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Article

A First Step to Zero Waste Nuclear – Advanced Strategic Thinking in the Light of iMAGINE

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Abstract: Traditionally there is a gap between reactor operation and the consideration of nuclear waste in the final disposal. Fuel is produced and fuel must be disposed of in the view of the reactor operator, fuel has to be cleaned in the reprocessing and new solid fuel has to be produced in the view of the chemist. iMAGINE is designed to overcome this separation through the breakthrough development applying an optimized, integrative approach from cradle to grave of nuclear energy production as a first step to come as close as possible to the vision of zero waste nuclear power. It is described here the first time all in three the steps: reactor, fuel cycle, waste, providing the ratio behind each of the choices taken to come to the overall solution to open the discussion and thinking process on what could be achieved by a really innovative approach to integrated nuclear energy production. The opportunities regarding the handling of the remaining waste will be discussed with a view on the expectation of the final disposal community, the study 'Nuclear waste from small modular reactors', and the IAEA report 'waste from innovative types of reactors and fuel cycles - a preliminary study. The aim of the is not to find answers to each of the raised points, but to identify first potential approaches and potentially promising ways to go, as well as to stimulate a discussion among experts. In the best case this could lead to a change of track for nuclear to become an even more sustainable and at least as important, trusted technology to help solve the net-zero challenge.

Keywords: nuclear; nuclear energy; nuclear waste; final disposal; nuclear reactors; reactor physics; molten salt reactors; nuclear chemistry; fission products; salt clean-up

1. Introduction

Traditionally there is a gap between reactor operation and the consideration of nuclear waste in the final disposal. Fuel is produced and fuel must be disposed of. There were only two options for the fuel cycle, the open fuel cycle with direct disposal versus the partially closed fuel cycle with reprocessing and disposal of vitrified waste. The key driver was at this point the reactor operation scheme and the planned approaches for closing the fuel cycle through the reuse of Pu in fast reactors [0] without considering the effects on the final disposal.

The discipline of nuclear waste management tried for some time to bridge the gap between reactor operation and final disposal with more or less success, at least it led to a discussion between the disciplines and partly joint studies of P&T technologies, e.g. Eurotrans [2] & Europart [3] as P&T projects and their impact on final disposal of the leftovers e.g. RED-Impact [4]. However, often the views significantly diverge between the P&T community, mainly driven by the approach to reduce the radiotoxicity [5] of the spent fuel and the final disposal community observing the potential dose released over very long term from a final disposal facility. Unfortunately, both concepts lead to very different optimization objectives. The P&T community is concentrating on ways to deal with reducing the Transuranium content in the waste stream, while the final disposal community is observing the migration of highly soluble long-lived fission products to the environment to understand paths leading to potential dose exposure to humans in a far future. Even joint studies like the German P&T study moderated by an independent body, acatech, the National Academy of Science and Engineering of Germany [6] facilitated intensive discussions, but was not successful in overcoming this gap.

Meanwhile other countries decided against closed fuel cycle strategies, and the related required reprocessing of spent nuclear fuel, due to different drivers, e. g. US, Canada, Sweden, Finland with the latter currently at the forefront of licensing and construction of a deep geological repository [8]. The reasons for these different national approaches are in part driven by our understanding and capability with respect to the underpinning technologies. Reprocessing, fast reactors, and geological disposal are technologically demanding, with respective performance characteristics. They are characterized through large uncertainties making objective decisions on the system very difficult. As a result, decisions on fuel cycle have tended to move into political discussion, either within the scientific bodies or at a governmental level.

However, when thinking about new, advanced reactor technologies, there is a fresh tendency, at least formulated in the UK and discussed in the academic community and authorities [9] – the aim is not only to design a reactor, but also to consider how can the fuel be produced? as well as how can it be disposed of? We could see this as a first, important step, if we think about potential breakthrough technologies or game changers like iMAGINE [10, 11], the view has to go much further. This led to iMAGINE – a breakthrough nuclear technology to operate on spent nuclear fuel without reprocessing.

The question must be, like in other technologies can we find reasonable ways for the re-use of the by-products (deliberately not calling them spent fuel or separated fission products waste) instead of declaring it as waste. Even if comprehensive solutions are not available yet, it is at least worth to think about and to research it. Can a breakthrough technology reduce the final disposal challenge and if yes, to what extent? What could be done already in the reactor and the fuel cycle to minimise the effort for dealing with the waste later? Can we achieve these goals only for the future by applying the new technologies or will it be possible to treat legacy wastes? If this is possible, to which extent, will it be limited to spent fuel only or will it also encompass already vitrified remainders of reprocessing?

These points will be discussed with a view on the expectation of the final disposal community [12], the study ‘Nuclear waste from small modular reactors’ [13], and the freshly published IAEA publication ‘WASTE FROM INNOVATIVE TYPES OF REACTORS AND FUEL CYCLES - A PRELIMINARY STUDY [14]. The aim of this publication is not to find an answer to each of the points raised, but to identify first potential approaches and potentially promising ways to go. Core point is to stimulate a discussion which could lead in the best case to a change of track for nuclear to become an even more sustainable and at least as important, trusted technology to help solve the net-zero challenge.

2. The way to iMAGINE

The way of the development of iMAGINE can be described through a thinking process from the inside – the reactor technology, to the outside – the complete nuclear system from cradle to grave as shown in Figure 1 with some of the major decision criteria given in each of the steps. The different steps in the decision criteria and the thinking process are described in the following paragraphs of this chapter in more detail.

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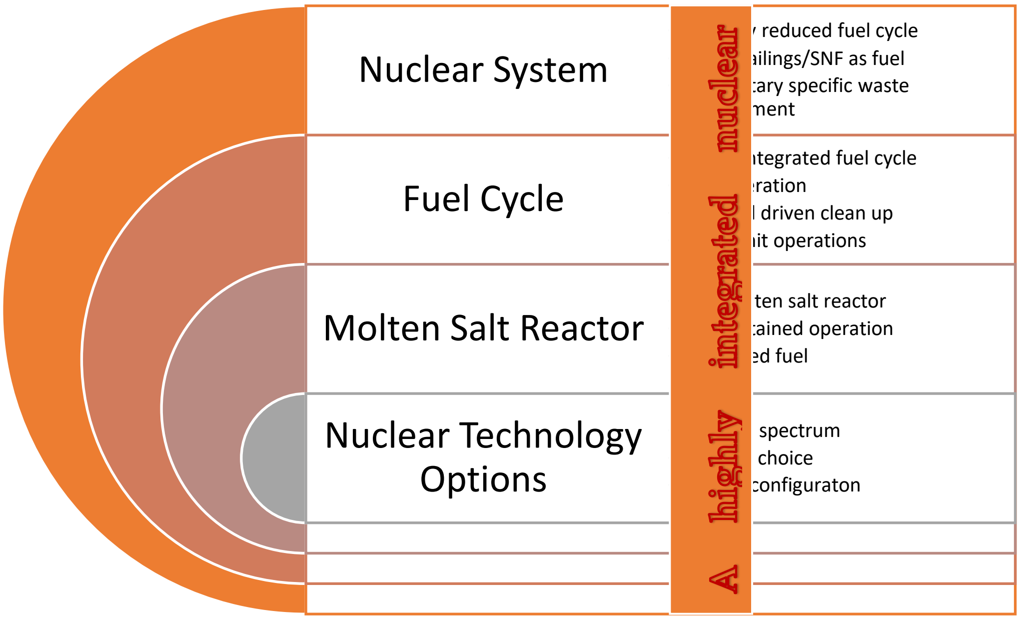

Figure 1. The different levels of decision making and optimization applied through the development of iMAGINE as a highly integrated nuclear system.

