

The Current Status of Partitioning & Transmutation and How to Develop a Vision for Nuclear Waste Management

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Introduction The waste management strategy of partitioning and transmutation is currently the cutting edge development of nuclear technologies, which has been intensively researched over the last several decades, as it is highlighted in the following two excerpts on the history of partitioning and transmutation given below.

“Nuclear Partitioning and Transmutation is one of the most promising fields of nuclear technology. We expect that partitioning and transmutation technology would contribute to the enhancement of the efficiency of high-level waste disposal and the utilization of resources in the spent fuel. We believe that the basic research and development effort in this field would be beneficial for the future generations although it is not quite an alternative to the present back-end policy” [1]. These are the words of T. Yamamoto of the Atomic Energy Bureau Science and Technology Agency, taken from the welcome address at the First International Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation (IEM P&T) 1990 in Mito, Japan. This meeting initiated a series of exchange meetings organized by the Nuclear Energy Agency (NEA), with the 15th Information Exchange Meeting recently held in Manchester, UK. The NEA is an agency, specialized in the support of the development of nuclear systems, within the Organization for Economic Co-operation and Development (OECD).

“One of the greatest challenges in the use of nuclear energy is the highly radioactive waste which is generated during power production. It must be dealt with safely and effectively. While technical solutions exist, including deep geological repositories, progress in the disposal of radioactive waste has been influenced, and in many cases delayed, by public perceptions about the safety of the technology. One of the primary reasons for this is the long life of many of the radioisotopes generated from fission, with half-lives on the order of 100,000 to a million years. Problems of perception could be reduced to an essential degree if there were a way to burn or destroy the most toxic long-lived radioactive wastes during the production of energy” [2]. These are the words of Victor Arkhipov a consultant in the IAEA division of nuclear power and the fuel cycle, in the nuclear power technology development section which highlights the importance of the topic for the future acceptance of nuclear technologies, published in the IAEA Bulletin number 39 in 1997.

History of Partitioning & Transmutation

The discussion of partitioning & transmutation of nuclear waste, mainly concerning transuranic nuclides, is a phenomenon which received a large amount of attention in the late 1980's and early 1990's. The increasing importance of the topic is reflected by the creation of the Information Exchange Meeting on P&T organized by the NEA every second year, as outlined in the previous section. The title of the meeting ‘Exchange Meeting on Actinide and Fission Product Separation and Transmutation’ already points out that the vision of P&T is much larger than just recycling and burning Plutonium. However, the first experiments to insert minor actinides containing fuel

into nuclear reactors to burn these isotopes are much older. *“In the 70's, minor actinide (Np, Am) containing mixed oxide fuels were designed and successfully irradiated in fast reactors: KNK II and PHENIX. The composition of the fuel covered the homogeneous as well as the heterogeneous recycle of minor actinides”* [4].

The first strong push to the technology of partitioning & transmutation (P&T) of transuranium isotopes has been given by two important international projects: the OMEGA program in Japan and the CAPRA/CADRA project in France. The OMEGA program, launched in 1988: *“In addition, the Japan's Atomic Energy Commission submitted in October 1988 a report entitled “Long-Term Program for Research and Development on Nuclide Partitioning and Transmutation (P&T)”, from the viewpoints of conversion of HLW into useful resources and its disposal efficiency. The program plots a course for technological development up to the year 2000 and is called “OMEGA” which is the acronym derived from Options for Making Extra Gains from Actinides and fission products”* [5] is the first major project related to P&T. Shortly after the OMEGA program, the CAPRA/CADRA project started in France in the begin of the 1990's: *“CAPRA/CADRA was created in response to the 1991 French law which mandated a 15-year research programme to investigate the technical options available for the nuclear fuel cycle in France”* [6] with the aim to investigate future opportunities to apply fast reactors for the burning of excess plutonium. In contrast to the Japanese OMEGA project focused on partitioning as well as on transmutation, the French CAPRA/CADRA project was only dedicated to transmutation, with a major focus on the reuse and incineration of Plutonium in Superphenix. *“The potential of fast reactor systems to burn plutonium and minor actinides (MAs) (Np, Am, Cm) is studied within the CAPRA/CADRA program (Barré, 1998)¹. CAPRA mainly deals with managing the plutonium stockpile and CADRA is related to the burning/transmutation of MAs and long-lived fission products”* [7]. The next major step in Europe was the launch of the Integrated Project EUROTRANS as a part of the 6th EU Framework Program (FP). *“Among the prior research and development topics of EURATOM 6th Framework Programme is the management of high-level nuclear wastes. In particular, the development of technical solutions of nuclear waste management is considered important”* [8]. IP EUROTRANS has been followed by a large number of smaller projects in FP 7 and now in HORIZON 2020. The focus is on specific problems related to partitioning and transmutation or integrated into projects with a wider focus like fast reactor development.

