be observed for U-235 (- 0.09 %) and U-236 (- 0.3 %) which is equivalent to higher masses calculated by ORIGEN 2.2. Lower masses were calculated by ORIGEN 2.2 for Pu-239 (0.06 %) and Pu-240 (0.3 %) which result in positive differences (Fig. 9). Again the similar difference values for the mother – daughter nuclei connection Pu-240 → U-236 and Pu-239 → U-235 indicate larger decay constants for Pu-239 and Pu-240 used in ORIGEN 2.2.

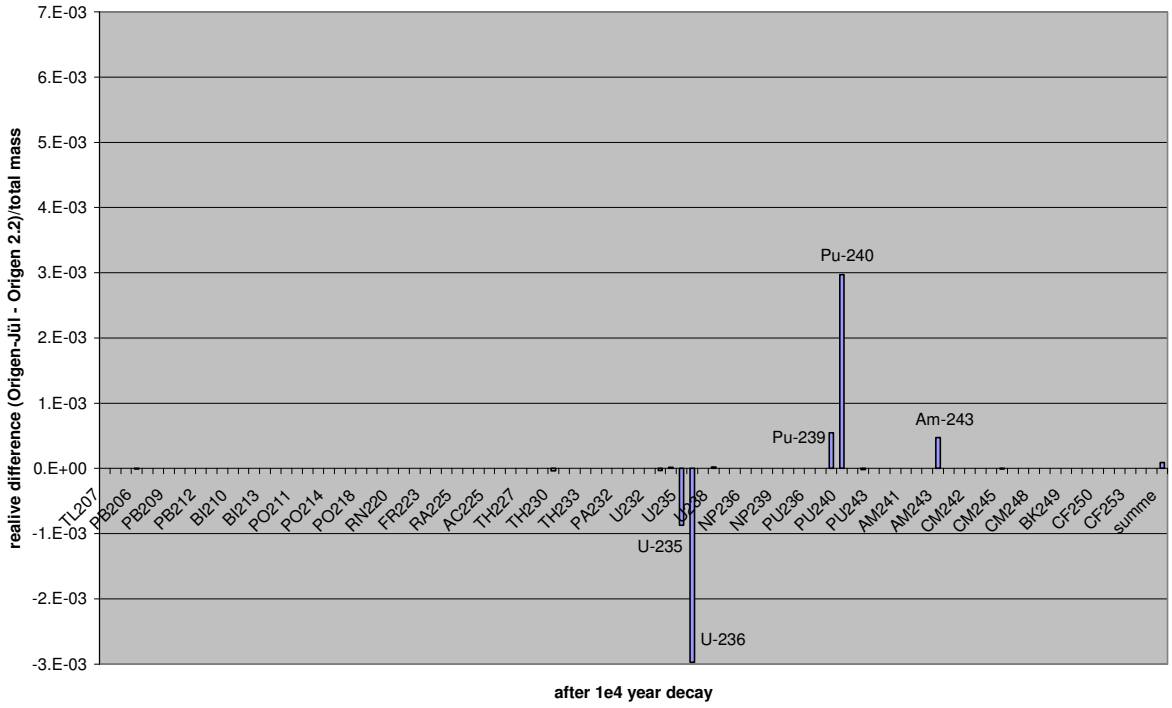

Fig. 9: Isotope mass difference after 10^4 year decay, normalized to total heavy metal mass

After the final decay time period of 10^6 years significant differences have to be observed for U-235 (0.6 %) and the total amount of heavy metal (0.7 %) (Fig. 10). The lower amount of U-235 calculated by ORIGEN 2.2 indicate a higher decay constant for U-235. The production of new U-235 via decay of Pu-235 is negligible for this long time period therefore the decay of U-235 is dominating. Smaller differences can be observed for Bi-209 (- 0.03%), U-233 (0.04 %), U-238 (0.04 %) and Pu-242 (- 0.04 %). The similar differences for Pu-242 and U-238 and their mother – daughter connection indicate a smaller decay constant for Pu-242 used in ORIGEN 2.2.

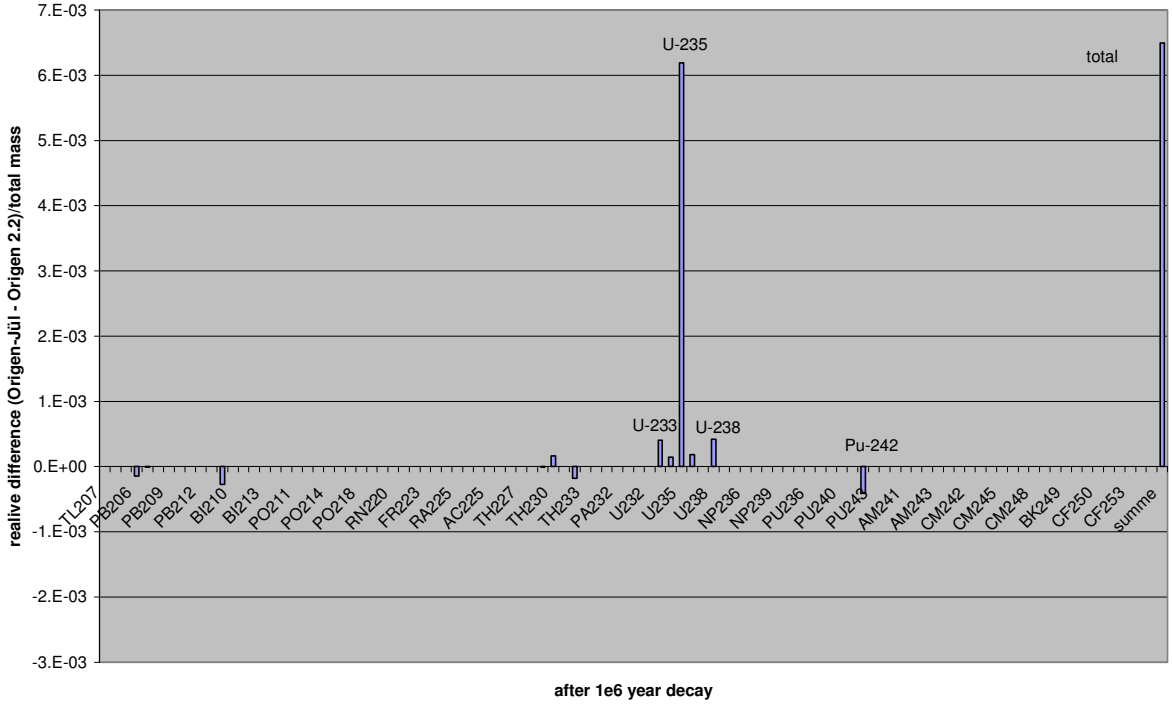

Fig. 10: Isotope mass difference after 10^6 year decay, normalized to total heavy metal mass

3. Decay of burned low enriched uranium fuel

In contrast to burned Pu-MA fuel considered in chapter 2 burned low enriched uranium (LEU, 9.7 wt% enrichment) has a different isotope composition (isotope composition see Appendix A) at a average final discharged burn-up of 102 MWd/kgHM fuel composition. The second comparison of ORIGEN-Jül and ORIGEN 2.2 therefore is done with this discharged fuel composition to separate general differences between the code and material specific effects. The kernel size of the coated particles used in this calculation was 0.5 mm in diameter.

3.1 Detailed analysis of decay for selected actinides

For the dominating actinides of the burned fuel the time dependent decay is analysed in more detail. The corresponding values of half lives are given in Tab. 2.

isotopes	$\tau_{1/2}$ (year)	isotopes	$\tau_{1/2}$ (year)	isotopes	$\tau_{1/2}$ (year)
U-235	7.038 e+8	Np-237	2.14 e+6	Pu-241	14.35
U-236	2.343 e+7	Pu-239	2.41 e+4	Pu-242	3.75 e+5
U-238	4.468 e+9	Pu-240	6563	Am-241	432.2

Tab. 2: Half life of selected isotopes based on Karlsruher Nuclide Chart 2006 (may differ from the data used in ORIGEN-Jül or ORIGEN 2.2)

For the isotopes considered in chapter 1.1 (Np-237, Pu-239, Pu-240, Pu-241, Pu-242, Am-241) the time dependent behaviour is very similar to burned Pu-MA fuel (Fig. 11). Main effects are the maximum and the following decrease of the difference of Pu-241 for the time period of 10 to 10^5 years which again correspond to the decrease for Am-241 at 10^4 years.

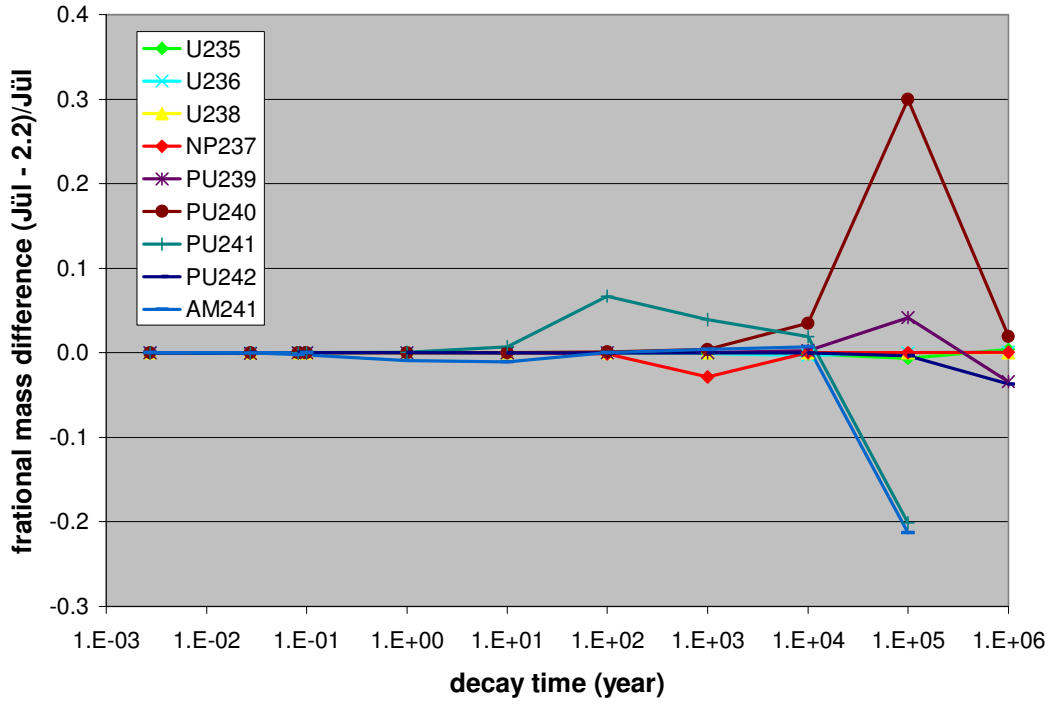

Fig. 11: Mass differences after decay for selected isotopes, normalized to the specific isotope mass calculated by ORIGEN-Jül, for burned LEU

Also the maxima of Pu-239 and Pu-240 at 10^5 years and the minimum of Np-237 at 10^3 years are nearly equivalent to the results in chapter 1.1. For the uranium isotopes the relative difference are negligible (< 0.7 %). The consistent behaviour of the differences of chapter 2.1 and 2.2 indicate that the differences are caused by different decay constants of the considered isotopes and not by different masses or types of their mother nuclei.