2.1. Optimizing the reactor technology

The whole concept of iMAGINE is based on molten salt reactor technology operating on spent nuclear fuel without prior reprocessing and demand driven salt clean-up. The system optimization for iMAGINE starts by the selection of a specifically attractive nuclear process technology options given in the scheme of potential molten salt reactor configurations as presented e.g. in [15] to make the best possible use of the advantages of a reactor based on liquid fuel, bypassing the challenges and cost of solid fuel production related to conventional closed fuel cycle systems like sodium cooled fast reactors [16].

The driver for the choice for a molten salt reactor was based on the demand for self-sustained long term operation so as to achieve sufficient breeding such that no fissile material needs to be supplied after the start-up phase [0]. The main requirement here is to identify a salt composition with a reasonable melting point and a very high loading of fertile material to assure the required breeding efficiency [17]. Here, the uranium-plutonium (U-Pu) cycle with shorter breeding cycle defined through the decay of the interim product Np-239 of ~2.3 days seems to be a practically better option than the thorium-uranium (Th-U) cycle with longer breeding cycle of ~27 days defined through the half-life of Pr-233. In addition, there is higher experience of handling chlorides of U (e. g. through the usage of uranium chloride salts for pyro-reprocessing) compared to Th, no to forget that there is a significant amount of Uranium already available in the form of spent fuel and tailings form LWR operation. These points are making uranium chloride a better option for molten salt reactor. A further advantage of U/Pu cycle is the significantly higher number of access neutrons in the U/Pu system compared to the Th/U system which will ease reactor design by allowing a higher leakage of neutrons and providing a better breeding efficiency. The challenge with U/Pu system is the requirement for a fast reactor configuration with the significantly higher fuel amount to achieve criticality compared to a Th/U based system which would operate in a thermal spectrum.

Collected opportunities of molten salt reactor technology based on liquid fuel U/Pu salts, compared to currently operating LWRs:

- Online feeding to avoid excess reactivity which is required to operate a LWR core through one cycle.

- Operating on already used fuel instead of fresh fuel required for the operation of a LWR. In a molten salt reactor fresh and used fuel will mix in the reactor due to the liquid state of the fuel
- Internal closed fuel cycle operation through self-sustained breeding instead of a pure converter system like LWR which requires fresh, enriched fuel for every cycle and a complex fuel cycle, typically based on aqueous reprocessing to re-use Pu once in MOX fuel which is then sent to final disposal
- Temperature level control, and resilient operation due to strong feedback effects, instead of burnup compensation based on boric acid in LWR
- Online salt clean-up for closed fuel cycle without waiting times in contrast to cycling times of several years until Pu in form of MOX fuel can be once cycled back into an LWR
- Avoiding costly fuel production of solid reactor systems for closed fuel cycle operation, which is reflected in the high cost of MOX fuel for LWRs and the even higher cost for fuel production in potential fast reactors [16]

2.2. Highly integrated fuel cycle incorporated in a reactor system

The reactor designed for self-sustained operation will require a salt clean-up system or reprocessing of the salt. The approach here is to perform salt clean-up or "reverse reprocessing" i.e. to separate elements which prevent the reactor from long term operation (fission products) instead of separating the elements which are required for reactor operation (fissile material) [18]. This approach will significantly reduce proliferation risk, since there will be no separated fissile material stream while allowing for completely new optimization of a molten salt system based on liquid fuel. In contrast to classical solid fuel, this clean-up approach in liquid fuel will allow reactor operation an online mode without cooling times and transports of the fuel from the reactor to the reprocessing facility. Finally, through the separation of fission products from the ***'fuel-stream'***, instead of fissile material from the ***'waste-stream'***, potential losses will just stay in the reactor reducing the demand for the separation rate which will be extensive when trying to avoid the carryover of fissile material of TRU elements into the final disposal.

Technologies in simplifying the fuel cycle while harvesting additional gains from *'fuel-stream'*, compared to the currently established nuclear system based on LWR and aqueous reprocessing:

- Operating on spent fuel without prior reprocessing, instead of demand for clean fuel, aqueous reprocessing and incomplete Pu burning in MOX fuel
- Tailored salt clean-up to remove fission products that prevent long-term reactor operation
- , instead of separation of fissile material creating proliferation issues
- Avoid costly solid fuel production and potential multi-recycling, compared to the requirement for fresh, enriched fuel for each cycle in a LWR
- Avoid mining and enrichment for new LWR fuel through internal closed fuel cycle operation
- Deliver breeding of fissile material from U-238, to use available resources currently seen as waste – spent nuclear fuel as well as tailings; reuse of reminders of the LWR technology for energy production instead of mining for new, raw materials

Avoid interim storage of SNF in cooling ponds required in solid fuel based technologies, thus reducing demand and costs.

The use of an integrated reactor in self-sustained breeding mode allows an extremely high efficiency in fuel use creating a massive amount of newly available resources for future generations, while it is based on already mined materials reducing the waste production, see data in Table 1. While in iMAGINE only ~100 kg fuel, e.g. Uranium from SNF are required for producing 1 TWh electricity, based on 42.5 kg/TWh thermal [19, 20] and a thermal efficiency of 42.5% (reasonable for a temperature level achievable in the MSFR). In contrast, a classical light water reactor requires ~2500 kg of fresh fuel in fuel assemblies, based on an average burnup of 50 GWd/tHM for the fuel assembly and a thermal efficiency of 33% (saturated steam process). The LWR fuel must be produced through enrichment of 20 to 25 tons of natural uranium, depending on the enrichment process applied [21] to achieve the target burnup of 50 GWd/tHM. To extract the required 20 to 25 tons of uranium, about 28500 to 72000 tons of raw material has to be removed from the ground for separating the uranium oxide, (data from Olympic dam, the largest Uranium mine with Uranium grades average from 0.07 to 0.035% U [22]. However, there is the opportunity to extract not only Uranium, many mines in the modern world will extract different metal ores in parallel.

However, there is the opportunity to extract not only Uranium, many mines in the modern world will extract different metal ores in parallel. In the iMAGINE concept we already have the raw materials in the form of spent nuclear fuel that is currently viewed as a liability, requiring $10s billions of investment to deal with the legacy in the UK alone.

