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ADVANCED NUCLEAR FUEL CYLCES AND NUCLEAR WASTE DISPOSAL

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ABSTRACT
Extracting transuranic elements from nuclear waste (partitioning - P) to burn them in dedicated nuclear reactors (transmutation - T) essentially holds the promise of reducing the one-million-year risk of highly radioactive nuclear waste disposal. That would solve one of the main conundrums of nuclear energy production. Here, we argue that P&T will not significantly change the safety requirements and risks of geologic disposal for spent fuel and high-level nuclear waste. We will assess the maturity of P&T technologies such as reactors, separation technologies and fuel fabrication plants. A sensitivity analysis will be presented on the time scale and effects of a P&T treatment of nuclear waste fuel cycle choices like fast reactor, molten salt reactors or accelerator driven systems. This will include an estimate of the number of required fuel cycle facilities, and the composition of the final waste stream, depending on separation and transmutation efficiency, irradiation and cooling down times and the build-up of problematic fission products with very long half-lives. We compare homogeneous P&T strategies with improved proliferation resistance and heterogeneous P&T strategies with different actinides being treated separately in a more flexible fuel cycle design.

INTRODUCTION
In geological repositories planned to date, transuranium elements (TRU) and long-lived and short-lived fission products (FP) are to be disposed of together. The requirements for the repository are therefore determined by the properties of these waste groups. The idea of partitioning and transmutation (P&T) is to separate the material groups and change their respective amounts to reduce the requirements for disposal.
Advocates of P&T point out that P&T essentially holds the promise of reducing the one-million-year risk of highly radioactive nuclear waste disposal or even eliminate the requirement for a geological repository altogether.
In this publication, we will give a short overview over the status of P&T technologies which still need to mature substantially to even begin to address the most problematic isotopes. We will then argue that P&T will not significantly change the safety requirements and risks of geologic disposal for spent fuel and high-level nuclear waste. In order to do so, we compare the results of two P&T implementation models.

P&T AND FINAL DISPOSAL
Today, geologic repositories are considered the gold standard for spent nuclear fuel (SNF) and nuclear waste disposal. The requirement on these repositories is supposed to be eased by partitioning and transmutation (P&T). In the P&T literature radiotoxicity plays the prominent role as measure to identify the most significant isotopes (Acatech 2014). Separating transuranic elements reduces the radiotoxicity of SNF significantly.
But radiation doses to humans based on radiotoxicity assume that the radioactive waste is completely ingested. Using radiotoxicity ignores all processes that happen between the geologic site and human exposure (Schmidt et al. 2013). Therefore, the standard methodology to assess radiological risks to humans from underground spent fuel repositories is a long-term safety

assessment. This includes geologic and man-made barriers, dissolution rates, radionuclide retention in the underground, (non-)existence of groundwater pathways to the biosphere, radionuclide mobility in the biosphere and its accumulation and discrimination in food chains. Long-Term safety assessments predict that residual radiation exposure to humans is mainly caused by radionuclides such as carbon-14, chlorine-36, selenium-79, tin-126, iodine-129 and radium-226 (depending on host rock formation) that are comparatively mobile in the geo- and biosphere due to their physicochemical properties and have a half-life of more than a few thousand years.
P&T instead addresses mostly plutonium and other actinides that are highly immobile under saturated and chemically reducing conditions. P&T of plutonium and other actinides will therefore not significantly reduce the potential radiation exposure of future generations from a geological repository. This has been known for decades (ORNL 1977; Croff und Blomeke 1980; OECD-NEA 2007).

STATUS OF P&T TECHNOLOGY

The technical and economic challenges and the disadvantages of P&T are well known (Croff und Blomeke 1980; National Research Council 1996). Recent overview studies on the state of research in P&T especially regarding the German situation are (GRS 2014; Acatech 2014; Oeko-Institut/ZNF 2015; Brenk 2015; Frieß 2021) and internationally e.g. (IRSN 2019; OECD/NEA 2018), that also include overviews of international developments and historical overviews of individual separation processes and reactor concepts as well as impacts on disposal options and pathways. In the following, the status for the most important P&T technologies is briefly discussed.