Also for the burned LEU fuel the negative differences of Pu-241 and Am-241 observed for a time period far beyond from their half life is not consistent with the higher residual amount of Cm-245 calculated by ORIGEN 2.2 (Fig. 12).

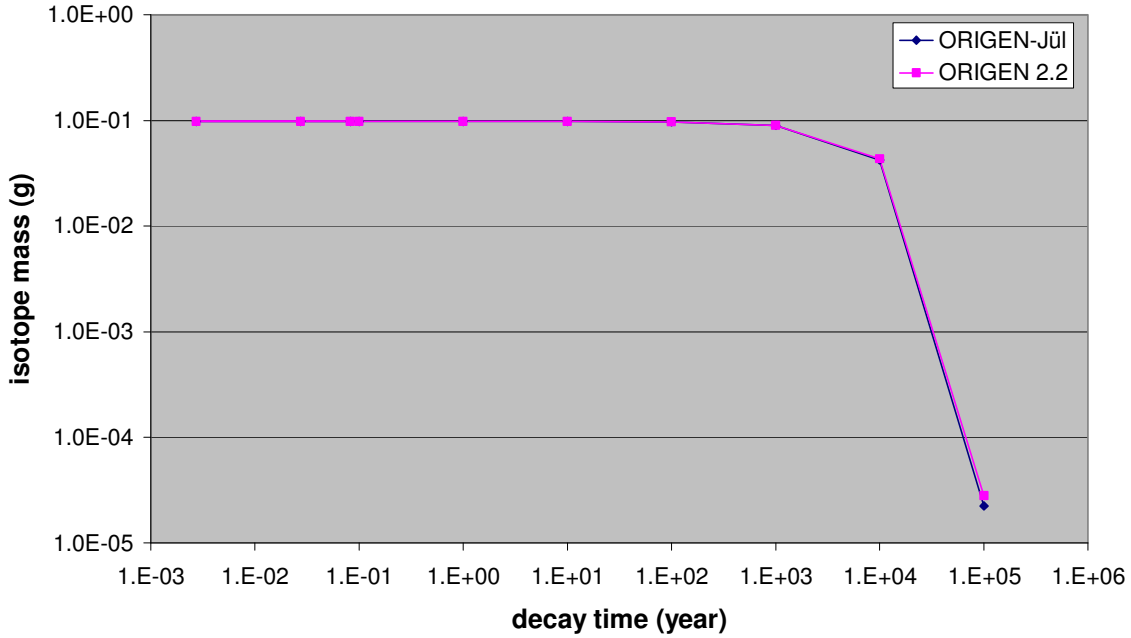

Fig. 12: Total mass of Cm-245 after certain decay time periods (α decay, $\tau_{1/2}$ = 8500 years)

3.2 General analysis of decay for heavy metal isotopes (mass number greater 205)

Again the decay of all actinides and their correlated daughter nuclei are calculated by the two codes ORIGEN-Jül and ORIGEN 2.2 and analysed with respect to the isotope mass difference for the selected time periods 1, 100, 10^4 and 10^6 years. The differences are normalized in the first step to the specific isotope mass calculated by ORIGEN-Jül to see the individual difference of the isotope masses (Figs. 13-16). Equivalent to chapter 2.2 in the second step the isotope mass differences are normalized to the total heavy metal mass for the specific time period (Figs. 17-20).

3.2.1 Isotope mass differences normalized to isotope mass

For a decay time period of 1 year again Po-214 (17 %) and Es-253 (12 %) show the largest differences while the differences of Am-244 and Bk-250 are influenced by numerical effects or small differences of the decay constant. (Fig. 13).

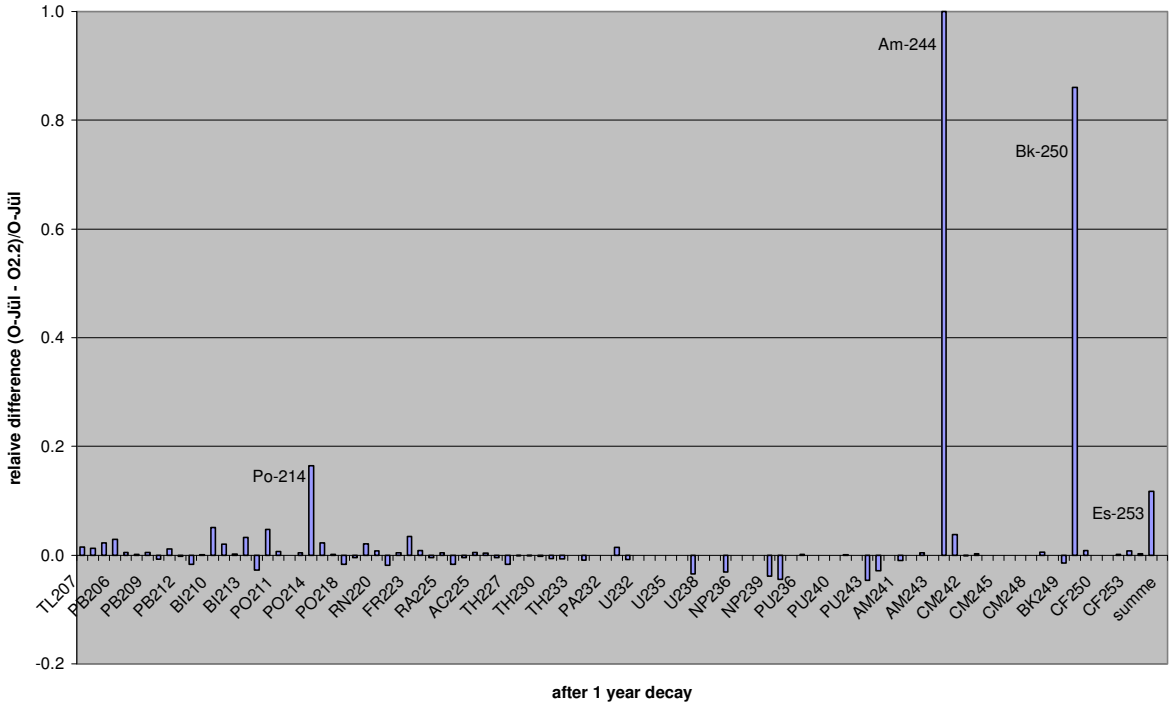

Fig. 13: Relative isotope mass difference after 1 year decay

The decay calculations for 100 years again generate the largest differences for the isotopes Po-214 (14 %), Cm-243 (24 %) and Cf-252 (10 %) (Fig. 14). The differences for Am-244, Bk-250 and Pu-236 are dominated by numerical effects or small differences of the decay constants.

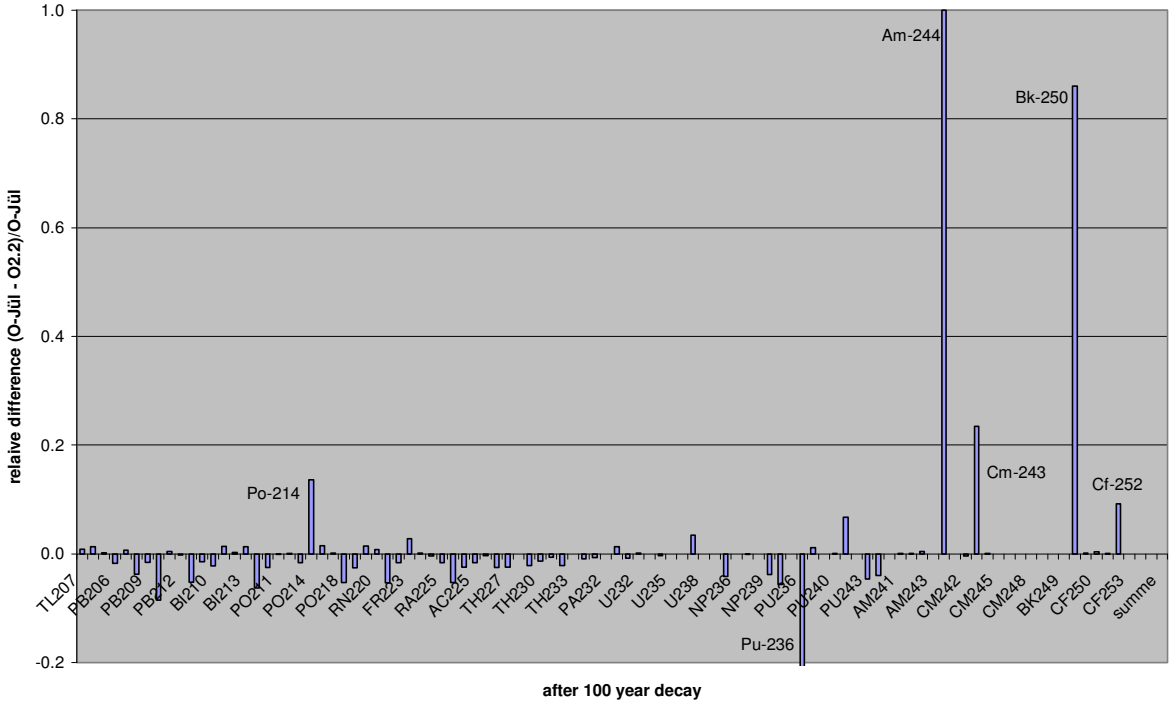

Fig. 14: Relative isotope mass difference after 100 year decay

After 10^4 years the same trend as for burned Pu-Ma fuel can be observed (Fig. 15). ORIGEN 2.2 calculates for this time period predominantly higher isotope masses than ORIGEN-Jül. In contrast Po-214 (14 %) and Cf-249 (10 %) show positive differences. Numerical effects or decay constant differences can be observed when determining the differences for Am-244, Cm-244, Bk-250 and Cf-250.