Focussing the view back to the UK, the potential resources, currently declared as nuclear waste materials give the following picture. NDA data from the radioactive wastes report 2016 [23] indicates 7000 tons spent fuel and 200000 tons of Uranium tailings as waste. iMAGINE can turn these ‘waste materials’ into an energy resource. These already existing materials can deliver electricity for the UK for about 7500 years based on the 100kg uranium required per 1TWh and on the data given above and the current UK electricity consumption [24]. No further mining is required for a substantial time. Mining is currently seen as the main source of ecotoxicity [25] and CO_2 release together with the very energy intensive enrichment process. Mining is often one of the activities heavily criticised when discussing nuclear in the public. For countries which do not have vast amounts of spent nuclear fuels and tailings it could be attractive to avoid mining through the use of non-conventional uranium resources [31] after the start-up of the system based on enriched Uranium.

Table 1. Fuel required and waste produced for delivering 1 TWh of electricity (30 h electricity for whole UK) or the equivalent of 350 000 tons of coal.

	iMAGINE	**LWR**
Fuel usage	**100 kg** from existing sources like SNF or tailings transformed into uranium chloride	**2500 kg** in the form of UOX pellets in fuel assemblies
	Using up already extracted/mined materials	made from **20 to 25 tons** of natural Uranium through enrichment
		separated from **28500 to 72000 tons** of mined material
Waste production	**100 kg** of fission products to be separated through new processes to assure long term operation Opportunity to optimally condition separate elements to limit mobility	**2500 kg** spent fuel in Fuel Assemblies containing TRU elements requiring to be stored and later finally disposed in a deep geological repository
		~ **20 tons** of tailings which have to be stored
		28500 to 72000 tons of mined material with low activity creating major source of radiotoxicity to the environment

The significant gain in available resources for future generations is visualized in Figure 2. The data for this calculation is based on the energy production and resources taken from [26, 27] whereas the sum of the reserves and reasonably exploitable resources is used for hydrocarbons as specified in the publications [26, 27], a number which is anyway somewhat speculative. Anyway, the figure should be seen as an illustration since resource projections show significant differences, see e.g. [28] delivering significantly higher numbers for the fossil fuel reserves. The general numbers coincide with the number delivered by Rosatom [29] for their closed fuel cycle analysis with 94 to 6% Hydrocarbon to nuclear now and 14 to 86% for the closed fuel cycle. The differences can be explained by the different closed fuel cycle efficiency and different resource estimations for hydrocarbons.

The application of the iMAGINE technology is active sustainability improvement. iMAGINE delivers more opportunities for future generations through technology development, providing a significantly larger flexible and 24/7 available energy resource which is of essential interest following COP26 [30].

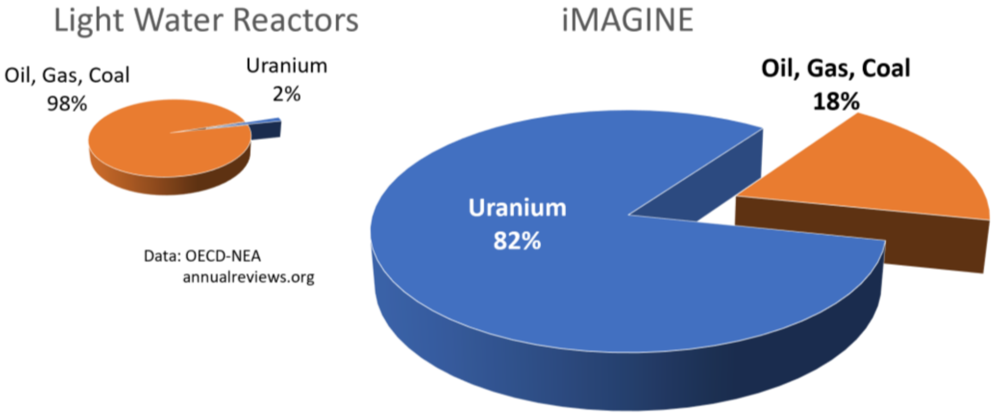

Figure 2. Availability of resources using LWR technology versus the application of iMAGINE (the overall size of the chart replicates the overall amount of available resources).

Following the opportunities of the iMAGINE approach for the reactor and the related highly integrated fuel cycle, the discussion will be opened to investigate potential influences on the nuclear waste management. What could be achievable if we think of iMAGINE in an innovative way from cradle to grave?

2.3. Waste production and final disposal opportunities

Spent nuclear fuel from light water reactors and vitrified wastes from reprocessing form the major contribution to high level nuclear waste which is foreseen for deep geological disposal, depending on the strategies of the countries directly as fuel assemblies or in the form of vitrified waste. According to the IAEA, at the end of 2020, 430+ kt (HM) had been discharged from NPPs, out of which about 300+ kt (HM) are stored in facilities at reactor sites or storage sites, while the remainder has been reprocessed. The annual discharge worldwide is currently ~ 10 kt (HM) [33]. The composition of spent fuel depends on the reactor system, the initial enrichment, and the achieved target burnup. However, a general view is given in Figure 3 with some bandwidth of the composition, delivering the main characteristics with the by far leading component U-238, a share of fission products and smaller shares of bred Pu and still available U-235.

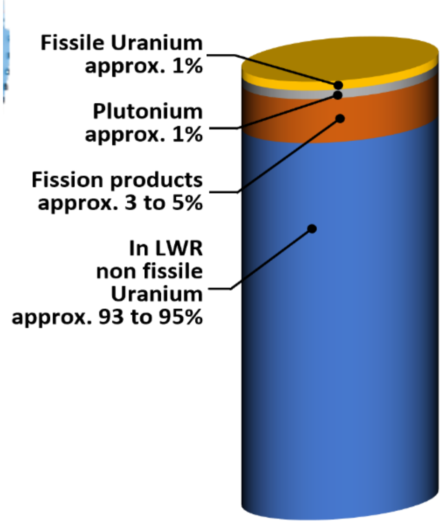

Figure 3. Uranium fuel composition after power generation in a LWR (not to scale).

This spent fuel can serve in a future iMAGINE system as fuel without prior reprocessing using the still available amounts of Uranium (U-235 and U-238) as well as Plutonium. thus 95+ % of a material which is currently considered as waste. The fission products will have to be separated out by the demand driven salt clean-up system during operation. Uranium and Plutonium will be completely utilized relieving the maximum amount of energy from the fuel. iMAGINE leaves a only 100kg waste per 1 TWh in fission products instead of 2500 kg in spent nuclear fuel from a LWR. It releases a factor of 25 times more energy from each fuel unit and the related produced waste units.

When discussing about nuclear waste, the P&T community delivers the reduction of radiotoxicity over time normalized to one ton see e. g. [35, 36] with the caveat that radiotoxicity is a weighted unit overemphasizes the effect of heavy isotopes due to a higher toxicity index. An approach often criticized in the final disposal community [37]. To take this into account and to facilitate a broader discussion, the following figures are based on the SI unit activity over time per unit waste mass (Figure 4) in addition to the mass of waste per unit energy produced, as already discussed above.

Up to now nuclear waste management through partitioning and transmutation has been concentrated on reducing the activity per ton of spent fuel as given in Figure 4, using a hard to understand double logarithmic presentation. For better understanding it is here complemented by Table 2. Figure 4 indicates that the fission products provide by far the largest contribution to the activity, up to 100 years (up to a factor of 10 and higher than the actinides), after about 200 years the main contribution changes from the fission products to the actinides. The following period shows the biggest difference, actinides contribute by a ~100 more to the activity than fission products (500 to 1000 years) with this difference slowly reducing over time to a factor of ~5 after 1 mio years and then growing again. The core explanation for these changes are in the different half-lives of the contributors to the long-lived waste: bulk of fission products ~300 years, leading actinide Pu-239, ~25000 years, small share of long lived fission products, e. g. I-129, ~15 mio years, Se-79, between approx. 1.24 10^5 years and 1.13 10^6 years [38].