Partitioning

The industry standard for hydrochemical separation of uranium and plutonium from spent nuclear fuel (SNF) is the PUREX process (Plutonium-Uranium REdox eXtraction). Current research efforts focus on modifications to PUREX to also extract other actinides such as neptunium (co-extraction) (OECD/NEA 2018). For more advanced fuel fabrication homogeneous separation of minor actinides is not sufficient and actinides such as americium and curium need to be separated. These processes have been tested in the laboratory, but stability and efficiency on industrial scale remain to be demonstrated.
An alternative technology to the hydrometallurgical chemical separation processes is pyroprocessing. Possible advantages of pyroprocessing include the high chemical and radiological stability of the used solvents, the possibility of high concentrations of fissile materials, the applicability to many different fuel types and the absence of liquid high-level radioactive process waste. This must be contrasted primarily with the lack of technical maturity of the various processes, many of which have been researched for a number of decades (OECD/NEA 2018).

Fuel Fabrication

MOX fuels have been used internationally in power reactors for decades. MOX is manufactured and used on an industrial scale. MOX fuels with an increased content of minor actinides are being developed. Their fabrication is challenging and has been successful only on laboratory scale.
The presence of uranium in MOX fuel inevitably leads to plutonium production. The use of uranium free, inert matrix fuel (IMF) prevents this. IMF fuels have been under development for decades. They have not been used on industrial scale yet.

Transmutation

Current light water reactors (LWR) are not designed to burn minor actinide bearing MOX fuels. In general, the thermal neutron spectrum of LWR is inefficient for transmutation. For large scale P&T applications fast reactors (FR) are much better suited. Today FR development is part of an international effort to deploy a fourth generation of nuclear reactors around 2040-2050. Six advanced reactor systems have been selected so far by the Generation IV International Forum (Gen-IV) (GIF, 2002). These fast reactor types are being developed worldwide within the framework of the forum. All these reactor designs still need significant R&D efforts.

Only the sodium cooled fast reactor development has progressed to demonstration phase in recent decades. The French fast sodium-cooled ASTRID concept, however, has been cancelled recently (NEI, 2019).

Minor actinides affect the neutron spectrum in the reactor. Their use in the reactor is limited by the negative influence on the reactivity coefficients. An alternative concept to the use of reactors are accelerator driven systems (ADS), a hybrid between particle accelerator and nuclear reactor. An ADS will always be subcritical and is not in danger of a criticality accident. Thus, a much higher content of minor actinide in the fuel is feasible. It is controlled by the neutrons produced by high energy reactions of accelerated protons that hit a heavy metal target in the core of the reactor and spall atoms. If the accelerator is switched off, the reactor shuts down. However, also in ADS accidental release of radioactivity is still possible. The main technical challenge for ADS is reliability. Today, these reactor concepts only exist as design studies. Only Belgium has concrete plans since several decades to build an accelerator driven research reactor (MYRRHA) with substantial financial help from the European Union.

A SIMPLE MODEL FOR P&T

The most important parameters for assessing a P&T scenario are the achievable reduction of the initial waste inventory and the time required for this. These parameters are not independent of each other: rather, the higher the targeted reduction of the initial inventory, the greater is the required effort. Using a simplified model, the mutual dependencies will be briefly explained below and substantiated using representative numerical examples in Table 1.

The model is based on a standard scenario for Germany (Oeko-Institut/ZNF 2015). Germany currently phases out of nuclear energy. Therefore, reactors will only be used to transmute most efficiently but not to produce new fissile material. The standard scenario assumes the use of accelerator-driven systems (ADS) and the use of uranium-free fuel (IMF). These technologies are not available yet. The choice can be considered to be close to a best case scenario. As example we use the EFIT conceptual design with a thermal output of 400 MW (Artioli et al., 2008). The amount of TRU to be converted under a P&T scenario for the German situation was estimated to be 140 t.