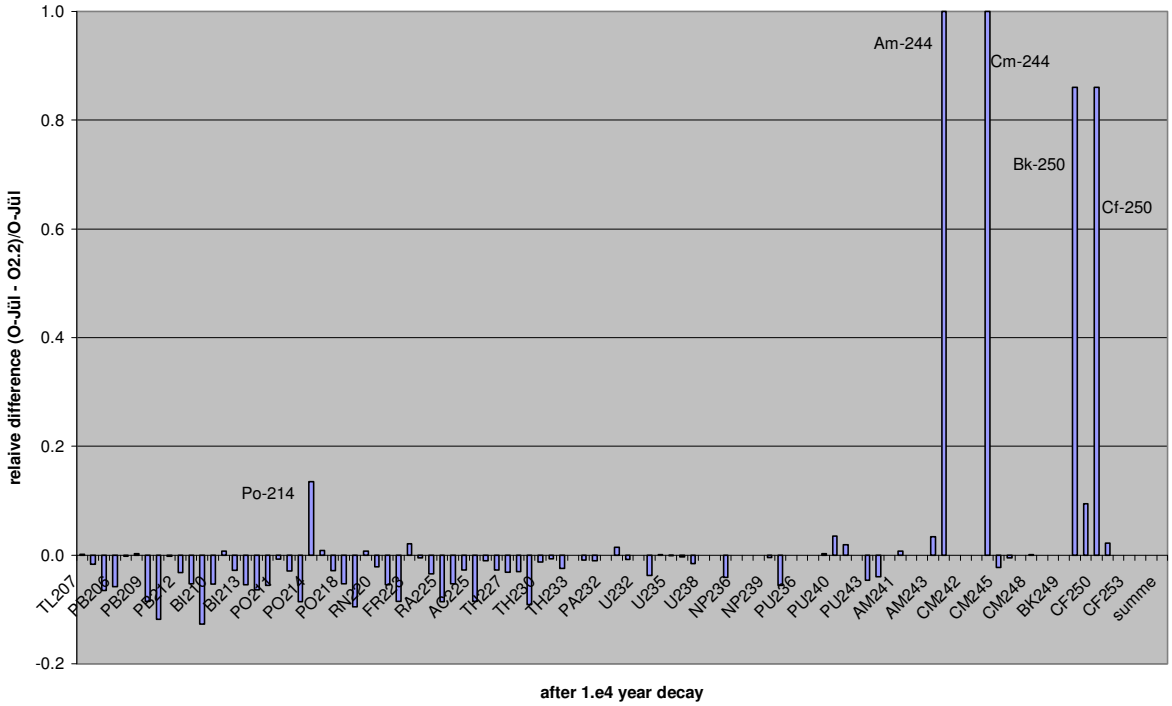

Fig. 15: Relative isotope mass difference after 10^4 year decay

For the final decay time period of 10^6 years the trend again is vanished, the main difference can be found for Po-214 (20 %) (Fig. 16). The differences of the isotopes Am-244, Cm-244, Cm-246 and Cf-250 are influenced by numerical effects or small differences of the decay constant.

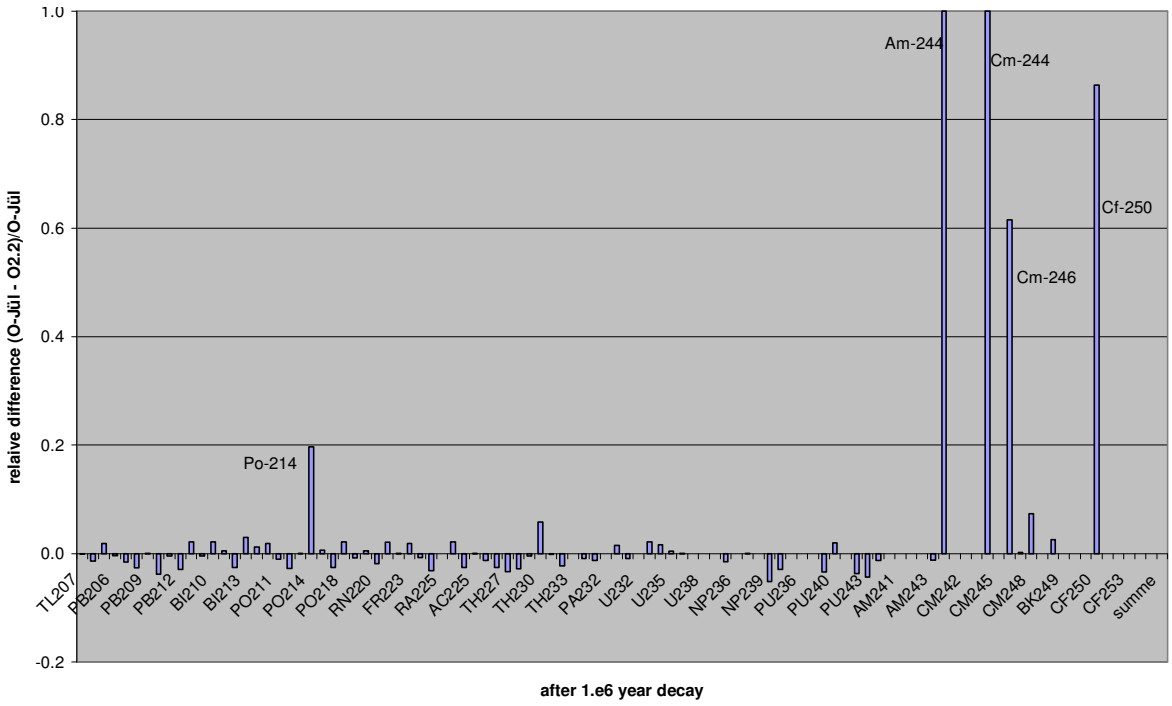

Fig. 16: Relative isotope mass difference after 10^6 year decay

3.2.2 Isotope mass differences normalized to total heavy metal mass

Normalizing the difference to the total heavy metal mass for the specific decay time period shows a even better agreement of both codes than for burned Pu-MA fuel.

After 1 year decay time the differences are negligible (< 2.e6) and even smaller than for burned Pu-MA fuel (Fig. 17). The highest difference occur for Pu-241 (1.8e-6) and Am-241 (-1.7e-6) which again indicates a larger decay constant for Pu-241 in ORIGEN 2.2.

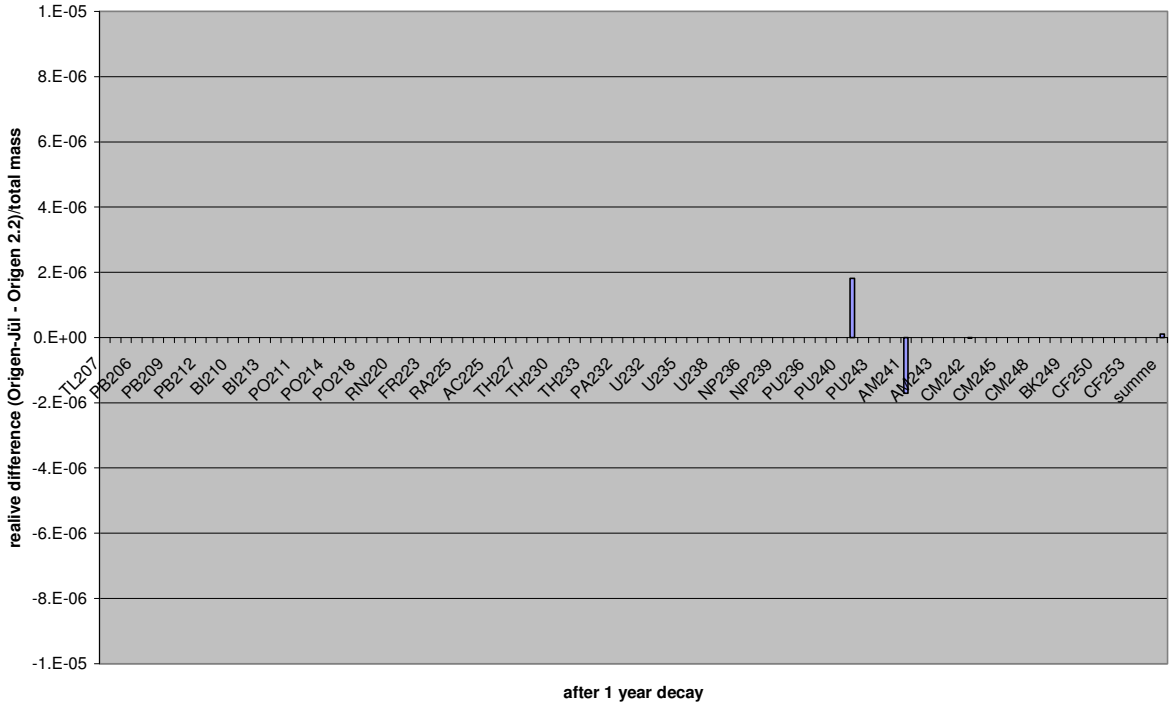

Fig. 17: Isotope mass difference after 1 year decay, normalized to total heavy metal mass

For a decay time period of 100 years again the largest differences can be observed for U-234 (- 3.e-6), Np-237 (- 2.e-6), Pu-238 (3.e-6), Pu-240 (4.e-6) and Pu-241 (2.e-6) (Fig. 18). The values of the differences for the mother–daughter connections Pu-238 → U-234 and Pu-241 → Np-237 are in very good agreement. The isotope Pu-240 shows a difference which is not consistent with a appropriate difference of the main mother nuclei Cm-244. Therefore the difference probably is induced by a difference in the branching ratio of both codes.