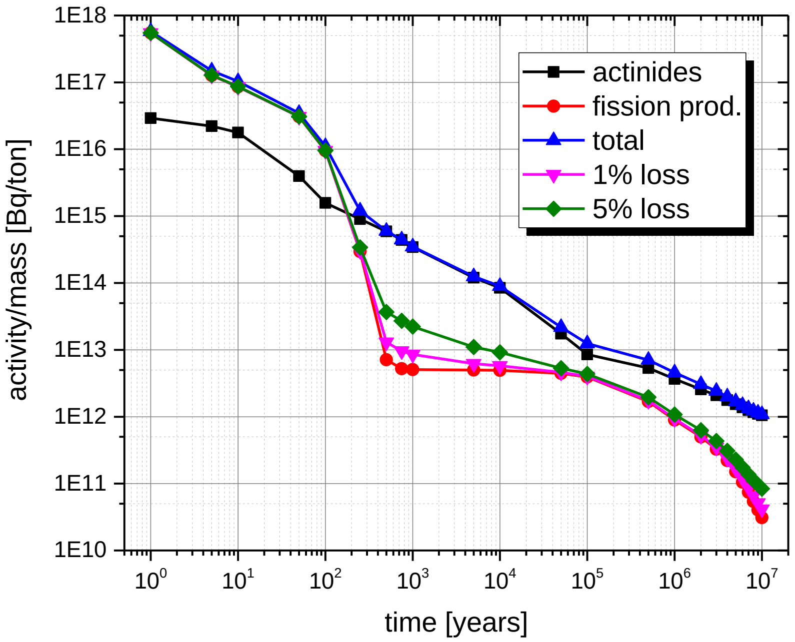

Figure 4. Spent nuclear fuel activity over time separated for actinides and fission products for one ton of fuel and the consequences of losses of actinides into the waste stream during reprocessing.

Analysing time periods instead, helps to get a deeper understanding what can be achieved through P&T, or as side effect through iMAGINE. The activity of the waste decreases naturally in the period from 1 to 500 years by a factor of 966, this can be significantly enhanced by P&T to a factor of more than 81000, P&T would reduce the activity of the waste by a factor of 84 disposing spent fuel. In the second time period from 500 to 1 mio years, the activity reduction after P&T is only a factor of 16, due to long lived fission products which do not significantly change before 100 000 years. In this time period (500 to 1 mio years) the activity of the spent fuel reduces due to the decay of the actinides with the leading actinide being Pu-239 with a half life of a bit more than 24000 years, which is utilized through P&T. In this time period, the advantage of P&T slowly reduces from the factor of 84 (after 500 years) to a factor of 5. After the observation period of the deep geological disposal the gain of P&T increases again, which shows that the activity of the waste never disappears completely, which is the nature of the exponential decay function. The reason for the increase of the P&T gain on this very long period is the absence of uranium since this very long term activity is formed through the decay products of the uranium. However, this has to be seen with a caveat, in P&T strategies, the burning of the Uranium is never discussed. Thus, it has to be asked where the uranium separated in the reprocessing will go to. iMAGINE delivers here a solution, delivering a full use of the uranium. The changes of the P&T gain over the whole period is given in Figure 5.

Table 2. activities at selected time points for the open fuel cycle as well as for the application of P&T (without losses) and the resulting potential reductions.

Time period [years]	Activity reduction factor
1-500 with P&T	81036
1-500 without P&T	966
500 gain P&T	84
500-1mio with P&T	8
500-1mio without P&T	131
1 mio gain P&T	5.1
500-10mio with P&T	230
500-10mio without P&T	552
10 mio gain P&T	35

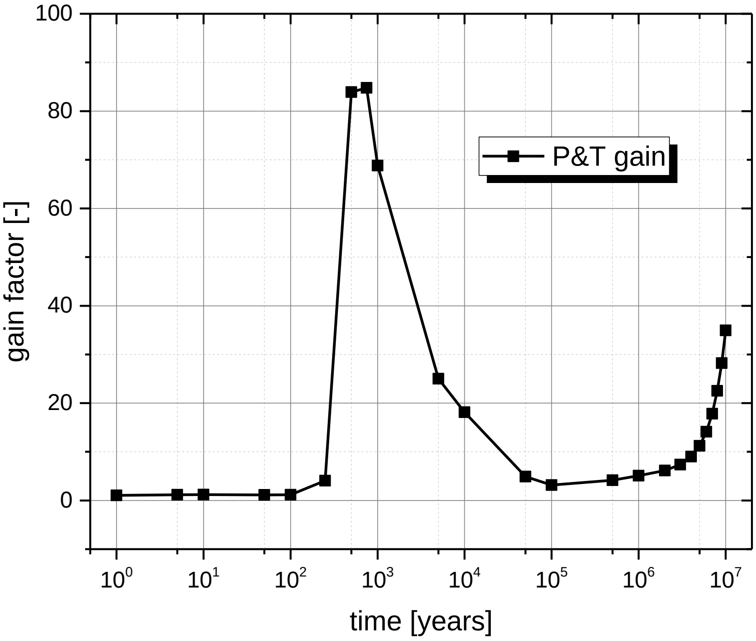

Figure 5. Gain factor of achieved reduction of activity in a final disposal through the application of P&T.

The comparison changes to a much stronger, more meaningful picture, considering the massively reduced amounts of fuel used per produced amount of energy and thus the amount of waste created in iMAGINE This approach seems to be more natural, since the final product aimed for is not waste, it is low-carbon energy to be produced. This means the amounts of waste will be reduced by a factor of 25, but it should be kept in mind that the activity is in a first order approximation proportional to the amount of energy produced. The real volume change cannot be reliably justified at this stage since the final volume of the waste is not only determined by the mass and the density when heavy metal is turned into fission products since the conditioning methods and the limitations in the achievable packing density have to be considered. This general change in the waste properties leads to a wider discussion of the potential influence of the iMAGINE approach on the design of a deep geological disposal facility (GDF).

The current situation is that either spent nuclear fuel is brought into the final disposal (e.g. Finland [39], Sweden [40] or plans of the US and many other countries [41]) or fuel is reprocessed (e.g. in UK [42], France [43], Russia [44]) and the fission products left over after the solvent extraction process are separated from the chemical solution and vitrified in boron silicate glass as high level waste. It should be considered here, that due to the stochastic nature of the fission process almost all elements of the periodic table are contained in the product solution in strongly different concentrations. Thus, the conditioning of the fission product solution in boron silicate is an engineering compromise, which though good and reliable, is not ideal for each of the individual elements.