In the following, the isotope-specific composition of this TRU quantity over time is not considered further. However, in a real transmutation system, the isotopic composition would change significantly over time. In particular, isotopes that are more difficult to fission in fast reactors would accumulate relative to isotopes that are easier to fission. It is not clear, whether such a P&T scenario would be possible at all without the supply of fresh fissile material.

A key parameter for evaluating a P&T scenario is the *implementation time*, i.e., the time from the start of a P&T campaign until the desired target reduction is achieved,

Based on the *initial inventory*, a *target reduction* can be defined to be achieved by a P&T scenario. This quantity affects the extent to which the requirements for geological disposal could be influenced by P&T. For example, it was assumed under the standard scenario that either the initial inventory is to be reduced by 90% (remaining inventory 14 t, column S in Table 1) or is to be reduced to a value of 5 t (variation 1). In this case, the required implementation period would increase from 149 years according to column S to 242 years.

Parameter \ Variation	S	1	2	3	4	5	6	7	8	9	10
Irradiation Time (y)	3				5	3	5	5			
Interim Storage + Processing time (y)	3				3	5	5	8			
Cycle Length (y)	6				8	8	10	13			
Reactor Inventory (% actual TRU)	50				63	37	50	38			
Transmutation efficiency per cycle (%)	10		15	20							
Conversionfactor	0										
Thermal Power at t=0 (GW)	6.4		9.6	12.8	4.8	4.8	3.8	3.0			
Reactor Power (MW)	400										
Nr. of Reactors at t=0	16		24	32	12	12	10	8			
Final Reduction (t)	14	5									
Implementation Time (y)	148	242	97	72	199	199	249	325	270		
Separation Factor (%)	99.9								99.5		
Sum Separation Losses (t)	1.2	1.3	0.8	0,6					6,4		
Reactor lifetime (y)	40									30	60
Number of Reactors	33	37	37	41	30	30	30	30	34	39	25

Table 1. Variation of key parameters and feedback on target achievement starting from an initial inventory of 140 t TRU.

In a transmutation step, the fuel remains in the reactor for a number of years, *the irradiation time*, typically 3-5 years. During this time, a certain percentage of the initial TRU inventory is converted by fission. Typically, the longer the fuel remains in the reactor, the greater is the percentage of the initial inventory converted by fission. The irradiation time may be limited by the fact that the remaining portion of TRU as fissile material is no longer sufficient to operate the reactor, or that the fuel itself can no longer be used.

After use in the reactor, the spent fuel must be stored prior to reprocessing to allow the contained radioactivity and associated heat generation to decrease. Only after this interim storage period is the fuel reprocessed and separated TRU produced. Typically, the longer the fuel has been irradiated in the reactor, the longer the required *interim storage time*.

The separated TRU must be processed into new fuel and this fuel must be used for the next cycle in the reactor; the time required for this represents the *processing time*. For the sum of intermediate storage and processing time, a time requirement of at least 3-5 years can be assumed.

The sum of the irradiation time, the intermediate storage time and the processing time forms the *cycle time*. If the cycle time increases from 6 years according to column S to 8 years or 10 years, the implementation period also increases from 148 years to 199 or 249 years (variations 4-6). In particular, the interim storage period required prior to reprocessing can also assume significantly higher values. Already with an increase of the required interim storage time to 8 years and a resulting cycle time of 13 years, the implementation time increases to 325 years (Variation 7). This shows that the cycle length is a critical variable for the feasibility of a P&T scenario.