After 10^4 years of decay Pu-240 has the largest difference (< 5.5e-5, Fig. 19, see also Fig. 11). The corresponding mother nuclei Cm-244 doesn't show such large difference while the daughter nuclei U-236 (- 5.5e-5) nearly exactly meet this difference. This is a strong indication that ORIGEN 2.2 is using a larger decay constant for Pu-240 than ORIGEN-Jül. The reason why this correlation is not clearly observable for the decay time period of 100 years is the significant longer half life of 6563 years. Therefore after 100 years only a small fraction of Pu-240 is decayed to U-236 and their actual amount is still dominated by the initial mass isotope composition. Also U-235 (- 1.8e-5) and Pu-239 (1.1e-5) show larger differences which indicate a lager decay constant for Pu-239 used in ORIGEN 2.2.

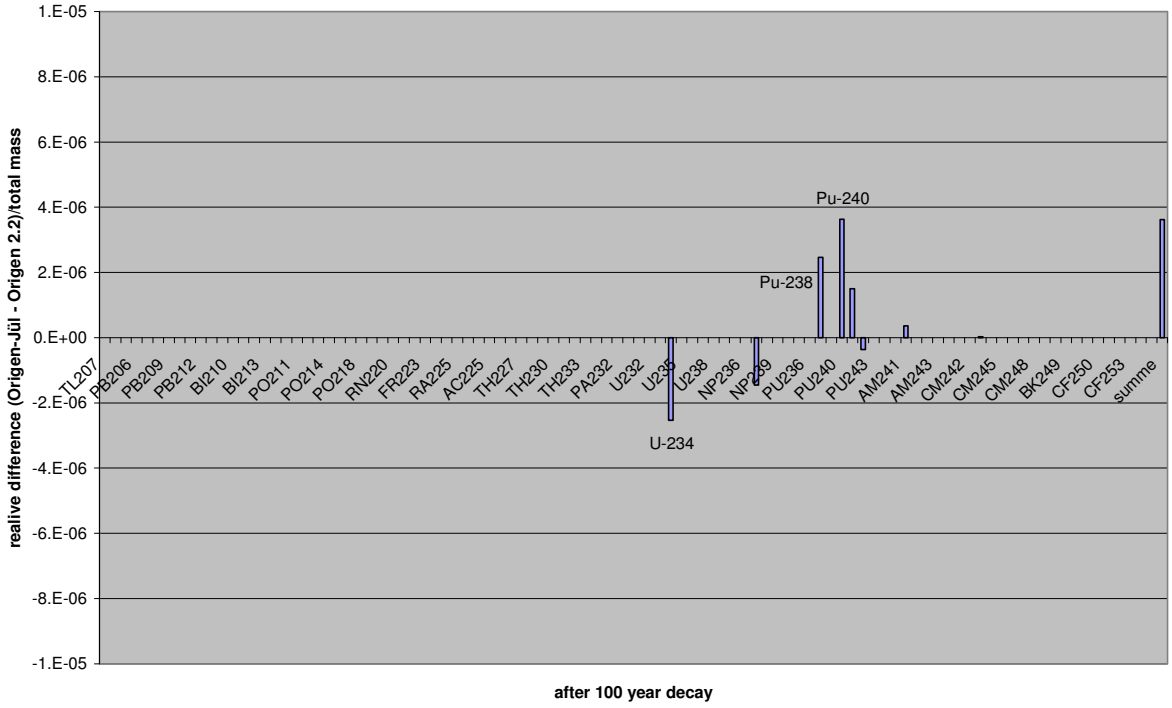

Fig. 18: Isotope mass difference after 100 year decay, normalized to total heavy metal mass

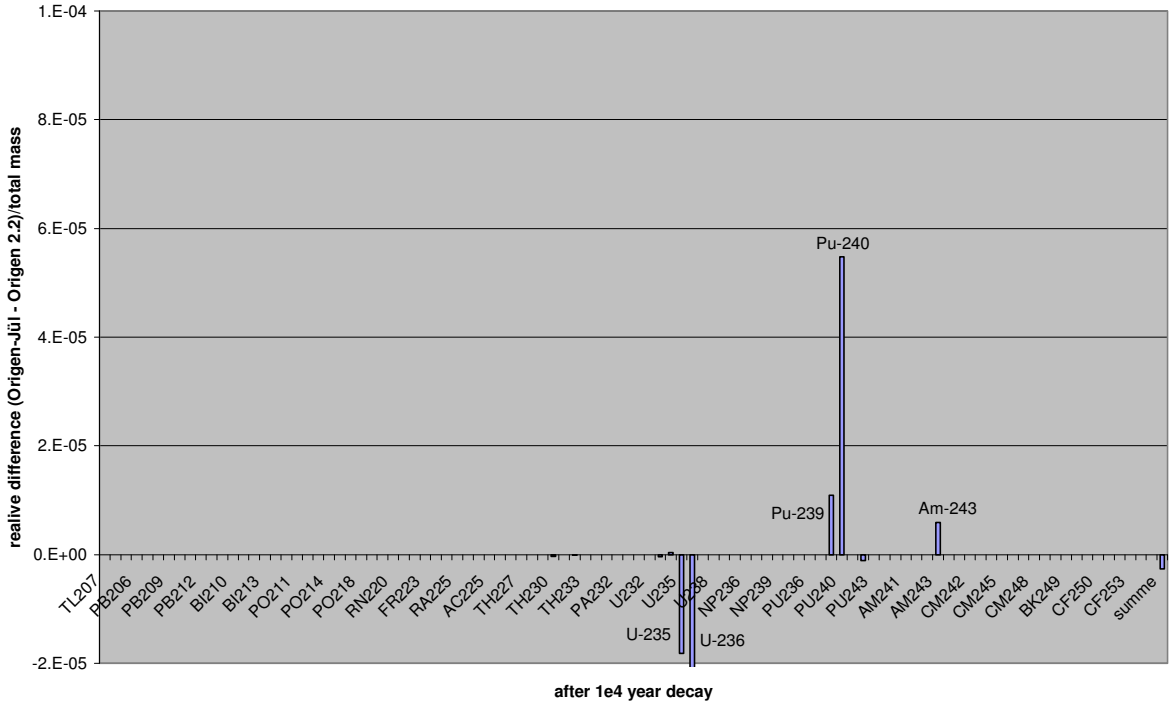

Fig. 19: Isotope mass difference after 10^4 year decay, normalized to total heavy metal mass

After the longest calculated decay time period of 10^6 years the main differences occur for U-235 (7.6e-5, positive in contrast to time period 10^4) and the total heavy metal mass (6.1e-5) (Fig. 20). Neither the mother nuclei Pu-239 nor the next relevant daughter nuclei Pa-231 (U-235 → Th-231 →) show such a large difference. The difference of Pa-231 could be masked by the relative fast decay of Pu-231 ($\tau_{1/2}$ = 3.276e+4 years) and their daughter nuclei compared to the considered decay time period but also for the final stable isotope Pb-207 no corresponding difference can be observed. Therefore the probably reason for the difference of

U-235 is a larger decay constant and a higher spontaneous fission fraction used by ORIGEN 2.2 keeping the α decay constant (Tab. 6). This would be consistent with the difference in the total heavy metal mass (the missing mass then should be found at the mass of the fission products). The production of new U-235 via decay of Pu-239 has only a small effect because of the long time period. Significant differences can also be observed for Pu-242 (- 1.7 e-5), U-236 (1.1e-5) and Th-232 (1.2e-5). The similar difference of the mother-daughter nuclei connection U-236 → Th-232 indicates a larger decay constant for U-236 used in ORIGEN 2.2. The correlated difference to the daughter nuclei of Pu-242 → U-238 as seen for burned Pu-MA fuel may be is superimposed by the large mass fraction of U-238 in the considered burned fuel composition. Because of the almost complete decay of Pu-240 its large difference does not exist anymore.