The options will be discussed liked to a conference presentation and the question "how might alternative nuclear fuel cycles impact geological disposal" [12]:

Based on a given amount of electric power produced, the authors have produced the following list of potential changes:

- Changes in the radionuclide inventory and materials, e. g. due to reprocessing or advanced reactors
- Changes in the volume of waste, e.g. due to reprocessing or through a new waste to be expected from an advanced reactor (e.g. fuel graphite combination like from a pebble bed reactor)
- Changes in the thermal power of the waste, e.g. through separation of minor actinides or due to a higher burnup
- Changes in the durability of the waste in the specific repository environment, e.g. more durable conditioning

And recommend to investigate the effect of the changes of their impact on:

- The repository safety
- The repository cost and efficiency

This very welcomed, comprehensive list will be used to facilitate the discussions of effect of the iMAGINE approach on a geological disposal facility along the list above:

Changes in the radionuclide inventory and materials

The radionuclide inventory will change, in a first approach to a configuration close to what is expected to be the end product of P&T, thus no Uranium, no Plutonium and no higher transuranics will be delivered to the GDF. Looking deeper into the opportunities some more far reaching statements can be made. The amount of fission products to be disposed of per unit energy will not change. However, do all fission products need to be sent to a GDF? Especially when considering the significant changes due to the reverse reprocessing approach, separation of fission products from the ***'fuel-stream'***. Thus, it would have to be asked which of the separated fission product elements really have to go into a final disposal and which of the elements with shorter half-lives could be handled in a different, maybe less costly way.

Changes in the volume of waste

The volume of waste per unit of produced energy will change significantly compared to SNF. It will be closer to the volumes expected after reprocessing, since there will be no U-238 put into a final disposal. It is one of the key features of the iMAGINE technology to make U-238 accessible as an energy resource. Looking deeper into the opportunities some more far reaching statements can be made. It is again the change to reverse reprocessing which offers new opportunities. Many of the elements separated will be clean and could have potential further use while some of the elements with shorter half-lives could be handled in a different, maybe less costly way helping to reduce the volume. If the volume becomes small enough, this can open the mind to think if there are other technologies available which have maybe historically been excluded due to the waste volumes. The easiest and closest could be deep boreholes.

Changes in the thermal power of the waste

The thermal power of the waste per unit of produced energy will almost not change compared to SNF, but the waste materials producing power will be significantly more concentrated, since only specific fission products will be separated from the reactor to go into the waste stream. Looking deeper into the opportunities some more far reaching statements can be made due to the reverse reprocessing. The most relevant heat producing elements can either be kept in the reactor, to be directly transmuted in the reactor (e. g. Americium) or they can be separated (e. g. Strontium). Separation would allow their elimination from thermal power production in the final disposal almost completely whild letting them decay in a short term storage. In this case only elements with long lived isotopes which have a very low power production, would be brought into a final disposal. This would allow a massive densification of the waste for disposal which should allow a reasonable cost reduction.

Changes in the durability of the waste

The durability of the waste is in general dependent on the conditioning. The waste form of iMAGINE is planned to be clearly more stable than SNF, since the waste form will consist of conditioned elementary fission products. The conditioning opportunities for the fission products are different compared to conditioning of a variety of elements in an engineering compromise. Tailored, highly robust approaches can be used for elements which demand long term storage, e. g. iodine in silver iodide [54], which has the potential to reduce the impact of highly mobile elements on the safety case of a final disposal. Thus, the enhanced durability of the waste form, which can be chosen specific for elements which demand high attention e. g. Selenium, Technetium or Iodine will help to improve the safety case. First works are currently already on the way to investigate improved conditioning for Tc.

Risk of misuse and theft of Pu

Another topic which has not been raised in [12], but in [6] “The danger of plutonium being stolen from the repository and misused is diminished [through the application of P&T]”. The iMAGINE technology helps to improve the final disposal safety in the case of human intrusion scenarios since no fissile material is sent to the final disposal anymore.

The study nuclear waste from small modular reactors [13], points out that the amount of secondary wastes per delivered energy for small modular reactors is significantly higher than for large scale reactors, which seem to be absolutely reasonable since it follows the economy of scale. Almost the same observation is made on the amount spent fuel left over from the reactors. This again seems to be reasonable, since due to the increased neutron leakage from a small core the achievable burnup will be reduced. However, the authors did not have a look into the opportunities for the re-use of spent fuel, besides in the case of a very small SFR. This is limiting, since especially molten salt systems offer a much better opportunity for the reuse of spent fuel due to the absence of the massive pre and post processing, which will allow a much quicker and cheaper turnaround of the fuel after clean-up.

The IAEA document ‘Waste from Innovative Types of Reactors and Fuel Cycles - A Preliminary Study’ [14], concentrates very much on classical fuel cycles using the established reprocessing technologies, either aqueous reprocessing or pyro reprocessing applied to different advanced reactor types (LMFRs, VHTRs, GCFRs, AHWRs, and MSRs) as well as the decommissioning of the reactors and the related fuel cycle facilities. In addition, design methods are discussed and the resulting waste from different advanced reactors, water cooled reactors, gas cooled reactors, liquid metal cooled reactors, and dedicated actinide burners - molten salt reactors. However, the discussion of the opportunities of molten salt reactors is limited to the appearance of the two different waste streams from gaseous and soluble fission products, while the salt clean-up is discussed is expected to be delivered through classical pyro-reprocessing: ‘The reactor operation wastes are mainly divided into two streams: noble fission products that are extracted on-line during operation, and fission products that remain in the fuel salt (mainly rare earths) and are extracted by a pyrochemical reprocessing of the fuel at a small rate (on the order of 10 L/d/GW(e)). The pyrochemical reprocessing of the fuel salt is compulsory for such reactors. Thus, a solution must be developed for safely handling the salts, and for separating the actinides and fission products from the base salt. Several flowsheets for this are being developed, but a large number of experimental studies are needed to reach laboratory scale feasibility. Waste forms and packages will need to be developed and demonstrated to contain the highly corrosive nature of some salt materials, such as fluoride salts.' [14] At least the opportunities of the MSR as breeder or burner as well as the online reprocessing with the substantially reduced fuel cycle operation times are recognized, here.

In general, the really innovative approach of operating directly on spent fuel without prior reprocessing as well as the opportunity of the "reverse reprocessing" as part of the game changer technology are not discussed or analysed in all three publications. This discussion will follow later in this document, after a first look into potential technologies for the reverse reprocessing.

2.4. Potential clean-up approaches for managing undesirable fission products

The core of the reverse reprocessing approach is the removal of fission products in elementary form from the reactor salt. The fission products can be separated from the liquid fuel on the basis of their physico-chemical properties through different processes.

Separation of the fission gases

The separation of the fission gases from the liquid fuel has been treated in detail by Degueldre, et al. (2021) [45]. Actually, their solubility in the molten chloride is smaller at low temperature e.g. around 500°C than in high temperature conditions. Consequently, such separations would be better to perform after the salt away from the reactor core region and preferably the fission gas should be extracted directly downstream of the heat exchanger, where the lower molten salt temperatures are expected and therefore enable reductions in the dissolved fractions of the gases .