The difference between the initial amount of TRU loaded into the reactor and the amount of TRU unloaded represents the transmuted fraction during a reactor deployment. Related to the initial amount of TRU, one speaks of the transmutation fraction or the *transmutation efficiency*. The higher the transmutation fraction achievable in one step, the fewer steps are necessary to achieve the desired target reduction of the initial inventory. Achievable values in the range of 10-20% are discussed for this purpose. If the transmutation share can be raised from 10% according to column S to 15% or 20% (variations 2-3), the required implementation time is reduced from 148 years to 97 or 72 years. Conversely, a lower achievable transmutation fraction would lead to correspondingly increased implementation times.

Since the cycle time achieved for each transmutation step in order to eliminate the respective transmutation fraction, these two quantities together with the target reduction essentially determine the required implementation period.

Of the total TRU inventory available at any given time, only a certain proportion can be in use in the transmutation reactors; this proportion represents the *reactor inventory*. The remaining TRU inventory is in spent fuel or is in the process of being processed into new fuel. The ratio of the respective reactor inventory to the available TRU inventory is determined by the ratio of the irradiation time to the total cycle time. Thus, if the intermediate storage time and the processing time were equal in sum to the irradiation time, 50% of the inventory on hand could be in the reactors at any given time (Variations S and 5). If, on the other hand, the intermediate storage time and the processing time were in sum only half as large as the irradiation time, two thirds of the inventory currently present could be in the reactor and vice versa (Variations 4 and 5).
If a uranium-free fuel (IMF) is used, the transmutation fraction is equal to the amount of TRU converted by fission, which in turn is proportional to the fission energy released. Since this energy is produced within the cycle length, this simultaneously determines the thermal power of the required transmutation reactors. If a constant transmutation fraction is assumed, the available amount of TRU and thus also the amount of TRU fissioned per year and the resulting thermal power decreases exponentially over time, cf. figure 1.

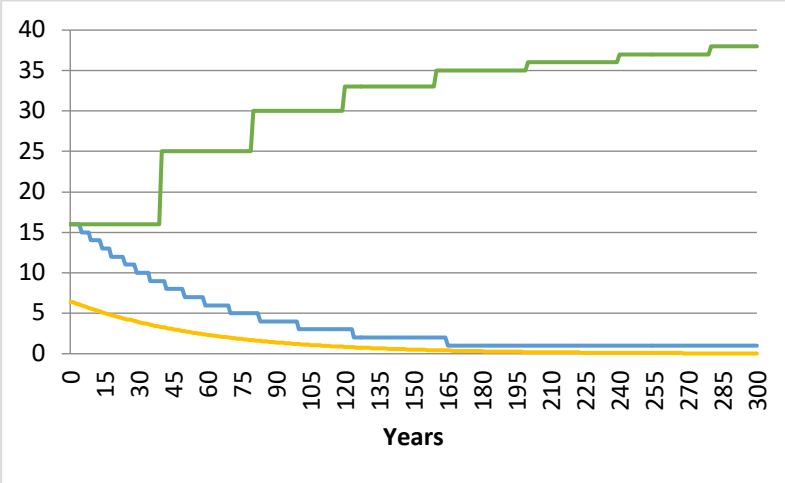

Figure 1: Simple Scenario. Development of thermal power (yellow), required number of reactors (blue) and required total amount of reactors (green) for transmutation of 140 t TRU. The scenario is optimized for throughput as some reactors only operate for a few years.

It also follows that the number of reactors required at each point in time would decrease according to the thermal power produced. While typically the required number of reactors is calculated as a time average of the total energy to be produced, in real terms a relatively large number of reactors is required at the beginning of a P&T scenario, which decreases over time. Since a reactor, once built, is available for a certain lifetime, this leads to the fact that the existing reactor capacities cannot be fully utilized in this simple scenario. If it were assumed that a reactor is available for a service life of 40 years, the existing reactor fleet would have to be replaced by new reactors after every 40 years. At this point in time, as many reactors would have to be built to be able to produce the thermal power still to be generated at this point in time, cf. Figure 1. If one assumes deviating reactor lifetimes of 30 or 60 years, this would result in a correspondingly larger or smaller number of reactors to be built in total (variations 9 and 10 in Table 1).
If one were to realistically attempt to reduce the required number of reactors and the associated construction costs, the implementation period would conversely be increased.
In each cycle, the spent fuel must be reprocessed once. The *separation factor* that can be achieved in this process determines what fraction of the remaining TRU inventory is transferred