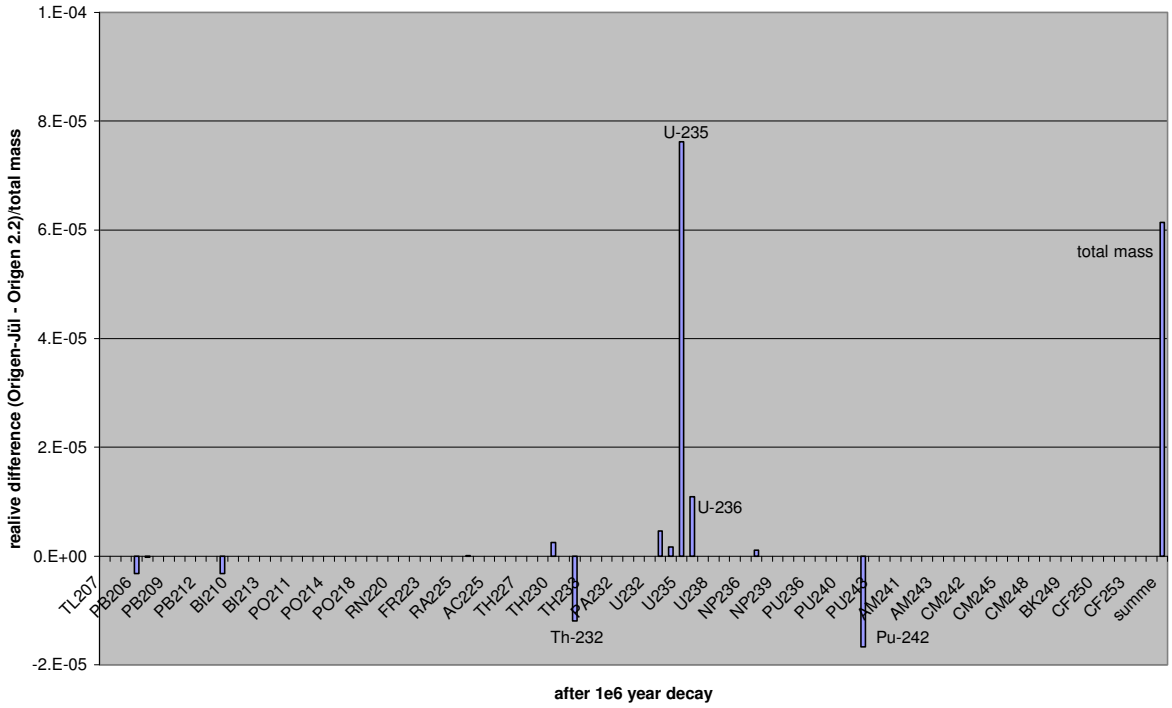

Fig. 20: Isotope mass difference after 10^6 year decay, normalized to total heavy metal mass

4. Comparison of decay constants for mass numbers higher 205

As shown by the comparison of pure decay calculations by ORIGEN-Jül and ORIGEN 2.2 (chapter 2 and 3) the differences of the decay constants are large enough to significantly influence the isotope masses after certain decay time periods. The relative differences of the decay constants

$$\Delta\lambda = (\lambda_{\text{ORIGEN-Jül}} - \lambda_{\text{ORIGEN 2.2}})/\lambda_{\text{ORIGEN-Jül}}$$

for all considered isotopes with mass numbers higher than 205 (actinides and their daughter nuclei) are given in Fig. 21/22 (numbers are given in Appendix C). The largest differences can be observed for Np-236 and Am-244. For Np-236 the difference result from the fact that ORIGEN-Jül use the decay constant for the exited isotope while ORIGEN 2.2 use the decay constant of the isotope at ground state which have a significant lower decay constant (7 orders of magnitude). For the exited isotope Np-236m ORIGEN 2.2 has a separate decay constant. Also for Am-244 ORIGEN 2.2 use separate decay constants for the ground state and the exited isotope while ORIGEN-Jül use the decay constant of the exited isotope.

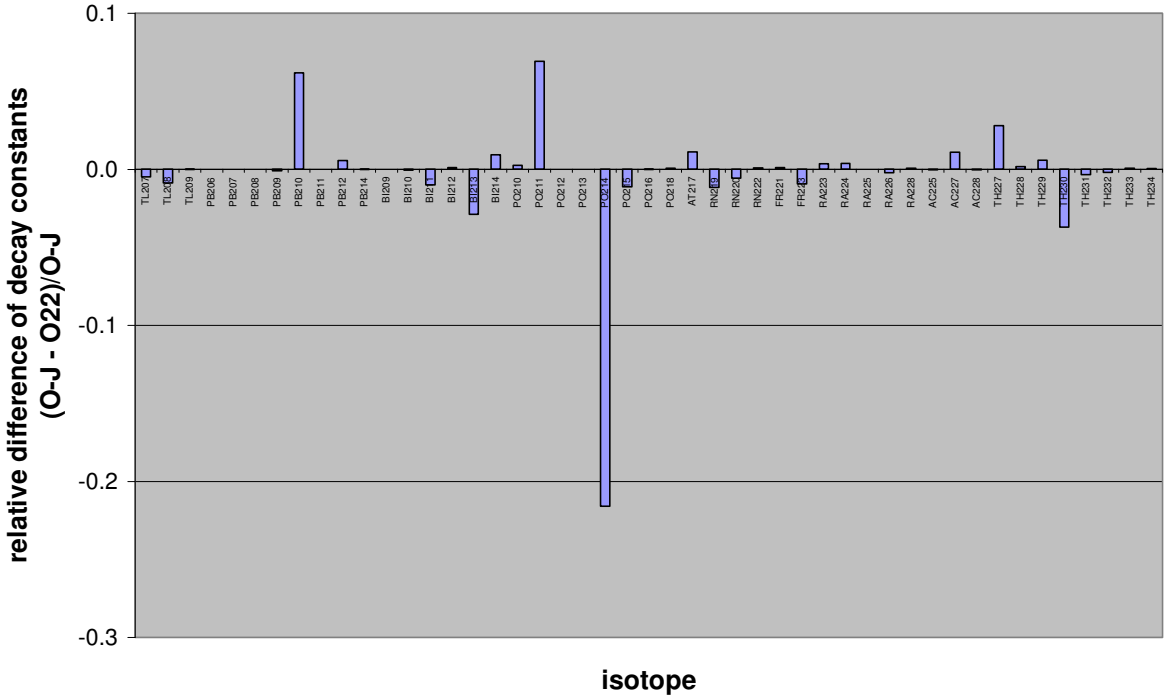

Fig. 21: Differences of the decay constants for the elements thallium (Tl) to thorium (Th), normalized to the decay constant of ORIGEN-Jül

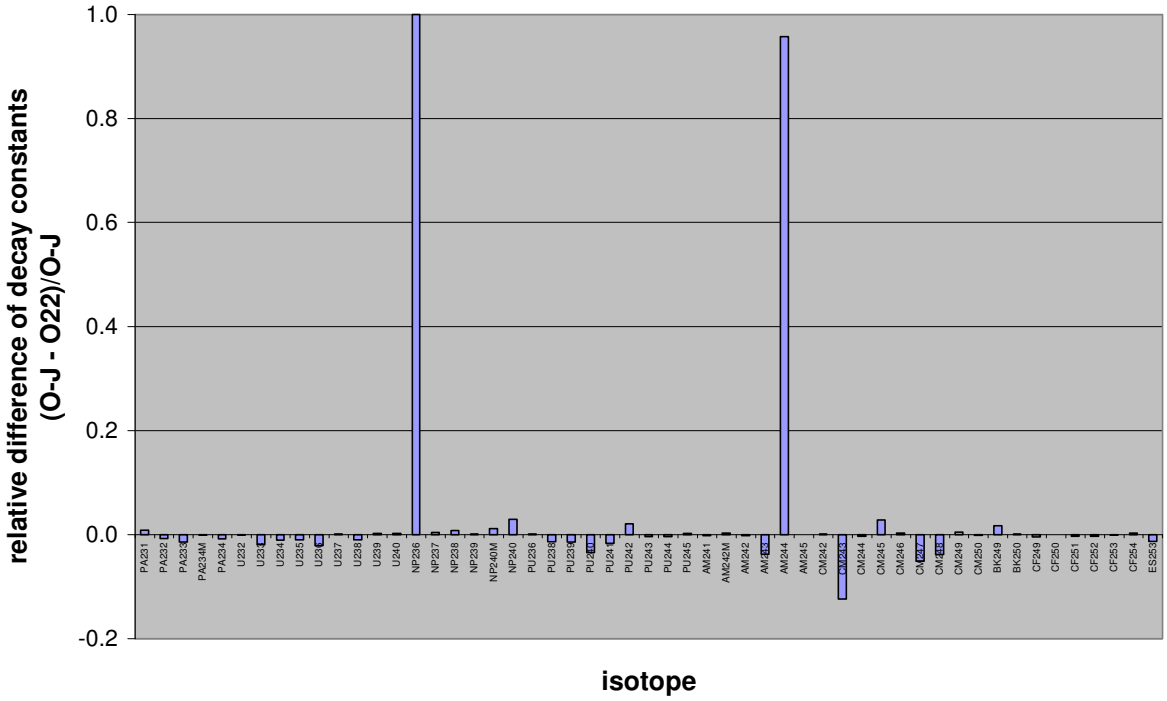

Fig. 22: Differences of the decay constants for the elements protactinium (Pa) to einsteinium (Es), normalized to the decay constant of ORIGEN-Jül

As mentioned the calculations of ORIGEN 2.2 use a larger decay constant then ORIGEN-Jül for Po-214 (22 %) and Cm-243 (12 %). The differences of the decay constants of the other isotopes are below ± 7 %. With respect to the decay calculations of the burned fuel compositions for selected isotopes the proposed difference of the decay constants can be found in Tab. 3.

Isotope	relative difference $\Delta\lambda = (\lambda_{ORIGEN\text{-}Jül} - \lambda_{ORIGEN\ 2.2})/ \lambda_{ORIGEN\text{-}Jül}$
U-235	-0.008
U-236	-0.020
Pu-238	-0.014
Pu-239	-0.013
Pu-240	-0.033
Pu-241	-0.013
Pu-242	+0.021
Am-241	-0.001
Am-243	-0.036
Cm-245	+0.028

Tab. 3: Relative differences of decay constants for selected isotopes

Some additional isotopes can be found in the ORIGEN 2.2 data library (see Appendix B). But only Pa-234 is generated by these decay calculations and only with a very small fraction compared to the total heavy metal mass (< 2.1e-16) for both burned and decayed fuel compositions (Pu-MA (10^6 years): 5.5 kg, LEU (10^6 years): 27.6 kg). Another more artificial isotope is used in ORIGEN 2.2: Sf-250 which seems to reflect the amount of stable isotopes generated by spontaneous fission.

5. Comparison of branching rations for selected isotopes

The branching ratios can be found in *decay.lib* for ORIGEN 2.2 and *orglib992.lib* for ORIGEN-Jül. For the branching ratio of α decay the corresponding variable is FA (value 8) and for the branching ratio of spontaneous fission FSF (decay.lib: value 2 in the second line, orglib992.lib: value 9) /6/. The β decay fraction FB then is calculated by

$$FB = 1.0 – FP – FA –FT –FSF$$

(FP: fraction of positron emission, FT: fraction for exited-to-ground state)

While the precision of the decay fractions are limited to 3 digits for isotopes dominated by one decay fraction the corresponding value is rounded to 1.000. Nevertheless for other relevant decay fractions numbers can be found. For selected isotopes the branching ratios for α decay and spontaneous fission are given in Tab. 4.