The effective separation of Xe and Kr has been traditionally been achieved by the distillation method, e.g. Banerjee, et al. (2018) [46]. More efficient alternative approaches such as pressure swing adsorption and temperature swing adsorption, as well as membrane-based separations are based on physical sorption where specific porous adsorbents can be used to preferentially adsorb either Xe over Kr or vice-versa. The core challenge will be that the porous material must be totally inert with the molten salt components, specifically the corrosive salt.

Separation of the noble metal and metalloid components from the liquid fuel.

The noble metals have been shown to be the most criticality relevant fission products dissolved in the fuel salt. Thus, their separation will be one of the core challenges [57]. The molten salt turns through the energy production into $NaCl - UCl_3 - UCl_4 - FpCl_\xi$, where $FpCl_\xi$ are the chloride of fission products (Fp), which includes the noble metals (Nm), Tc, Ru, Rh, Pd and other elements such as Ag, Cd, In, Sn and Sb that can precipitate under a reducing atmosphere (e.g. 1% H_2 in Ar). The general reaction for the reduction of the noble metal chlorides is written as:

$$NmCl_\xi + \xi/2H_2 \Rightarrow Nm + \xi HCl$$

The reduction process is guided by the values of the metal ion reduction potentials, $E°$, which are given as a function of temperature for comparison in Figure 6. There is also the chance for the co-precipitation of inter-metallic compounds such as NmU_3 at lower temperature e.g. 500°C, which must be controlled.

The precipitation may be completed by additional reduction, e.g. using H_2 doped Ar atmosphere, with the production of elementary Mo, Ag, Cd, In and Sn. However, Sb and

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Te as well as Se and Br may form SbH_3, H_2Te and HI as well as H_2Se and HBr respectively and they can be collected separately.

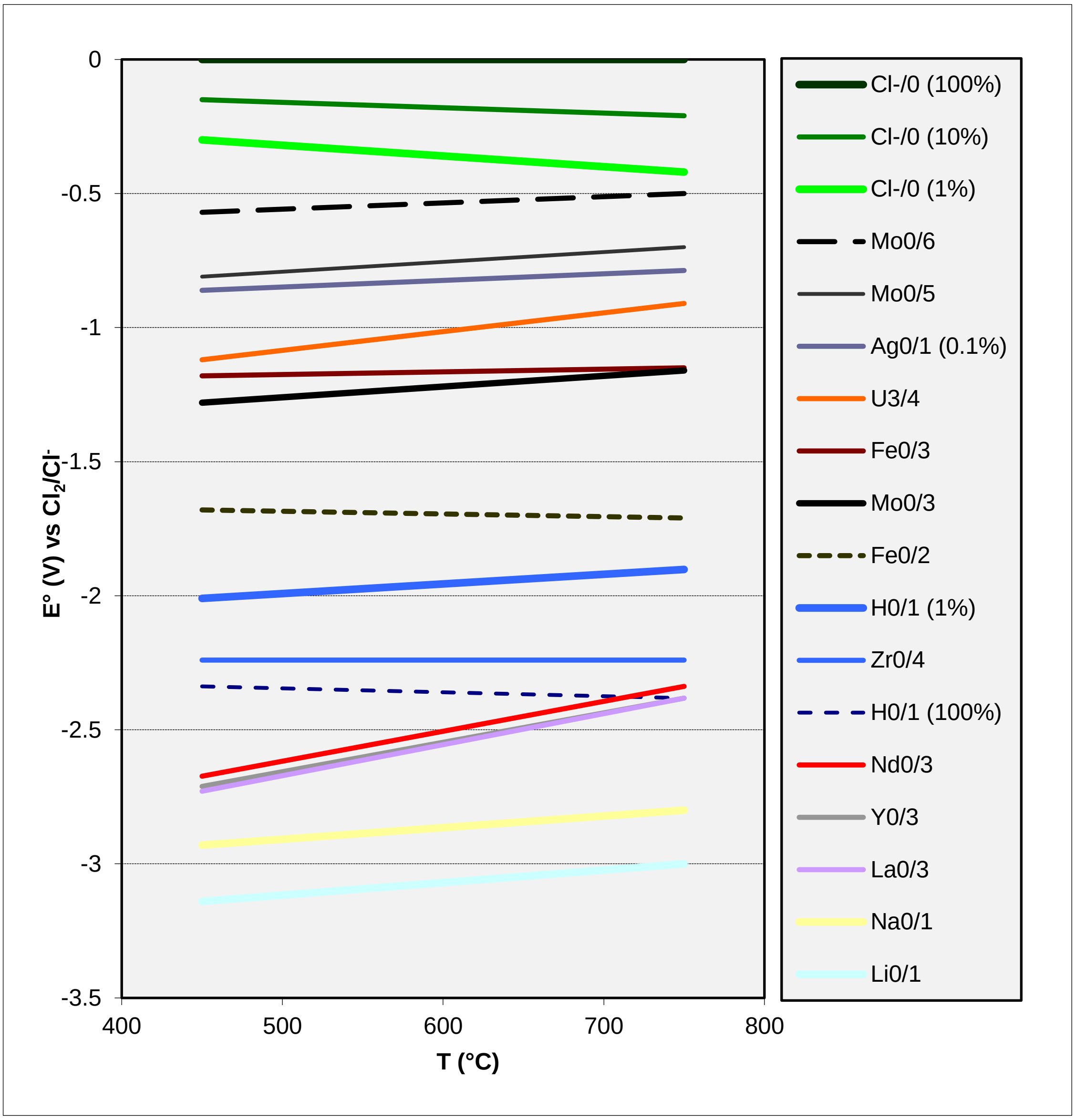

Figure 6. Redox potential Vs temperature of relevant metal couples in chloride salts relevant for the reverse reprocessing (Conditions Redox potentials (V) vs $E^0(Cl2/Cl-)$, adapted from Young & Sham (2018)[47] and from Koyama, et al. (1997) [48]).

Separation of the volatile chlorides

High valence metals such as penta- and tetra- valent fission products or actinides are volatile and can be vaporised or sublimed from the melt. Their production by oxidation reaction with chlorine can be written as:

$MCl_\xi + ½(n-\xi)\ Cl_2 = MCl_n$, where n=4 or 5

Higher valence chlorides are generally more volatile than those produced at low valence state. This is not a formal rule, but thermodynamic data in Table 3 shows that this is at least valid for $MoCl_n$ and UCl_n. It is also valid for $SnCl_\xi$ (with T_B $SnCl_4$: 114°C) and tentatively for chlorides from the same row: AgCl, $CdCl_2$, $InCl_3$ and $SnCl_4$. This is also the case for $ZrCl_3$, which readily oxidises in the presence of chlorine to give $ZrCl_4$, for which T_M: 437°C and T_B: 331°C (see Table 3). Such low temperatures suggests that $ZrCl_4$ could readily evaporate from the molten chloride mixture.

