A DEMAND DRIVEN WAY OF THINKING NUCLEAR – NEUTRON PHYSICS OF A REACTOR DIRECTLY OPERATING ON SPENT FUEL FROM LWRs

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Abstract. The current generation of nuclear reactors are evolutionary in design. The aims of GenIV form a bridge to future generations of reactors. Here, the aims are extended to encompass the ultimate and universal vision for energy production, the 'perpetuum mobile' – at least as close as is practical. To come as close as possible to this vision, we propose to rethink nuclear reactor design to develop a system which neither requires fresh resources nor produces no fresh waste during operation and continues to generate power safe and reliably in an economic way. The results demonstrate, from a theoretical perspective, that it is feasible to fulfil this vision through the reuse of spent nuclear fuel from currently operating reactors as the fuel for a new reactor. Consequently, there is no burdensome waste production than current spent nuclear fuel which is used as feed to the system. The extended goals which will need to be demonstrated are the safe, reliable and economic operation to create the basis for the long term success of nuclear reactors as a major carbon free, sustainable, and applied highly reliable energy source.

Key Words: Fast Reactor; Spent Fuel; P&T; Innovation Strategy.

1. Introduction

Innovative research for the energy sector should be strictly driven by demand since the customer has no contact to the technology and thus the interest in specific technologies is not very high. The key demand for the future years will be characterized by the UN sustainability goals and the request of reducing the carbon footprint. Nuclear faces in addition the challenge of the increase of the spent fuel pool from light water reactors in most of the countries operating light water reactors. Bringing these two requests together forms the strategic development goals for the future. Already in the 1960s, Everett Rogers described the strategic development of innovations through S-curves and formulated the effects in the theory of diffusion of innovations [1]. He argues that the application of a technology to a market follows an S-curve.

However, Fredmund Malik has extended this thinking in a "Symphony of S-curves: Seeing the Future Clearly" in his book on demand driven strategy development [2] to motivate the people to leave the beaten track of purely evolutionary development when changed boundary conditions require disruptive development. In this case strategic development requires the change to a new S-curve. In a first example this concept will be applied to derive a deeper understanding of the idea of strategic development of industrial innovations. A short excursion will be given on a day-to-day issue with a well-known piece of gardening equipment which is taken from [3]. In the next part the development concept is applied to illustrate the current development of nuclear reactor systems. The historic boundary conditions of nuclear development are reviewed against the already mentioned current boundary conditions. These boundary conditions will be used to propose a reactor system which is as close as possible to the requirements for a worldwide, wide spread future electric energy production using nuclear power. Fulfilling the future requests like reduced CO_2 emission and handling the spent fuel create the basis for the long term success of nuclear reactors to act as a major contributor for the production of reliable carbon free, sustainable electric energy.

There are several good examples for the change from an existing S-curve to one of the next level available, like e. g. the development of the iPhone. Let's consider how Apple developed the iPhone. They started with the Apple Newton – where the required technology was not available to achieve their intended goals – a mobile device for managing your life, communications and entertainment when you are on the move. The product failed in the market place. However, in the next step they introduced the iPod where the technology goals were more limited – they dropped everything other than entertainment - but the product was much more successful. Finally, they introduced the iPhone which did everything the Message Pad tried to do and more and they made billions from it. They invented the move from the S-curve 'mobile telephoning' to the curve 'mobile life' when the technological boundary conditions were given. Of course they weren't the only company doing and trying the same thing but they were the most successful in it in that they became the most valuable company in the world.

The development of nuclear reactors has followed a long evolutionary history of the technical solution as well as the question the technology is trying to address. Thus we propose to address the new demands for a nuclear reactor by moving to a more advanced S-curve due to changed boundary conditions, too.

The Development of Nuclear Reactor Technologies

The development of nuclear reactors started in 1942, with development of "Chicago Pile 1" which was the world's first nuclear reactor, built in 1942 by Nobel Prize winner Enrico Fermi [4]. The development of the first reactors was based around three objectives [3]

1. Demonstration of a continuing chain reaction and its potential application
2. Generation of material for military purposes and/or commercial/medical applications
3. Energy generation from the chain reaction, either as electricity, most desirable, or heat

The main focus of nuclear reactor development is the generation of energy, i.e. objective 3. However, almost all reactor types go back to this early years of development where completely other request have been targeted. We have to think for the future development if these reactors are still the ideal ones for the wide spread nuclear energy production in 20 years. The evolutionary development has been shown already very often in the GEN IV project. Nevertheless, the GEN IV goals are already a strong step towards the future, see Figure 1. The only question is, 'Is this enough for a future success?'