		ORIGEN 2.2 (decay.lib)		ORIGEN-Jül (orglib992.lib)	
isotope	id	FA (α decay)	FSF (sf)	FA (a decay)	FSF (sf)
Po-214	842140	1.000	0.000	1.000	0.000
Th-230	902300	1.000	5.000e-13	1.000	0.000
Np-237	932370	1.000	4.400e-12	1.000	0.000
Pu-238	942380	1.000	1.840e-9	1.000	1.760e-9
Pu-239	942390	1.000	4.400e-12	1.000	0.000
Pu-240	942400	1.000	5.000e-8	1.000	4.910e-8
Pu-241	942410	2.450 e-5	0.000	2.400 e-5	0.000
Pu-242	942420	1.000	5.550e-6	1.000	5.340e-6
Am-241	952410	1.000	4.100 e-12	1.000	0.000
Am-243	952430	1.000	1.800 e-10	1.000	0.000
Cm-244	962440	1.000	1.346e-6	1.000	1.340e-6
Cm-245	962450	1.000	0.000	1.000	0.000
Cm-246	962460	0.9997	2.610e-4	0.9997	2.620e-4
Cm-247	962470	1.000	0.000	1.000	0.000
Bk-250	972500	0.000	0.000	0.000	0.000
Cf-249	982490	1.000	5.000e-9	1.000	0.000
Cf-250	982500	9.992e-1	7.700e-4	1.000	0.000

Tab. 5: Branching ratios (α decay, sf) for selected isotopes in ORIGEN 2.2 and ORIGEN-Jül

As proposed in the comparison of the decay calculation for Cm-245 the branching ratios used in ORIGEN 2.2 and ORIGEN-Jül are consistent. Differences can be found for several isotopes in particular for small fractions of spontaneous fission. For some isotopes even ORIGEN 2.2 use very small fractions for spontaneous fission while for ORIGEN-Jül the fraction for spontaneous fission is zero.

As mentioned in the previous chapters for some isotopes larger differences could be observed, typically far beyond their half live. Beside the fact that these isotopes are almost completely decayed the reason for these differences should be clarified. Possible reasons could be differences in the method of solving the coupled differential equations, numerical effects because of handling very small values or very small differences in the nuclear data (decay constant and branching ratios). The last point can be verified by using equivalent nuclear data for the main decay series in ORIGEN-Jül and ORIGEN 2.2. Any residual isotope mass difference then will be caused by the first two reasons mentioned. The nuclear data of the following decay series equalised for the decay calculations:

- Cf-249 → (α,sf) → Cm-245 / FP → (α) → Pu-241 → (α) → Am-241 → (α,sf) → Np-237
- Cm-242 → (α,sf) → Pu-238 / FP
- Cm-243 → (α, β^-) → Pu-239
- Cm-244 → (α,sf) → Pu-240 / FP
- Cm-246 → (α,sf) → Pu-242 / FP
- Cm-247 → (α → Pu-243 → (β^-)) → Am-243

The half life (decay.lib: value 4, orglib992.lib: value 2 (character 8 to 16), unit of the decay constant: value 3 (character 17)) and the branching ratios for α decay and spontaneous fission of the ORIGEN-Jül library are used for an equivalent ORIGEN 2.2 calculation (Tab. 6). The values for half live of ORIGEN-Jül are modified to the unit second and than used for the

ORIGEN 2.2 calculation. The half life used in ORIGEN 2.2 is given in the second column with its relative difference to the ORIGEN-Jül data. If different from the ORIGEN-Jül values the branching ratios of ORIGEN 2.2 are given in brackets in the corresponding column.

isotope	half life ORIGEN-Jül data	half life (s) (rel. diff. to O-Jül) ORIGEN 2.2 data	α decay fraction FA	spontaneous fission fraction FSF
Cf-249	1.110e10 s =352 a	1.106e10 (0.36 %)	1.000	0.000 (5.000e-9)
Cm-247	5.172e14 s = 16.4e6 a	4.923e14 (4.81 %)	1.000	0.000
Cm-246	1.486e11 s = 4.711e3 a	1.493e11 (-0.47 %)	0.9997	2.620e-4 (2.61e-4)
Cm-245	2.606e11 s = 8265 a	2.682e11 (-2.92 %)	1.000	0.000
Cm-244	5.708e8 s = 18.1 a	5.715e8 (-0.12 %)	1.000	1.340e-6 (1.346e-6)
Cm-243	1.009e9 s = 32 a	8.994e8 (10.86 %)	1.000 (0.9976)	0.000
Cm-242	1.408e7 s = 163 d	1.410e7 (-0.14 %)	1.000	6.18e-8 (6.80e-8)
Am-243	2.413e11 s = 7650 a	2.329e11 (3.48 %)	1.000	0.000 (1.8e-10)
Am-241	1.366e10 s = 433.0 a	1.364e10 (0.15 %)	1.000	0.000 (4.100e-12)
Pu-242	1.195e13 s = 3.79e7 a	1.221e13 (-2.18 %)	1.000	5.34e-6 (5.55e-6)
Pu-241	4.604e8 s = 14.6 a	4.544e8 (1.30 %)	2.400e-5 (2.450e-5)	0.000
Pu-239	7.695e11 s = 2.44e4 a	7.594e11 (1.31 %)	1.000	0.000 (4.4e-12)
Pu-238	2.807e09 s = 89 a	2.769e9 (1.35 %)	1.000	1.76e-9 (1.84e-9)
U-235	2.239e16 s = 7.10e8 a	2.221e16 (0.8 %)	1.000	0.000 (2.60e-9)
Np-237	6.749e13 s = 2.14e6 a	6.753e13 (-0.06 %)	1.000	0.000

Tab. 6: ORIGEN-Jül nuclear data used for a ORIGEN 2.2 calculation for selected isotopes (if different branching ratios of ORIGEN 2.2 are given in brackets)

Calculating the decay with ORIGEN 2.2 based on half life and branching ratios of ORIGEN-Jül a much better agreement and therefore significant lower differences can be observed (Fig. 23), except for Am-243 at 10^6 years where the difference is increased from -1.2 % to -4.9 %. Also the small negative difference maximum of Np-237 at 10^3 years is not affected by the nuclear data exchange. The relative differences of the half life for the main isotopes are lower than 3.5 %. Only for Cm-237 (4.8 %) and Cm-243 (10.9 %) the differences are larger.

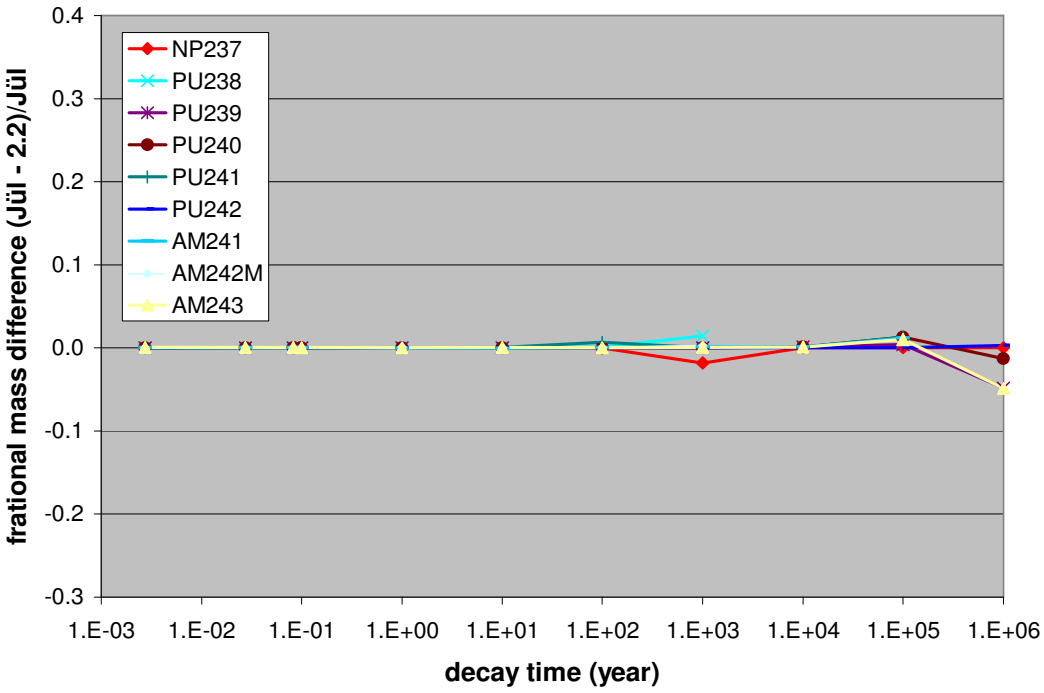

Fig. 23: Differences of isotopes masses by using ORIGEN-Jül nuclear data for the ORIGEN 2.2 calculation

This comparison indicates that the initially observed larger differences are mainly caused by differences of the nuclear data and not by numerical effects. In particular small differences of the half life result in larger mass differences up to 20 % when calculating the decay for time periods far beyond their half life.

The observed isotope mass differences of Am-241 and Pu-241 beyond 10^4 years were investigated separately by exchanging the nuclear data of ORIGEN 2.2 only for these isotopes by ORIGEN-Jül data.