A very specific project has been undertaken in Germany following the nuclear phase out decision taken in 2011. The Federal Ministry for Economic Affairs and Energy

¹Barré, B., 1998. The Future of CAPRA. 5th Int. CAPRA Seminar, Karlsruhe

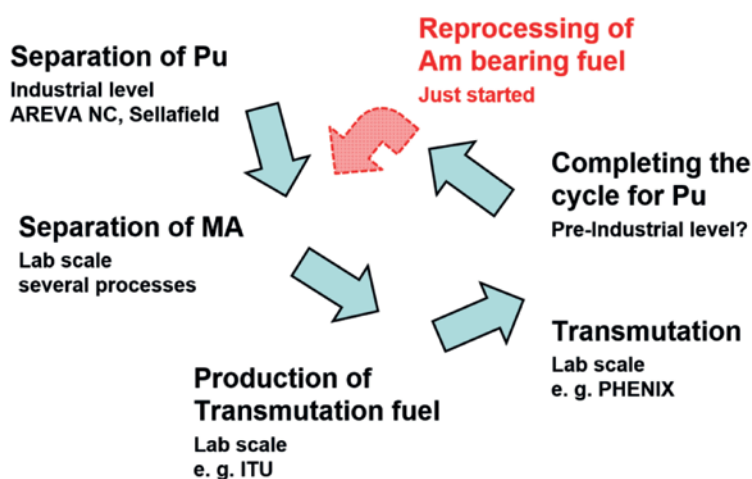


Fig. 1. Process steps of Partitioning & Transmutation as applied for the demonstration of the technology.

(BMW) and the Federal Ministry of Education and Research (BMBF) launched a comprehensive study managed by the National Academy of Science and Engineering (acatech) to create a robust scientific basis for an evidence based approach on the future of the P&T research under the boundary conditions of the nuclear phase out. The study consists of two parts: the first part is concerned with technological issues („Studie zur Partitionierung und Transmutation (P&T) hochradioaktiver Abfälle“ [9]) while the second part is concerned with an evaluation of chances and risks of the technology to the society (“Gesellschaftliche Implikationen der Partitionierungs- und Transmutationsforschung” [9]). The information collected in the German study on P&T [10] created the basis for the acatech POSITION “Partitioning and Transmutation of Nuclear Waste. Chances and Risks in Research and Application“ (“Partitionierung und Transmutation nuklearer Abfälle. Chancen und Risiken in Forschung und Anwendung” [11]). The results of this study, as well as the acatech POSITION have been presented at both national [12] and international levels [13].

At the last International Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation (IEM P&T) in Manchester in 2018, Russia announced its commitment to P&T [23]. Their approach will be based on aqueous reprocessing with downstream minor actinide separation in the Mayak facility and transmutation in homogeneous or heterogeneous mode in the sodium cooled fast reactor BN-800. This will incorporate investments into scaling of BN-800 spent nuclear fuel (SNF) management process (transportation, storage and reprocessing) in the first approach, complemented with the Brest-OD three stage development: 1) the construction and commissioning of a nitride fuel production facility; 2) the construction and commissioning of the reactor itself; and 3) the spent nuclear fuel reprocessing facility. In the same time, the long-term research will focus on minor actinide burning in molten salt reactors.