From the point of view of strategic developments in S-curves the GenIV goals build the bridge between the current S-curve and the future one. The current light water reactors are still essentially based on the technology developed to power nuclear submarines. They have undergone an evolution via GenII to GenIII/III+ reactors. They are to be seen as ideal under the currently given boundary conditions for a reliable and robust product to deliver electricity. However, from the point of view of sustainability – ideal use of the fuel – and from the point of waste production – once more going back to ideal use of the given fuel, a clear improvement would be possible. This improvement is typically addressed by proposing the use of fast reactors and a closed fuel cycle. The question is if we could do better in fulfilling the sustainability goal.

Sustainability
- Generate energy sustainably and promote long-term availability of nuclear fuel.
- Minimise nuclear waste and reduce the long term stewardship burden.

Safety and reliability
- Excel in safety and reliability.
- Have a very low likelihood and degree of reactor core damage.
- Eliminate the need for offsite emergency response.

Economics
- Have a life cycle cost advantage over other energy sources.
- Have a level of financial risk comparable to other energy projects.

Proliferation resistance and physical protection
- Be a very unattractive route for diversion or theft of weapon-usable materials, and provide increased physical protection against acts of terrorism.

- Gas-cooled Fast Reactor (GFR)
- Lead-cooled Fast Reactor (LFR)
- Molten Salt Reactor (MSR)
- Sodium-cooled Fast Reactor (SFR)
- Supercritical-Water-cooled Reactor (SCWR)
- Very-High-Temperature Reactor (VHTR)

Figure 1: Generation IV (GenIV) goals (left) and GenIV systems (right) [6]

However, seen from the historic point of view almost all GEN IV systems are based on the very early developments of nuclear when the development objective has been formed with completely different boundary conditions and requests as today – producing nuclear materials and powering submarines versus wide spread sustainable nuclear power production. Important decisions have been made on the way of the development which have been made on the, in this moment, historic requests to answer the given demand. A perfect example is the decision between SFR and the molten salt breeder reactor in the late 60ies [5]. The SFR is advantageous for the production of high quality nuclear material but will it be advantageous for the wide spread sustainable nuclear power production, too? Moreover, our attention should not only be drawn to the reactor – it is, in addition, the fuel cycle. We are still developing the PUREX cycle – plutonium recovery and extraction cycle which has been designed to the demand of recovering the produced high quality plutonium. From energy and waste point of view we are now trapped in the request of exorbitantly high recovery rates since we need the fissile material, but more important we don't want the TRU to be passed to the waste stream.

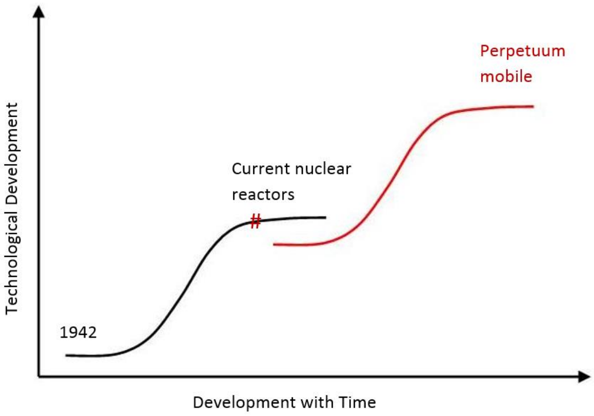

Figure 2: The nuclear reactor development in S-curves

Theoretically, applying the future request of wide spread sustainable nuclear power production, the developing next level S-curve should bring us to the ultimate, universal vision for electric energy production which is independent of the observed ‘power plant‘ system, see Figure 2. The vision is much more advanced and broad than the GEN-IV goals. This ultimate, universal vision can be given with one simple, old phrase – ‘perpetuum mobile’ [3], even if it is clear amongst physicists and engineers, the first law of thermodynamics and energy conservation law prevents such a design operating. Nevertheless, it provides a clear vision for engineers and scientists to drive research and innovation to the right direction – develop a reactor that can breed and burn its own fuel for a significant period of time, and come the vision as close as possible. The key words for such a vision are: no resources requested, no waste produced, highly economic, reliable, secure, and safe [3].