In the molten salt mixture, there will be combination of Mo(II), Mo(III), Mo(IV) species, such as $MoCl_2$, $MoCl_3$ as well as corresponding complexes and cluster salts (e.g. $[Mo_6Cl_{14}]^{2-}$,$^{-}$ $[MoCl_6]^{2-}$), Those species can be oxidised further to give dimer $(MoCl_5)_2$. Indeed, the high temperature reactions of molybdenum with chlorine in molten alkali metal chlorides were investigated between 400 and 700°C by Volkovicha, *et al.* (2003) [49] in selected melts: LiCl–KCl, NaCl–CsCl and NaCl–KCl. In these melts Mo reacts under 1% Cl_2 – Ar atmosphere and initially produces $MoCl_6^{2-}$ and then a mixture of Mo(III) and Mo(V) chloride-complexes, the final proportion depends on the reaction conditions as reported by Volkovicha, *et al.* (2003). The Mo(V) content can be separated from the melt under vacuum as $MoCl_5$, and because of its low boiling point, T_M: 194°C and T_B: 268°C (see Table 3) can be vaporised from the molten NaCl–KCl.

Similarly, technetium can also form low valency polymeric and/or cluster chlorides (α,β-$TcCl_2$, α,β-$TcCl_3$, $[Tc_2Cl_8]^{2-}$) in the chloride melts as well as volatile $TcCl_4$ depending on the concentration of chlorine gas and temperature [50]. Because of its low boiling point: T_B: 300°C technetium tetra-chloride can be separated from the melt.

Subsequently, the uranium separation may be suggested by oxidising U(III) in U(IV), U(V) and U(VI) e.g. using Cl_2 diluted in Ar. Consequently, the part of UCl_3 present in the molten salt may be oxidised according to

$$UCl_3 + (n-3)/2Cl_2 = UCl_n$$

If it does not already happen in the reactor due to the chloride ions or atomic chlorine set free from the fission reactions. The free chloride ions or atomic chlorine results from the formation of elements with lower valance states, e.g. $BaCl_2$ and Kr. The Na(Rb)-UCl_4 binary mixture of the main components is comparably easy to separate by vaporisation of UCl_4 that evolves from the molten salt and crystallizes to enable it to be recuperated for reutilisation in the melt.

Table 3. Comparison of the phase transformation (M: melting, B: boiling) temperature for increasing valence state of chlorides Data from Yaws (2015) [52].

$FpCl_x$,	T_M (°C)	T_B (°C)
NaCl	801	1465
RbCl	718	1390
AgCl	455	1547
CsCl	645	1297
$SrCl_2$	873	1249
$MoCl_2$	530	1427
$CdCl_2$	564	960
$SnCl_2$	247	623
$BaCl_2$	962	1560
$SmCl_2$	855	1950
$EuCl_2$	731	2190
YCl_3	721	1507
$ZrCl_3$	627	330
$RhCl_3$	450	717
$InCl_3$	497	586
$SbCl_3$	73	220
$LaCl_3$	858	1000
$CeCl_3$	817	1727
$PrCl_3$	786	1710
$NdCl_3$	758	1600
$PmCl_3$	737	1670
UCl_3	837	1657
$PuCl_3$	760	1793
$ZrCl_4$	437	331
$MoCl_4$	552	322
$TcCl_4$		300
$TeCl_4$	224	387
UCl_4	590	791
$NbCl_5$	205	254
$MoCl_5$	194	268
UCl_5	287	
UCl_6	177	527

The separation includes first $MoCl_5$ followed by $TcCl_4$, $ZrCl_4$ and $TeCl_4$, and, finally by UCl_4 with $AnCl_4$ ($NpCl_4$, $PuCl_4$ and $AmCl_4$), where the actinides should not be separated. Thus, a careful control of the processes and the related temperature will be required. Purification can also be carried out during condensation and crystallization.

Separation by electro-reduction

For electro-reduction, specific voltamogramms have already been recorded for example for uranium in LiCl-KCl by Masset, *et al.* (2005) [51] and for neodymium in LiCl-KCl also by Kim & Lee (2020) [53], thus the data is already available in the community. They clearly display the waves due to the electro-deposition of U^0 and of Nd^0.

The electrodeposition of U^{3+} would be the first to take place according to the voltamogram. The electrodeposition potential is −1.5 V (vs Ag/AgCl at 430°C) for the reaction, which highlights the requirement to oxidise the U^{3+} as described above to avoid the reaction:

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$U^{3+} + 3e^- \Rightarrow U^0$

Which is similar to the E° of the $U^{3+/0}$ couple: 1.68 V (vs Ag/AgCl) (see Figure 6). For Nd^{3+} the electroreductions start by the production of Nd^{2+} at -1.7V followed by the electrodeposition of Nd^0 at -2.2 V (vs Ag/AgCl).

$Nd^{3+} + 1e^- \Rightarrow Nd^{2+}$

$Nd^{2+} + 2e^- \Rightarrow Nd^0$

The electrodeposition of U can be discriminated from that of Nd in the case of a non-successful U^{3+} to U^{4+} transfer before the invitation of the electroreduction.

2.5. Opportunities for improved waste management

The above described changes are the result when analysing the challenges with a singular view. Changing to a more global approach and taking forward the demand driven thinking following the vision of zero waste nuclear , it would be reasonable to ask what would be needed to either significantly reduce the final disposal challenge, or in the best case to make a final disposal of the currently planned form obsolete.

Defining the demand by understanding the challenges in a much more open discourse in contrast to the final disposal concepts that have partly been defined in the 1980s but based on even earlier plans [55, 56]. These concepts have mainly been driven by the waste forms provided by the reactors and the reprocessing technology developed at the same time. Understanding the demand will allow the development of the relevant processes in the most appropriate way to support tailored final disposal solutions e. g. through the specific separation of elements which contain long lived isotopes, or elements which are carrying the isotopes responsible for the major decay heat production. This can be delivered in the same way as identifying the elements which have the major influence on the reactor long term operation [18, 57]. This approach has identified elements to be separated through the salt clean-up system e. g. Ru, Mo, Pd, Cs, Nd, Tc [57] using the processes described above while most other fission products will be kept in the salt as long as the solubility limits allow. Adopting a study based on final disposal requests will support the discussion. It will help to understand the effect of the separation of specific elements and their optimized conditioning on a future strategy for final disposal. In addition, some of the volatile fission products elements, e.g. Iodine, or noble gasses will naturally bubble out of the salt and will have to be captured in the off-gas treatment and conditioned out of this waste stream.

Recycling of fission products for alternative end use applications

Reverse reprocessing' based on the processes described in potential clean-up approaches for managing undesirable fission products will deliver clean elementary streams of materials. This will allow reuse of fission products in alternative end use applications. Some initial research on separation of noble metals and Technetium demonstrates that there is initial growing interest [58, 59, 60]. Other opportunities would be to investigate into the rare earth elements and noble metals even if they will contain radioactive isotopes. In addition, India is already planning on the reuse of nuclear waste materials “Main fission products like Cs-137 & Sr-90 present are recovered using in-house developed technologies and deployed for societal applications covering medical applications, external irradiators and other medical applications.”[61].

In general, the fission products are produced in comparably small amounts, roughly 800 to 900 kg per year in a reactor delivering a power of 1 GWe. Table 4 gives a list of fission and activation products to be expected with the concentration of the different elements after a full operational time of ~30 days without any clean-up. The percentages in the third column have to be seen as a share of the overall 800 to 900 kg of fission products produced in one year to facilitate future discussion of the potential use for other potential applications instead of directly forwarding these elements to a final disposal. Some of the elements will have to be separated due to criticality issues, but other elements could be separated in case there is a path that can be developed for further use.