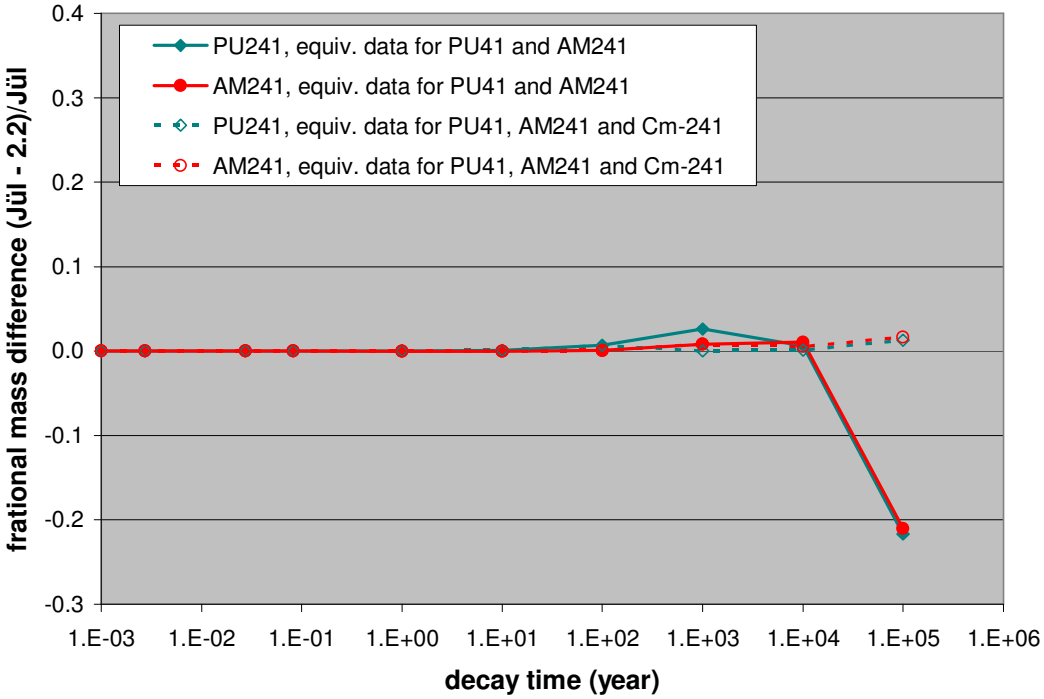

Fig. 24: Differences of isotopes masses by using ORIGEN-Jül nuclear data for the ORIGEN 2.2 calculation for the isotopes Cm-245, Pu-241 and Am-241

While the differences stay nearly unchanged when using only the data of Pu-241 and Am-241 the differences for the whole considered decay time period nearly vanish when using the data of Cm-245 (Fig. 24). While the decay constant of Cm-245 in ORIGEN 2.2 is smaller than in ORIGEN-Jül which is equivalent to a higher residual amount of this mother isotope, this comparison indicate that the decay calculation of Cm-245 however is responsible for the higher residual mass of Pu-241 and Am-241 beyond 10^4 years. The reason for this effect should be analysed in more detail.

6. Conclusion:

Calculations done by ORIGEN 2.2 to determine the residual mass of burned fuel compositions after different decay time periods generally are in good agreement with calculations done by ORIGEN-Jül. This was demonstrated for two different isotopes compositions. For the considered burned fuel compositions (LEU, Pu+MA) decay calculations for isotopes with mass numbers > 205 were done by ORIGEN-Jül and ORIGEN 2.2 and the relative differences of the isotope masses were compared.

The differences are specific for the individual isotopes - no general trend can be observed. While the comparisons show differences for different decay time periods for some isotopes their contribution is rather small (< 0.7 %) compared to the total heavy metal mass. Highest relative differences are observed for isotopes nearly completely decayed (mass reduction ≈ 10^6) in the considered burned fuel compositions and for time periods far beyond their half life. The largest contribution to these large differences is caused by small differences of the decay constants (mainly lower than 3.5 %) which increase the mass difference in particular for time periods far beyond the individual half live (> 10 $\tau_{1/2}$).

When normalizing the difference to the actual residual heavy metal mass isotopes can be identified which are dominate for the assessment of radioactivity. Isotopes with very small mass fractions will be neglected for this time and for longer decay time periods. Corresponding differences of mother and daughter nuclei observed with this normalization indicate different decay constants in ORIGEN-Jül and ORIGEN 2.2 (Pu-238 → U-234, Pu-239 → U-235, Pu-240 → U-236, Pu-242 → U-238). The difference is calculated as:

$$\Delta = (m_{isotope}(O\text{-}J) - m_{isotope}(O2.2))/m_{total}(O\text{-}J)$$

So a positive difference correspond to a smaller residual isotope mass calculated by ORIGEN 2.2. And this is equivalent to a faster decay or a smaller production of this isotope by ORIGEN 2.2.

For both burned fuel compositions consistency between the kind of differences and the decay constants can be observed for selected isotopes:

- different branching ratios (α, sf) for Cm-245
- a larger decay constant (faster decay) of Pu-238 used in ORIGEN 2.2 (significant for Pu-MA fuel)
- larger decay constants for Pu-239, Pu-240, Pu-241 and Am-243 used in ORIGEN 2.2
- a larger decay constant for Pu-241 used in ORIGEN 2.2 which explain the difference for a decay period < 10^4 years
- a smaller decay constant for Pu-242 used in ORIGEN 2.2 while the fractions of decay are constant
- a larger decay constant and spontaneous fission fraction for U-235 used in ORIGEN 2.2, the negative difference observed for U-235 for a time period of 10^4 years is caused by a higher production via decay of Pu-239 compared to the decay of U-235

The observed differences of the isotope masses after certain decay time periods are verified by differences of the decay constants of ORIGEN-Jül and ORIGEN 2.2 except for Am-241 and Pu-241. The higher amount of Pu-241 and Am-241 calculated by ORIGEN 2.2 for time periods beyond 10^4 years seems not to correspond with the also higher amount of Cm-245. A higher residual amount of Cm-245 combined with a smaller decay constant as used by ORIGEN 2.2 usually is correlated with a lower mass of the daughter nuclei. But anyhow by using the same decay constant of Cm-245 for ORIGEN-Jül and ORIGEN 2.2 the differences

for Pu-241 and Am-241 can be minimised. One reason for this behaviour could the superposition of the decay of the initial Pu-241 and Am-241 fraction with the fraction generated by the decay of Cm-245. This effect should be analysed in more detail.

Using the ORIGEN-Jül nuclear data for the decay calculation with ORIGEN 2.2 reduce the isotope mass differences significantly. This demonstrates that the observed mass differences are mainly induced by differences in the nuclear data. Numerical effects based on differences in the method of solving the coupled differential equations or handling of very small values if existing only have a small influence.

It can be concluded that decay calculations for the dominating isotopes of the considered isotope compositions done by ORIGEN-Jül and ORIGEN 2.2 result in very good agreement (-0.1 % $\leftrightarrow$ 0.7 %). With respect to the difficulty of calculating decay series for individual isotopes with ORIGEN-Jül it is reliable to apply ORIGEN 2.2 for these calculations.

Nevertheless a library update of ORIGEN-Jül seems to be preferable to have a consistent data set with the neutronic diffusion and transport calculations.

References:

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Appendix A:

Mass (gram) of isotopes with mass number higher than 205 of the discharged fuel for an initial plutonium/minor actinide fuel composition.

isotope	mass (g)	isotope	mass (g)	isotope	mass (g)	isotope	mass (g)
TL207	1.92E-19	AT217	4.43E-21	PA234M	2.68E-12	AM241	2.38E+02
TL208	2.59E-13	RN219	2.83E-21	U232	7.30E-05	AM242M	1.04E+01
TL209	3.87E-19	RN220	2.30E-13	U233	2.55E-04	AM242	1.30E-01
PB206	2.54E-18	RN222	7.28E-17	U234	2.49E+00	AM243	1.93E+02
PB207	3.24E-15	FR221	4.06E-17	U235	2.17E-01	AM244	1.78E-03
PB208	2.76E-08	FR223	1.95E-20	U236	1.64E-01	AM245	6.98E-17
PB209	1.58E-15	RA223	7.09E-16	U237	1.64E-04	CM242	5.24E+01
PB210	1.33E-15	RA224	1.32E-09	U238	7.43E-04	CM243	9.34E-01
PB211	1.48E-18	RA225	1.56E-13	U239	1.00E-09	CM244	6.56E+01
PB212	1.51E-10	RA226	1.18E-11	U240	7.84E-23	CM245	3.92E+00
PB214	3.42E-19	RA228	5.45E-20	NP236	1.12E-06	CM246	1.50E-01
BI209	8.73E-13	AC225	1.24E-13	NP237	7.35E+02	CM247	4.77E-04
BI210	8.11E-19	AC227	7.33E-13	NP238	4.37E-01	CM248	6.71E-06
BI211	8.79E-20	AC228	5.69E-24	NP239	1.60E-04	CM249	7.31E-12
BI212	1.43E-11	TH227	1.32E-15	NP240M	7.24E-25	CM250	2.75E-15
BI213	3.83E-16	TH228	2.65E-07	NP240	1.15E-09	BK249	1.72E-08
BI214	2.51E-19	TH229	2.96E-09	PU236	4.69E-04	BK250	1.91E-12
PO210	6.95E-18	TH230	3.60E-06	PU238	3.95E+02	CF249	2.41E-09
PO211	1.06E-24	TH231	5.01E-10	PU239	1.38E+03	CF250	1.14E-09
PO212	7.57E-22	TH232	2.91E-09	PU240	1.23E+03	CF251	1.98E-10
PO213	5.58E-25	TH233	1.33E-15	PU241	8.10E+02	CF252	2.21E-11
PO214	4.25E-26	TH234	9.87E-15	PU242	3.77E+02	CF253	6.98E-15
PO215	1.25E-24	PA231	1.18E-07	PU243	2.02E-02	CF254	2.26E-18
PO216	6.05E-16	PA232	2.13E-11	PU244	2.59E-12	ES253	6.88E-15
PO218	3.96E-20	PA233	2.54E-05	PU245	8.38E-19	sum	5.49E+03

Mass (gram) of isotopes with mass number higher than 205 of the discharged fuel for an initial LEU fuel composition.