State of the Art

Partitioning & Transmutation cannot be seen as a single process. In fact, it is a whole chain of interlinked processes which have to be worked through in a cyclic way, see Figure 1. The whole chain of the process has been followed for the successful lab scale demonstration of the feasibility of Partitioning & Transmutation using French facilities

for reprocessing, fuel production and the sodium cooled fast reactor Phenix [14]. However, it has to be kept in mind that almost each of the steps of demonstration is based on technologies which have been available at this time, even if developed for another purpose – the production and the separation of Plutonium.

The introductory step into the technology is what is currently known as aqueous reprocessing amended with some downstream processes to recover Americium. The discussion about reinsertion of Curium into the reactor has been at least postponed due to the challenges in the radiation protection during fuel production. The opening step: classical aqueous reprocessing of LWR fuel, is currently commercially performed on industrial level in at least three facilities, in France at La Hague, in Russia in Mayak, and in the THORP facility at the Sellafield site in the UK which has just been finished operation. There have been several downstream processes and advanced reprocessing flow sheets developed on lab scale. The down selection of possible processes for a future industrial application is currently part of the GENOIRS Horizon 2020 project [20]. Different styles of transmutation fuels have been produced, irradiated, and examined in IP EUROTRANS and several follow up projects [21, 22]. However, all these processes have only been demonstrated in laboratory scale and even the production of mixed oxide fuels (MOX) with plutonium loadings required for fast reactor operation has never been brought from the pre-industrial level used for the fuel production of Superphenix to the industrial level. New Russian studies [19] on the expected cost and the cost structure of closing the fuel cycle point out that the production of high loaded MOX will be a major cost driver. The study indicates that the front-end cost (fuel production) versus back end cost (reprocessing) will be shared ~75% to 25% based on the application of currently available technologies in Russia. The cost of Americium bearing fuels will be even higher due to the lack of experience. However, cost is only one of the challenges. Another important point is addressing the high radiation exposure [23] associated with the production of Americium targets (i.e. fuel rods with a high Americium content). It will require fully remote (automated) fuel fabrication and handling technologies due to an increase of the radiation exposure from the fuel assembly by a factor of ~100 compared to standard fast reactor MOX fuel, based on data given in [23].

Transmutation of transuranic (TRU) isotopes like Plutonium, Americium or Curium could theoretically be achieved in any kind of nuclear reactor where a high enough neutron flux is available for efficient burning. However, studies have shown that the efficiency of the transmutation process depends significantly on the amount of fast neutrons available in the specific reactor. “The fission/absorption ratios are consistently higher for the fast spectrum SFR. Thus, in a fast spectrum, actinides are preferentially fissioned, not transmuted into higher actinides” [26]. This is due to the strong energy dependence of the relation between the fission and the absorption cross sections. This relation thus depends strongly on the average neutron spectrum available in a specific nuclear reactor. The mentioned fission-to-absorption ratio describes the probability of a desired fission event – which will destroy the TRU isotope, compared to an undesirable absorption event – which will only lead to breeding of a heavier TRU isotope. Studies have shown a significant difference between the PWR and the SFR, with regard to transmutation efficiency [26]. In the PWR the main

isotopes which have a high fission-to-absorption ratio are U-235, Pu-239, and Pu-241. Thus these isotopes have a high probability to undergo a fission reaction. Typical other isotopes (Np-237, Pu-238, Pu-240, Pu-242, Am and Cm) indicate a low or very low fission-to-absorption ratio, thus these isotopes have a high probability to undergo an absorption reaction leading to a breeding process. In the fast neutron spectrum, almost all transuranium isotopes show a significantly higher fission-to-absorption ratio than in thermal reactors. This demonstrates that a fast neutron spectrum reactor, is essential to achieve reasonable transmutation rates for TRU isotopes.