Table 4. Specific element content in the fuel salt of a molten salt reactor based on 42.5 % NaCl – 17.25 % UCl_3 – 40.25 % UCl_4 fuel at a burnup of 100 GWd/itHM, structured by the top elements by concentration, the top of the rarest metals produced, to noble gasses, the rare earth elements, and other notable elements all with a concentration near to or greater than 1 ppm. ^{x}S is formed as an activation product of Cl^{35}, while a portion of H forms as a portion of the activation of Cl^{35}.

Top five activation and fission products	Concentration (ppm)	Percentage fraction of the activation and fission products
Zr	4526	13.1
Mo	3934	11.4
Xe	3736	10.8
Nd	3155	9.11
Cs	3093	8.93

Top four rarest elements	Concentration (ppm)	Percentage fraction of the activation and fission products
Ru	2562	7.40
Pd	1184	3.42
Rh	705	2.03
Te	425	1.23

Noble gases	Concentration (ppm)	Percentage fraction of the activation and fission products
Xe	3736	10.8
Kr	497	1.43
Ar	217	0.63
He	145	0.42
Ne	3	0.01

Lanthanides above 1 ppm	Concentration (ppm)	Percentage fraction of the activation and fission products
Nd	3155	9.11
Ce	1979	5.71
La	1010	2.92
Pr	913	2.63
Sm	924	1.98
Y	600	1.73
Pm	89	0.26
Eu	56	0.16
Gd	33	0.1
Tb	1.35	0.004
Dy	0.96	0.003

Other notable elements above 1 ppm	Concentration (ppm)	Percentage fraction of the activation and fission products
Ba	1336	3.86

Sr	1072	3.10
Tc	900	2.60
H[x]	657	1.90
Rb	473	1.37
I	182	0.53
S[x]	99	0.29
Se	79	0.23
Ag	74	0.21
Cd	60	0.173
Sn	58	0.167
Sb	15	0.04
Nb	9	0.03
In	5	0.015
Ge	1	0.003

In any of the opportunities, the volumes of the solid fission products to be separated and conditioned would be very limited, thus other opportunities than non-retrievable deep geological repository could be envisaged, thus should be researched and discussed.

Applying and researching these new opportunities including exact determination of the masses of different elements to be expected, combined with a plan for future amounts of 'energy production' from nuclear energy, and potential conditioning technologies, would open a new approach to deliver at least partly on the vision of zero waste nuclear. This should lead to an approach, which should be much closer to other technologies where the primary solution is to find a reasonable use for the by-products instead of defining these products as waste without considering other approaches first, like the reuse of e. g. Ruthenium, Niobium/Neodymium in magnets or maybe Radio-isotopes for therapies. This approach is used in other industries. In the area of industrial chemistry, the worldwide market for noble metal catalysts is expected to reach $17 billion by 2026 [62]. Catalytically active by-products include Rb, Nb, Ag, Ce, La, Ba and Sr. With nuclear energy being viewed as a potential energy source for hydrogen and synthetic fuel (and chemical) production, there would be an opportunity to re-use the by-products from nuclear fission to drive downstream processes, including the reverse water-gas shift reaction for synthetic hydrocarbon production from atmospheric CO_2. With the potential that these processes could occur on the same site or adjacent to the reactor, the mechanics of re-sue would be simplified and even the use of radioisotopes cannot be ruled out given that the site would already be licensed to handle radioactive material.

Researching these opportunities in parallel to research and studies on the classical deep geological disposal should open new ways in the handling of materials which we currently tend to call nuclear waste. Using the described approaches could bring the old dream of the P&T community, the 500 year final disposal, into reality and the world closer to the vision of zero waste nuclear. There may be a way to tackle the problem via a completely different approach. We could separate out some materials with short half-lives of typically around 30-50 years, which could be almost completely decayed after more than 10 half-lives within 500 years. We can then take some small quantity of well-conditioned long-lived elements with a very low radiation/activity level that could be safely stored while we could research their potential re-use [63].

3. Conclusions

Nuclear technologies have a great potential to contribute to future low carbon societies and at least some governments have decided that nuclear should play a significant role in their low carbon strategies. In today's world most of these strategies are built on using light water reactors. However, when thinking about new, advanced reactor technologies, there is a new, very important tendency, at least formulated in the UK and discussed in the academic community and authorities – the aim is not only to design a future reactor, but also to consider where does the fuel will come from, as well as where the fuel is going to be disposed of? This has motivated to look into more integrated system development, instead of concentrating only on very focussed reactor development. This widened view is essential to understand the opportunities of an innovative, highly integrated nuclear system. It is given in three the steps: reactor, fuel cycle, waste.

Optimizing the reactor part through innovative thinking has led to the strategic decision to set on molten salt reactors with fast spectrum to assure sufficient breeding to allow operation on spent fuel only without prior reprocessing. This requirement led to the choice of a chlorine based molten salt which allow an exceptionally high load of fertile material to support the required breeding.

The highly integrated fuel cycle from cradle to grave all incorporated into a reactor system leads to the next step of optimization. Due to the specifics of the molten salt reactor it will be possible to operate on spent fuel from LWR without prior reprocessing, since the anyway required salt clean-up system can be designed in a way that it can deal with this extra amount of fission products inserted. The integrated fuel cycle will take advantage of the absence of the traditional pre and postprocessing required for solid fuelled reactors (cooling and dissolution of solid fuel and solid fuel production). The salt clean-up will be based on ‘reverse reprocessing’ (separating fission products form the fuel stream) which will help to avoid proliferation discussion since no fissile material will be separated. As side effect the new processes will open new opportunities in the future waste management.

The waste production in the form of fission products for any nuclear reactor is more or less proportional to the amount of energy delivered. However, the proposed reverse reprocessing has the potential to deliver massive, new opportunities for the final disposal. It delivers in contrast to aqueous reprocessing clean streams of separated fission products instead of ‘the soup’ containing a wide mix of different elements. The analysis has shown that the radionuclide inventory and materials, will change since only the separated fission products will have to be stored while all fissile and fertile materials stay in the reactor until they are completely burnt. This will lead to a significant reduction in the volume of waste, even if some new form of process waste will potentially be produced. The thermal power of the waste to be disposed can be brought down to extremely low levels, since the major contributors to the decay heat production can be separated and stored in a cooled environment instead of being part of the general waste stream. This should allow significantly denser packing of the waste canisters. Finally, the separated elements will allow to develop and apply specific conditioning methods instead of a compromise for a wide mix of elements. This will help to increase the durability of the waste in the specific repository environment. All the proposed changes will have a significant effect on the repository safety, maybe this will even lead to a massively changed set of requirements for the disposal – this should be investigated in the future with an open mind to harvest as many opportunities as possible to come as close as possible to the vision of zero waste nuclear. In addition, this new approach has the potential to lead to clear reduction of the repository cost helping to deliver a more efficient way of dealing with the residual products of nuclear energy generation – again dimension of the changes are on such a wide level that more detailed investigations will be required for the future to understand all impacts in detail.

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