to the waste stream and is no longer available for a further transmutation step. Thus, the higher the separation factor, the lower the total losses that ultimately have to be transferred to a final repository. For transmutation, separation factors of 99.9% are targeted. If, on the other hand, only a separation factor of 99.5% can be achieved, the losses from reprocessing would increase by a factor of 5 (variation 8 in Table 1). Since the losses from reprocessing would already be greater than 5 t at this separation factor, a target reduction to 5 t would no longer be possible. High target reductions are only conceivable with very high separation factors.
The achievable target reduction is limited by the fact that in each cycle step a proportion of the TRU present is transferred to the waste stream. In practice, at the end of a P&T scenario, a fraction of TRU will still remain in the fuel of the last reactor in use. Thus, the remaining amount is composed of the total losses of TRU during reprocessing and the reactor inventory remaining at the end of the scenario.
Therefore, in order to still achieve the same target reduction with a lower separation factor, the reactor inventory remaining at the end of the scenario must be reduced accordingly. This also increases the required implementation time (also Variation 8 in Table 1).
The variations show that depending on the P&T technologies available, implementation scenarios differ significantly in various aspects.

REFINED MODEL

The simple model used to assess the impact of different parameters on P&T scenarios does not reflect the isotopic properties of the different (fissile) nuclides in the fuel.
To better account for those properties, a more refined model has been introduced in (Frieß et al., 2021). In this model, the properties of the single elements in the nuclear materials are taken into account. The initial inventory has been updated to the most recent data of approximately 150 t of transuranium elements. The limiting condition in this model is the fraction of the different actinides in the fuel: it must stay the same over the implementation time. This model still lacks a proper consideration of the changes in isotopic composition. But it gives first insights into the effects of the transuranium mixture on transmutation scenarios. It further shows how the boundary conditions build into the model shape the outcome.
(Friess 2021) analyzes three different scenarios: the use of critical fast reactors with MOX fuel (FR), the use of accelerator-driven systems with inert-matrix fuel (ADS), and the use of molten salt reactors (MSR). The second scenario (ADS) will be presented in more detail since it is comparable to the simplified model discussed in the previous section. Likewise, it is based on ADS similar to the European EFIT concept. Inert-matrix fuel is used, and reprocessing is done by pyrochemical processes. The element-specific transmutation rates are from Chen et al. (2008).
The most important boundary conditions compared to the simple model above are:

- Reactors are only built and operated if their operation can be sustained over the whole estimated lifetime. Sufficient fuel must always be reprocessed to avoid costly "idling" of the transmutation plants or their early decommissioning. The capacity of the reprocessing plants has been chosen to ensure this.
- Element-specific reprocessing is possible and can be fitted to the requirements of the respective transmutation facility. It is assumed that each time a reactor is reloaded, the same composition of fuel is used in relation to the individual TRU. This does not change over the course of the scenario. This assumption is still simplistic because the isotopic composition of the individual TRU would change, and the individual isotopes would have quite different effects on the operation of the reactor.
- Element-specific transmutations rates are implemented in the model. Curium build-up under irradiation is thus mirrored in the simulation.

Parameter \ Variation	ADS
Implementation Time	88
Reactor Power (MWth)	400
Capacity (%)	74
Reactor lifetime (y)	40
Irradiation Time (y)	4
Number of Reactors during Scenario	2
Initial Pu Inventory (t)	128.6
Initial MA Inventory (t)	21.1
Pu reduction (t)	-1.2 (-1%)
Final Inventory (t) Pu, Am, Np	138
Final Inventory of separated Pu (t)	121

TABLE 2: Result overview of the scenario ADS+IMF.