Different kinds of fast reactors can be dedicated for the transmutation of TRU isotopes. Up to now only sodium cooled fast reactors have been envisaged and used for the demonstration of closing the nuclear fuel cycle as well as for the demonstration of transmutation, due to their availability - mainly the Phenix reactor in France – and due to the experience in the operation of this reactor type [14]. However, two problems have to be highlighted: 1) It would be necessary to demonstrate an industrial level of fast reactor operation, employing a closed fuel cycle whilst applying transmutation, but these efforts have received a significant setback due to the decision to delay the industrial demonstration of SFR technology to 2080 in the ASTRID project [25]; and 2) The insertion of transmutation fuel, mainly a high amount of Americium, will have a significant effect on reactor stability and thus operability. *“Increasing the minor actinide content [in a SFR core] enforces the positive coolant temperature and sodium void effect. Additionally, the absolute value of the negative Doppler effect and the delayed neutron fraction are decreased as well as the melting temperature of the MOX fuel. These changes degrade system safety, making the enhancement of the feedback effects mandatory for a transmutation system to enable it to attain the safety characteristics comparable to those of a classical SFR”* [14].

The first step which would be required to demonstrate P&T technology at an industrial level by completing the cycle for Pu, thus demonstrating closed fuel cycle operation, since plutonium forms more than 90% of the transuranic nuclides which are produced during LWR operation. However, due to the reconfiguration of the ASTRID project, the objective of industrial demonstration pertaining to closed fuel cycle operation, has been postponed until 2080. This will create uncertainties related to deployment due to the unavailability of any fast reactor irradiation facility for future steps related to the demonstration of Americium incineration in Europe. This delay will impact on the reprocessing of Americium bearing fuel/targets, too. There are two limitations, firstly there is no driver for the development and the manufacturing of Americium bearing fuel when it cannot be irradiated, and secondly there will be no irradiated fuel available for hot lab tests to develop the technology required for the specific fuel form which is not yet decided for.

To summarise, it becomes clear that the demonstration of P&T technology at the lab-scale has been deployed successfully. However, most of the steps have been based on technologies which have not been designed for the purpose. The demonstrations have been based on alternative application and extension of existing equipment like aqueous reprocessing and the sodium cooled fast reactor PHENIX. A major question for the future is how to proceed with the application of P&T technology at the pre-industrial and later industrial level. One should recognise the enormous challenges moving from small

scale to large scale particularly in applications where the uncertainties and the required investments are high. In this case we cannot predict accurately enough how the performance at scale will be influenced by small imperfections in the technological solution at the lower scale. Here the lack of a credible demonstrator at scale is laid bare. This leads directly to the major question for the future: Is it the right way to base the future development on existing technologies and solve the massive challenges or should cutting edge research in nuclear be focused on developing a system specially dedicated to the requirements of P&T?

To answer this question, we need to have a more holistic view of the socio-economic costs of each proposed solution. This is a much greater problem than a business deciding what product to bring to market. Here we have to pose the question – how much society is willing to ‘loose’ (in the form of an unattractive investment) to make a problem safe or to avoid another ‘imperfect’ solution which is seen as a worse choice by the society. P&T needs to beat the next best option for the problem to be viable.