isotope	mass (g)	isotope	mass (g)	isotope	mass (g)	isotope	mass (g)
TL207	1.49E-16	AT217	2.56E-21	PA234M	1.28E-11	AM241	1.69E+00
TL208	4.83E-14	RN219	2.20E-18	PA234	2.21E-11	AM242M	4.31E-02
TL209	2.24E-19	RN220	4.27E-14	U232	7.37E-06	AM242	1.75E-03
PB206	8.17E-15	RN222	1.02E-14	U233	4.03E-05	AM243	1.22E+01
PB207	9.55E-12	FR221	2.35E-17	U234	1.29E+01	AM244	6.42E-05
PB208	1.26E-08	FR223	1.14E-17	U235	3.79E+02	AM245	3.37E-17
PB209	9.15E-16	RA223	5.52E-13	U236	4.02E+02	CM242	1.01E+00
PB210	6.38E-13	RA224	2.44E-10	U237	1.03E-01	CM243	2.24E-02
PB211	1.15E-15	RA225	8.10E-14	U238	2.63E+04	CM244	3.32E+00
PB212	2.80E-11	RA226	1.61E-09	U239	5.99E-03	CM245	9.77E-02
PB214	4.80E-17	RA228	1.14E-15	U240	3.68E-23	CM246	1.95E-02
BI209	2.36E-12	AC225	7.16E-14	NP236	1.57E-08	CM247	7.64E-05
BI210	4.09E-16	AC227	4.27E-10	NP237	2.91E+01	CM248	2.89E-06
BI211	6.84E-17	AC228	1.19E-19	NP238	2.08E-02	CM249	1.53E-12
BI212	2.67E-12	TH227	9.17E-13	NP239	7.92E-01	CM250	1.03E-14
BI213	2.21E-16	TH228	4.84E-08	NP240M	3.17E-25	BK249	8.25E-09
BI214	3.53E-17	TH229	4.98E-09	NP240	4.18E-06	BK250	1.56E-12
PO210	6.94E-15	TH230	1.15E-04	PU236	1.12E-05	CF249	1.70E-09
PO211	8.27E-22	TH231	1.03E-08	PU238	1.20E+01	CF250	2.52E-09
PO212	1.41E-22	TH232	2.25E-05	PU239	1.44E+02	CF251	6.70E-10
PO213	3.23E-25	TH233	5.74E-12	PU240	1.16E+02	CF252	5.34E-10
PO214	5.98E-24	TH234	3.79E-07	PU241	7.10E+01	CF253	2.75E-13
PO215	9.72E-22	PA231	1.65E-05	PU242	7.74E+01	CF254	9.50E-16
PO216	1.12E-16	PA232	4.50E-09	PU243	2.22E-03	ES253	4.85E-13
PO218	5.57E-18	PA233	9.93E-07	PU244	1.93E-12	summe	2.76E+04

Appendix B:

Additional isotopes of the ORIGEN 2.2 data library beyond data library of ORIGEN-Jül

TL206	TH226	NP236M	AM244M	ES254M
BI208	PA235	NP241	AM246	ES254
BI210M	U230	PU237	CM241	ES255
PO211M	U231	PU246	CM251	(SF250)
RN218	U241	AM239	BK251	
RA222	NP235	AM240	CF255	

Appendix C:

Decay constants used in ORIGEN-Jül and ORIGEN 2.2 for isotopes with mass number higher than 205 (actinides and their daughter nuclei)

NUCLIDE	$\lambda_{ORIGEN\text{-}Jül}$ (1/s)	$\lambda_{ORIGEN\ 2.2}$ (1/s)	relative difference $\Delta\lambda = (\lambda_{ORIGEN\text{-}Jül} - \lambda_{ORIGEN\ 2.2})/ \lambda_{ORIGEN\text{-}Jül}$
TL207	2.41E-03	2.42E-03	-4.98E-03
TL208	3.73E-03	3.76E-03	-8.85E-03
TL209	5.25E-03	5.25E-03	-1.90E-04
PB206	0.00E+00	0.00E+00	0.00E+00
PB207	0.00E+00	0.00E+00	0.00E+00
PB208	0.00E+00	0.00E+00	0.00E+00
PB209	5.83E-05	5.84E-05	-8.58E-04
PB210	1.05E-09	9.85E-10	6.19E-02
PB211	3.20E-04	3.20E-04	0.00E+00
PB212	1.82E-05	1.81E-05	5.49E-03
PB214	4.31E-04	4.31E-04	-2.32E-04
BI209	0.00E+00	0.00E+00	0.00E+00
BI210	1.60E-06	1.60E-06	-6.25E-04
BI211	5.37E-03	5.42E-03	-1.01E-02
BI212	1.91E-04	1.91E-04	1.05E-03
BI213	2.46E-04	2.53E-04	-2.89E-02
BI214	5.86E-04	5.81E-04	9.39E-03
PO210	5.81E-08	5.80E-08	2.41E-03
PO211	1.33E+00	1.24E+00	6.92E-02
PO212	2.31E+06	2.31E+06	0.00E+00
PO213	1.65E+05	1.65E+05	0.00E+00
PO214	3.47E+03	4.22E+03	-2.16E-01
PO215	3.85E+02	3.89E+02	-1.14E-02
PO216	4.62E+00	4.62E+00	-2.16E-04
PO218	3.79E-03	3.79E-03	5.28E-04
AT217	2.17E+01	2.15E+01	1.11E-02
RN219	1.73E-01	1.75E-01	-1.16E-02
RN220	1.24E-02	1.25E-02	-5.65E-03
RN222	2.10E-06	2.10E-06	9.52E-04
FR221	2.41E-03	2.41E-03	1.24E-03

FR223	5.25E-04	5.30E-04	-9.33E-03
RA223	7.04E-07	7.02E-07	3.41E-03
RA224	2.20E-06	2.19E-06	3.64E-03
RA225	5.42E-07	5.42E-07	1.85E-04
RA226	1.37E-11	1.37E-11	-2.19E-03
RA228	3.28E-09	3.28E-09	6.10E-04
AC225	8.02E-07	8.02E-07	-3.74E-04
AC227	1.02E-09	1.01E-09	1.08E-02
AC228	3.14E-05	3.14E-05	-3.18E-04
TH227	4.41E-07	4.29E-07	2.79E-02
TH228	1.15E-08	1.15E-08	1.74E-03
TH229	3.01E-12	2.99E-12	5.65E-03
TH230	2.75E-13	2.85E-13	-3.71E-02
TH231	7.52E-06	7.55E-06	-3.32E-03
TH232	1.56E-18	1.56E-18	-1.92E-03
TH233	5.23E-04	5.23E-04	5.74E-04
TH234	3.33E-07	3.33E-07	3.00E-04
PA231	6.76E-13	6.70E-13	8.28E-03
PA232	6.08E-06	6.12E-06	-7.07E-03
PA233	2.93E-07	2.97E-07	-1.40E-02
PA234M	9.87E-03	9.87E-03	-4.05E-04
PA234	2.85E-05	2.87E-05	-8.42E-03
U232	3.05E-10	3.05E-10	-3.28E-04
U233	1.36E-13	1.39E-13	-1.91E-02
U234	8.89E-14	8.98E-14	-1.05E-02
U235	3.09E-17	3.12E-17	-1.00E-02
U236	9.19E-16	9.38E-16	-2.08E-02
U237	1.19E-06	1.19E-06	8.40E-04
U238	4.87E-18	4.92E-18	-9.45E-03
U239	4.92E-04	4.91E-04	2.24E-03
U240	1.37E-05	1.37E-05	2.92E-03
NP236	8.75E-06	1.91E-13	1.00E+00
NP237	1.03E-14	1.03E-14	3.88E-03
NP238	3.82E-06	3.79E-06	7.85E-03
NP239	3.41E-06	3.41E-06	1.17E-03
NP240M	1.58E-03	1.56E-03	1.20E-02
NP240	1.83E-04	1.78E-04	2.90E-02
PU236	7.71E-09	7.70E-09	7.78E-04
PU238	2.47E-10	2.50E-10	-1.34E-02
PU239	9.00E-13	9.13E-13	-1.42E-02
PU240	3.25E-12	3.36E-12	-3.38E-02
PU241	1.50E-09	1.53E-09	-1.67E-02
PU242	5.80E-14	5.68E-14	2.12E-02
PU243	3.87E-05	3.89E-05	-3.88E-03
PU244	2.65E-16	2.66E-16	-3.40E-03
PU245	1.82E-05	1.82E-05	2.20E-03
AM241	5.07E-11	5.08E-11	-2.37E-03
AM242M	1.45E-10	1.45E-10	3.45E-03
AM242	1.20E-05	1.20E-05	-1.67E-03

AM243	2.87E-12	2.98E-12	-3.69E-02
AM244	4.44E-04	1.91E-05	9.57E-01
AM245	9.30E-05	9.30E-05	-1.08E-04
CM242	4.92E-08	4.92E-08	8.13E-04
CM243	6.86E-10	7.71E-10	-1.23E-01
CM244	1.21E-09	1.21E-09	-2.48E-03
CM245	2.66E-12	2.58E-12	2.86E-02
CM246	4.66E-12	4.64E-12	3.65E-03
CM247	1.34E-15	1.41E-15	-5.07E-02
CM248	6.24E-14	6.48E-14	-3.81E-02
CM249	1.81E-04	1.80E-04	4.97E-03
CM250	1.26E-12	1.26E-12	-1.59E-03
BK249	2.55E-08	2.51E-08	1.69E-02
BK250	5.98E-05	5.98E-05	8.36E-04
CF249	6.24E-11	6.27E-11	-4.33E-03
CF250	1.68E-09	1.68E-09	5.95E-04
CF251	2.44E-11	2.45E-11	-2.46E-03
CF252	8.30E-09	8.33E-09	-3.13E-03
CF253	4.50E-07	4.50E-07	-8.89E-04
CF254	1.33E-07	1.33E-07	3.01E-03
ES253	3.87E-07	3.92E-07	-1.27E-02

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