Transferring the idea of the perpetuum mobile to real nuclear reactor operation would fulfil the major request for a long term success of nuclear reactors to become a reliable major carbon free, sustainable energy source. Obviously, no reactor can be operated completely without resources but it would be a smart option to better use already existing resources, the SNF. In this way a reactor could be designed which doesn’t require fresh resources, since it is operated on fuel which already exists in vast amounts. The smart point of this proposal, the reactor will not produce additional waste, since the waste mass should closely match the SNF which is used as fuel. The sustainable long term operation has to be solved within the core physics since such a system can only be achieved if the system can provide enough neutrons for a self-sustained operation on the basis of SNF used. This demand will request a fast neutron spectrum system. Demonstrating the feasibility of the proposed idea a first simulation will follow which is based on a molten salt fast reactor configuration. The potential advantageous safety behaviour and the excellent operational flexibility of molten salt fast systems have already been extensively discussed [7], [8], [9]. The remaining major challenge is in the implementation of such reactor technology which has already been described in several projects [10].

2. Materials & Methods

2.1 Reference Configuration

The calculations are based on the core dimensions and boundary conditions given in the EVOL benchmark definition (see Figure 3), and focus on a reference MSFR of 3000 MWth using a fast neutron spectrum [10]. The spent nuclear fuel composition is calculated with HELIOS 2.1 based on the “OECD/NEA MOX BENCHMARK” [12].

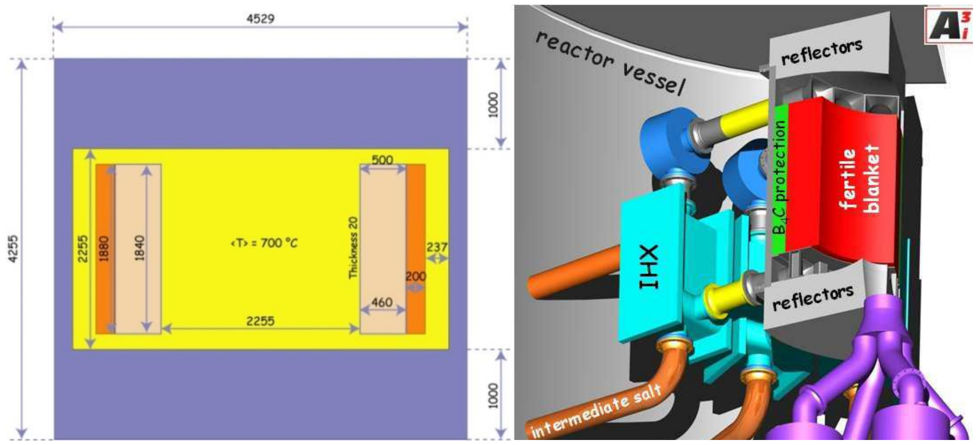

Figure 3: (Right): Simplified scheme of the MSFR system including the core, blanket and heat exchangers (IHX) – (Left): Benchmark definition [10].

The 4.5% enriched UOX fuel was burnt to 50 GWd/tHM the Pu-241 content has been halved and turned to Am-241, representing a postulated storage, no further adaption has been introduced. The TRU isotopic vector given for the feed is (Np-237, 6.3%; Pu-238,2.7%; Pu-239, 45.9%; Pu-240, 21.5%; Pu-241, 10.7%; Pu-242, 6.7%; Am-241; 3.4%, Am-243, 1.9%; Cm-244, 0.8%; Cm-245, 0.1%). The salt configuration is also based on the EVOL benchmark, and consists of LiF with mainly UF$_4$ (calculated as SNFF$_4$ as approximation) in the core. However, in contrast to the EVOL benchmark, the share of SNF has to be determined, to allow for acceptable breeding of fissile material keeping the reactor critical for long term operation without further feeding of fissile material. The blanket region is filled with pure LiF$_4$, with an overall fuel salt volume of 18 m^3 within the core and 7.7 m^3 within the blanket. The salt clean up system provides an additional degree of freedom for the optimization of the breeding.

2.2 Modelling and Simulation Tool

For the model evaluations, the HELIOS 2.1 licensing grade code system is used with the internal 177 group library [13]. The benchmark configuration is transferred to a volume corrected 2D HELIOS model (see Figure 4). The leakage in the third dimension has been fixed by a comparison of 2D and 3D calculations within the EVOL benchmark exercises, with the leakage in radial direction being directly modelled.

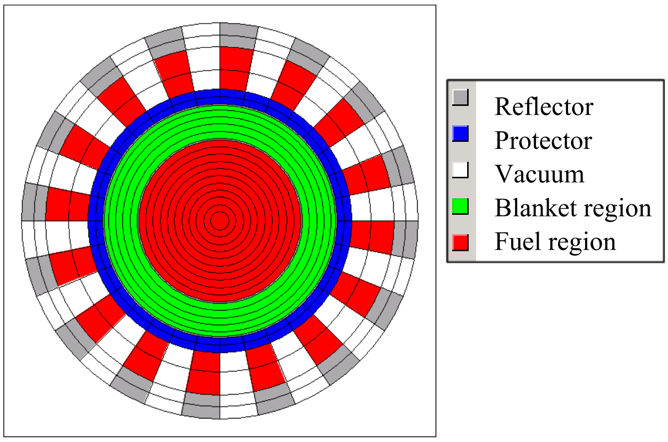

Figure 4: Volume corrected 2D HELIOS model of the molten salt reactor.