Key Challenges

The following **Figure 2** is used to collate the key challenges of P&T based on the current technologies, which are:

- The challenge in partitioning relates to the demand for very high recovery rates for the TRUs requiring multi-stage processes. However, these recovery rates are essential to avoid a carryover and accumulation of TRUs in the waste stream which would reduce the effects of P&T on the final disposal. In addition, reprocessing and especially the downstream processes form a costly challenge both for the development as well as for the application.
- The challenge in solid fuel production is due to the very high cost and significant increase in radiation levels during fuel production and handling. The high radiation levels will require remote handling and manufacturing technologies [23].
- The challenge in transmutation is the requirement for a solid fuelled fast reactor where the right balance between efficiency (requiring a high Pu and minor actinide content in the core) and the effect of the TRUs on reactor stability [27]. In addition, the currently high cost of fast reactor technology and the relatively limited experience in operating these reactors at demonstrator level also create further uncertainties.

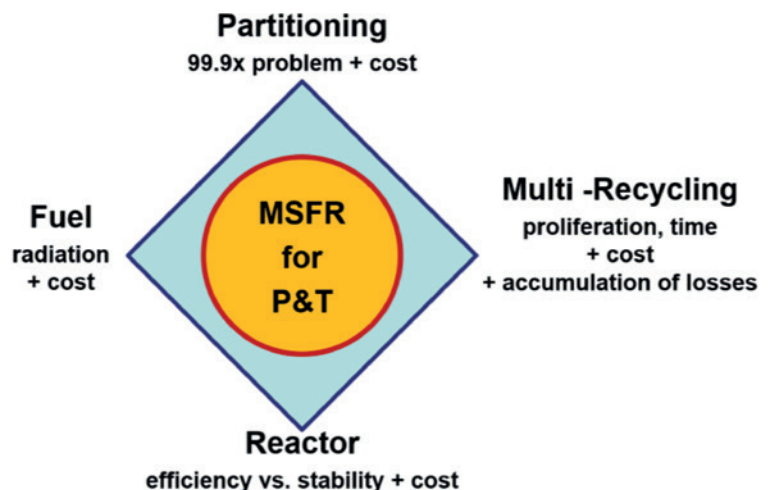


Fig. 2. The reverse quadrature of the circle or P&T between today and tomorrow.

- The challenge of the whole cycles lies in the request for a multiple recycling scheme since only a part of the TRUs or minor actinides can be transmuted during one operational cycle of a fuel assembly. The multi-recycling requirement will demand to run through the whole costly system in a multiple way accumulating cost and time while creating a huge number of handovers with each handover creating a proliferation risk. However, there is another limiting factor which has a consequence for the whole system, this is the accumulation of losses which depends on the one hand on the losses in the partitioning and the fuel production and on the other hand on the efficiency of the transmutation [24]. Thus the multi-recycling request poses high challenges onto the separation chemistry and fuel production as well as on the reactor design. In addition, multi-recycling will lead to a significant mass of fuel being resident in the fuel cycle due to the requested times for cooling, reprocessing, and fuel manufacturing.

Based on the mentioned challenges, it is important to at least consider other advanced technologies and proposals relating to how P&T technology could be made accessible without requiring a costly engineering solution for the above challenges. Recently, two different approaches have been proposed in the acatech POSITION [11] which has been developed in the aftermath of the acatech P&T Study [10]. The proposed solutions are either the application of accelerator driven systems (ADS) for transmutation or the application of molten salt reactor technologies for the whole P&T process.

In our view the application of ADS systems for transmutation has the potential to improve the transmutation of TRUs since due to the promised enhanced neutron physical reactor stability and the external neutron supply, a higher load of TRUs is possible which has the potential to improve the efficiency of the transmutation as well as to reduce the number of multi-recycling cycles. However, this approach tackles only the reactor part of the quadrangle and effects slightly the multi-recycling. A more comprehensive view onto the problem with the focus of avoiding most of the challenges leads to the application of molten salt reactors for P&T [15, 16, 17] since a liquid fuelled reactor offers the opportunity to integrate major parts of the fuel cycle into the reactor while avoiding some of the major challenges associated with solid fuel production. Taking into account all advantages of a liquid fuel technology associated with molten salt reactors, leads to the recognition that the molten salt fast reactor approach has the potential to eliminate some of the most costly steps of the P&T technology. If a molten salt reactor would be used, solid fuel production, one of the major cost drivers as well as one of the major time consuming research challenges would be eliminated and replaced by another process. The approach focuses research into another technology that may offer a superior solution based on current research priorities in nuclear technology. One of the keys to making of MSR technology available would be to produce nuclear fuel in the salt phase like, as is already required for the advanced reprocessing relying on pyro processes. Pyro processes are already under research in several countries, including in the UK as part of the REFINE project [28].