Key parameters and results of the model are shown in Table 2. The evolution of the minor actinide inventory over the scenario is plotted in Figure 2.
The high americium inventory in the reactor and the related transmutation efficiency for americium lead to the fact that in the considered scenario the americium inventory is the limiting factor: there is not enough americium available to load further plants. In total, only two irradiation facilities are deployed. These are commissioned one after the other to be able to transmute as much of the inventory as possible over the operating times of 40 years in each case. Parallel operation of both plants is not possible due to the limited americium inventory.
The key boundary condition that the fuel composition needs to be constant artificially reduces the possible throughput and the required facilities due to americium shortage. While the plutonium and neptunium (Figure 2) inventories change almost not at all, the americium inventory is reduced to just under one third of the original value (from 14.3 tons to 4.7 tons). Approximately 2 tons of Curium are produced over the implementation time. Figure 2 does not mirror the fact that the most important Curium isotopes have comparable short half-lives and decay to Plutonium. At the end of the scenario, only about 165 kg of Curium are left. Consequently, another 2 tons need to be added to the plutonium inventory (Fries 2021, 213).
Instead of Americium, more plutonium could also be used in the fuel. Thus, more plants could be operated and a larger proportion of the TRU inventory would be transmuted. Modelling a changing fuel composition requires detailed information on the ability of the ADS to adjust to changing fuel compositions and fuel evolution over scenario time – which is lacking.
Naturally, the amount of fission products increases due to continuous fission. The use of inert-matrix fuel and a fast spectrum leads to a different spectrum of fission products. Especially cesium-135 is produced in a higher fraction (Frieß, 2017). Due to the small throughput in this scenario, the produced amount is small compared to the already existing inventory and not shown explicitly.

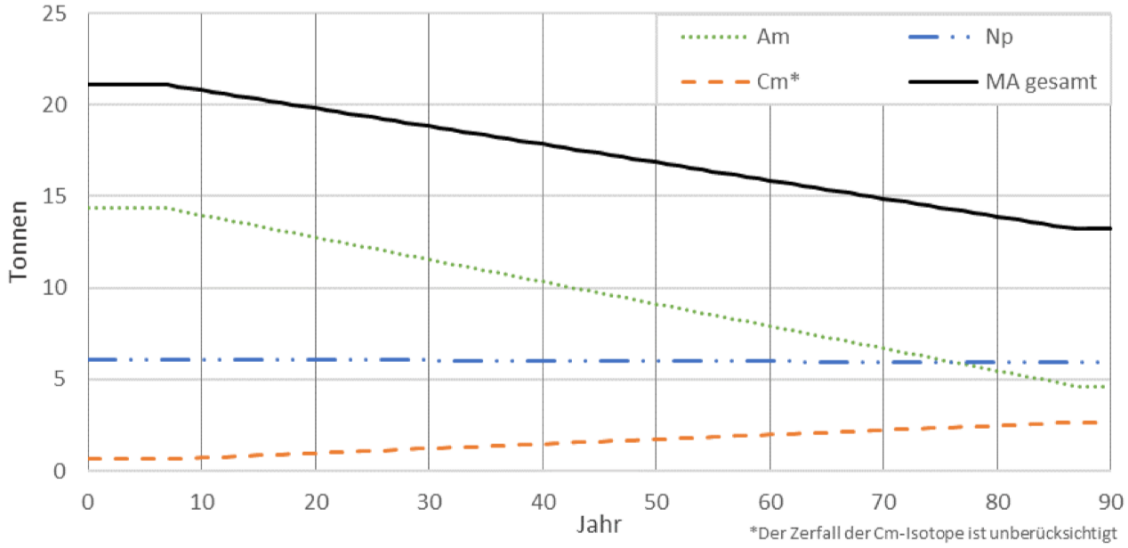

Figure 2 shows the evolution of the different minor actinides in the scenario. Due the limitation imposed by the fixed fuel composition, only a small amount of minor actinides is actually fissioned in the scenario.