The challenges in the reactor are partly reduced since a homogeneous system is employed with the fuel dissolved within the coolant. These systems provide much stronger feedback effects than solid fuelled fast reactors since in the case of an appropriate design, the amount of fissile

material in the core is reduced when the density is reduced due to temperature increase which will allow a significantly higher loading of transuranic nuclides as long as the solubility of the transuranics in the carrier salt can be assured. In

addition, there is neither an inhomogeneous core composition like it would be the case in heterogeneous Americium burning using targets nor a problem with the fission gas accumulation in the fuel pellets and rods which limits the burnup of minor actinides due to the pressure increase in the fuel rods. However, due to the release of the fission gas directly in the liquid fuel most probably a reliable off-gas treatment with the demands on a safety grade system will have to be operated.

The challenges of the multi-recycling will be eliminated completely in a molten salt reactor, since in these kinds of reactors, the salt has to be cleaned in an online process. This means a small share of the salt will be continuously withdrawn from the operating reactor. This stream will be cleaned from the fission products which prevent the reactor from achieving long term operability and the cleaned stream will be fed back into the reactor. This reduces the proliferation risk significantly if the processes are designed in an appropriate way since the fissile material will stay in the solution, the plutonium quality will always be reduced due to the mixing of all materials and the handovers of fissile material are eliminated. However, it has to be mentioned that the required clean-up processes have to be developed following a completely new approach, instead of the separation of fissile materials like in the conventional reprocessing, the new processes have to be designed to separate specific isotopes like Neodymium and Caesium which cause almost 50% of the effect on criticality, with Zirconium and Samarium causing further 30% of the fission product effect on criticality [29]. Nevertheless, a successful design of the salt clean-up system will eliminate the step of partitioning of fast reactor fuel completely since there is no demand to separate fissile material to produce new, clean fuel. In contrast, all material which is foreseen for transmutation will stay in the reactor until the nuclei have undergone a fission process.

The manuscript has been opened with the remark that P&T is the cutting-edge research topic of nuclear technology and that the lab scale demonstration of the technology has been accomplished successfully. In general, if we intend to get the technology into an industrial application, which will be required if we intend to reduce the long term challenges in final disposal, a significant amount of research and development work will be required [11]. It is now on the community to define the ideal strategic approach which will in our view require a careful evaluation of the available approaches to identify the ideal approach. All approaches require a strong development demand to achieve industrial application and the research community will have to face a wide set of new challenges. However, it will be more promising to invest into tailored approaches to solve existing problems with innovative approaches which are ideally based on the application of already existing skill sets (e. g. nuclear chemistry, advanced fluid dynamics) taking advantage of technological approaches which are already available or under development in other technologies (e. g. pyro-reprocessing for fuel production, or off-gas cleaning like in reprocessing facilities).

Before entering into the next step of development the community should undergo a strategic development

process for demand driven research planning [31] with an unbiased analysis of different options before taking major investments into R&D. Getting P&T into industrial application is a very long, time consuming mission requiring a massive investment even when we would rely on existing technologies. Why not focus on an improved solution to direct the future R&D into a more innovative dimension which promises some clear benefits. The focus should lie on creating innovative, demand driven solutions which on the one hand have the potential to provide new IP and market opportunities while promising the elimination of some major challenges which would require very costly engineering solutions. The focus on disruptive solutions has the potential to bring more innovation into nuclear technologies which have suffered from a lack of ground breaking innovation in the last several decades. The proposed application of molten salt reactors for transmutation is not a singular view of the authors, see MA burning in molten salt reactors proposed by Russia [29] and early proposals to use molten salt reactors for the burning of weapon grade plutonium in the 1990's [33].

A Possible Outlook

All thoughts mentioned up to now have focused on the issue of developing the P&T technology as a waste treatment technology separate from the development of power reactor technology. There are currently clear reasons for this approach since the current reactor fleets worldwide are based on light water reactor technology while P&T will require fast reactor technology. However, a slight change in the approach to the problem has the potential to create a new way of thinking. We currently tend to look at the spent nuclear fuel (SNF) of light water reactors as waste to be stored safely instead of the common approach of other industries when developing production chains – ‘up to now, this is a material we haven't found the ideal use for’. On this basis, we have to see SNF as a resource, a potential energy source for the future. The same approach can change the recognition of the fission products, a resource we have to find a better solution or use. Taking this alternative view and looking into the future of nuclear hopefully based on closed fuel cycle operation creates the opportunity to bring the major nuclear technologies – power production and waste management – closer together. The last consequences could be the recently proposed operation of molten salt reactors directly on spent nuclear fuel originating from light water reactors without prior reprocessing [30, 31, 32]. This approach has the potential to eliminate the very costly pre-step into closing the fuel cycle which has all the time been accepted as unavoidable – the reprocessing of the LWR fuel – thus eliminating one of the most challenging and costly hurdles to get closed fuel cycle operation into future application. Thus, this new approach can ideally provide society with the advantages of closed fuel cycle operation without the massive pre-investments into chemical separation of Plutonium and the related production of mixed oxide fuel with high plutonium content. The approach would allow society to solve P&T as a side effect of a new and disruptive, highly efficient and sustainable nuclear energy system which could serve as a reliable low carbon energy source for the world. The strategic thinking leading to this development and the process which would have to be established will be described in another article in the next edition of this journal [34].

Conclusions

The waste management strategy of partitioning and transmutation (P&T), encompassing reprocessing and reactor technology, is currently the cutting-edge development of nuclear technologies. It has been intensively researched over the last few decades with most effort spent in the IP EUROTRANS program and several follow up projects. Almost all required technological steps have been demonstrated at least at laboratory scale based on existing technologies like aqueous reprocessing with added downstream processes, mixed oxide fuel production and sodium cooled fast reactor operation. However, putting a deep look into the described challenges faced during these demonstrations as well as taking an outlook to the much larger challenges which will appear during upscaling of the technology to industrial scale gives rise to the question, ‘Is the chosen way for the demonstration the right, most efficient way forward or do we need to adopt a much more disruptive approach?? A detailed discussion of the key challenges as well as the evaluation of possible innovative approaches has been given working out approaches which would be required to make P&T an attractive choice for real industrial application.

The discussion leads the way to an innovative, demand driven re-thinking of the whole technological process typical for solid fuelled reactor systems and their related fuel cycle. The outcome of this discussion delivers an approach that avoids the most costly and challenging process steps associated with the solid transmutation fuel production by applying a demand driven technology development using a liquid fuel operated system like a molten salt reactor specially designed for the challenges of P&T.

Finally, overcoming frames of the historic separation of power production and waste management is proposed by applying wider out of the box thinking to improve the attractiveness of nuclear technologies. A disruptive approach of developing a molten salt reactor system with demand driven salt clean-up directly operating on spent nuclear fuel is worked out which will offer the potential to operate a nuclear reactor in closed fuel cycle mode without requiring prior reprocessing as initial step. Applying this approach would allow, to make the large energy amount available which is still contained in spent nuclear fuel, while the requests of waste management using P&T are fulfilled as a side effect. Both points together have the potential to make nuclear one of the most promising answers to the rapidly rising demand for low carbon technologies.

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