RESIDUAL NUCLEAR WASTE INVENTORY

The simple and the more refined model showed that the residual nuclear waste inventory after a P&T treatment would change significantly. Key results are:

- Since only a certain percentage of the respective inventory is transmuted per P&T cycle, a residual amount of TRU always remains in the high-level waste at the end of the P&T treatment for which a disposal option is required. Depending on how many P&T cycles are performed and how complex the fuel composition, this percentage can be further depressed. Especially the curium build-up during the P&T treatment needs to be considered.
- Process losses during reprocessing and fuel fabrication contribute to the residual amount of transuranium elements. Especially the efficiency of the separation processes influences this amount. With current transmutation schemes, the initial inventory of FP increases overall. The radioactivity of the FP strongly decreases in the first 300 years. The waste also no longer exhibits any relevant thermal output after this period. However, this goal could also be achieved by pure waiting and a long-term interim storage of the waste without P&T treatment. The time period would even be shorter, since no radioactive isotopes are produced during the transmutation implementation phase.
- Relevant proportions of very long-lived mobile FP remain in the waste stream, which continue to determine the requirements for disposal, especially in the long-term safety analysis. Theoretically, it would be possible to partition the relatively small volume of long-lived FP as well and dispose them separately (e.g., in deep boreholes). Practically, however, extremely high separation efficiencies would have to be achieved during partitioning. Appropriate processes do not yet exist or have hardly been developed (IAEA, 2004; OECD/NEA, 2018). Transmutation of these long-lived FP by neutron irradiation of targets made from the isotopes is currently not pursued worldwide and is considered technically difficult (OECD/NEA, 2002).
- Remaining waste streams such as ingot molds or waste from research reactors would most likely not be treated with transmutation and is thus not covered in the models.

Those aspect must be taken into consideration when assessing the requirements set on waste disposal sites.

CONCLUSIONS

In this publication, we briefly discussed the concept of partitioning and transmutation (P&T), the status of the technology, and its possible implications for a final repository using a simple and a more refined model.

The simple model treats all TRU without element specific discrimination whereas the more refined model considers element specific transmutation rates. However, both models do not cover the changing isotopic fuel composition, which is highly relevant to reactor operation.

Both models are based on roughly the same basic assumptions (German TRU inventory, use of ADS with IMF) but yield highly different results. It is therefore of utter importance to communicate boundary conditions and assumptions very clearly when presenting possible P&T implementation results. Further, to make reliable statements on the effects of P&T, much more elaborate models are needed.

In such scenarios, it is assumed that all necessary technology is available on an industrial scale, although many technologies necessary for full scale P&T implementation remain to be demonstrated. Especially industrial scale partitioning of trans-plutonium actinides and fuel fabrication will be an issue. Fast reactors and accelerator-driven systems are currently not in operation with sufficient reliability. But even if all necessary technologies are developed and put into operation for decades or even centuries: would this impact the requirements for geological disposal sites?

The main focus of this paper was on P&T implementation scenarios, and we did not discuss the impact of P&T on the design of a final repository. The waste forms and compositions that remain at the end of a P&T treatment of waste depend heavily on the specifics of the technology park and process schemes employed. The more elements to be treated with P&T, the more complicated, expensive, time-consuming, and technically challenging the P&T scheme becomes.

Both models, however, agree that P&T will still require the long-term storage of significant amounts of transuranium elements. The long-term mobile fission products that dominate the long-term safety analysis of final repositories are not targeted at all in these transmutation concepts. Further, certain waste forms such as radioactive waste from research reactors or process losses during reprocessing and fuel fabrication will likely not be treated with transmutation.

This result is no surprise since large studies such as (National Research Council 1996) concluded that it is “highly unlikely”, if not "absolutely impossible" that a repository can be dispensed with.

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