Originally, as HELIOS was written for the simulation of solid structured fuel assemblies, the possibility of online refuelling and online salt clean-up was not foreseen. To deal with these special features of molten salt reactors a PYTHON script has been developed.

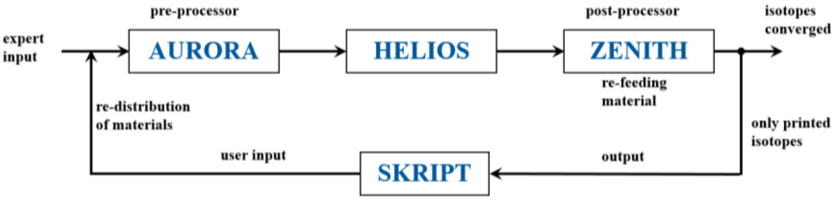

Figure 5: Description of the calculation cycle for the simulation of a MSR.

All information, which is constant during the whole reactor operation, is stored in a so-called expert input, the changing material configuration is given in the user input, and is written new in every 5 GWd/tHM cycle using the PYTHON script to create an updated input for the

HELIOS run, determining the neutron flux distribution and burnup of material. The results are evaluated in the post-processor ZENITH, where it is decided which isotopes will be fed back into the next user input, created with the help of the PYTHON script (see Figure 5). Theoretically, it is possible to simulate a molten salt reactor precisely by using small time steps in this calculation loop. However, in a real MSR two different time scales for salt clean-up can be observed, due to the different extraction methods for fission product removal. To simulate these different time scales a full removal of gaseous and volatile fission products takes place after cycle, but only a partial removal of dissolved fission products is established at the end of cycle. Due to the characteristics of HELIOS, some approximations have to be accepted, e.g. there is no fuel salt movement. HELIOS was designed for use for LWR reactors, but comparisons to SERPENT on the isotope accumulation during the burnup in SFR using different HELIOS libraries have shown an acceptable agreement for major isotopes [14]. The major uncertainties are predominantly given by the current preliminary design. Such design uncertainties are expected to impact significantly the spent LWR fuel configuration and the TRU or Pu feed quality.

3. Results

3.1 Initial Core

The initial core configuration for the simulation is based on the starting configuration with 65% mol LiF, 28.5% mol SNF, and 6.5%mol TRU. On core level, this corresponds to ~63 tons of SNF with ~15 tons of TRU altogether to 78 tons of HM in the core, while the blanket consists of pure LiF salt. This leads to a start-up core with an averaged $\Delta\overline{k}_{\mathrm{eff}}$ of 0 over the first burnup cycle of 5 GWd/tHM.

3.2 Simulation over Lifetime

The operation over longer term requests a variable TRU or Pu feeding to keep the $\Delta\overline{k}_{\mathrm{eff}}$ over the cycle in the range of ±400 pcm, Figure 6. In the initial ~3.6 tons of TRU are fed into the system within 20 years. Additionally, a constant amount of SNF is fed into the system at begin of each cycle to keep the U-238 level almost constant, see Figure 7. After transforming the system into a fast reactor configuration the TRU feed is not needed anymore. Only SNF is fed into the system from this point on and the system stays inside the iteration band of ±400 pcm and ~90 tons of SNF are fed in 80 years into the system.

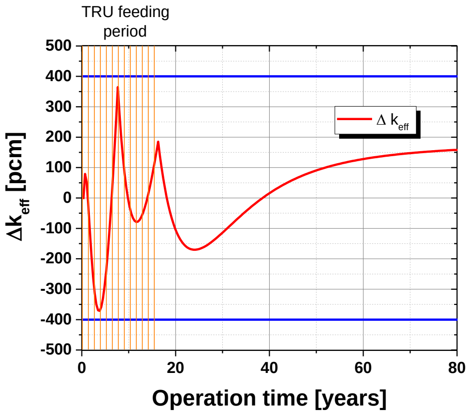

Figure 6: Change of the deviation of the averaged effective multiplication factor $\Delta\overline{k}_{eff}$ over a simulated operational period of 80 years within the iteration band of ±400 pcm

Throughout the observed period, the U-238 content remains almost constant, Figure 7. The isotopic number density of the most important fissile isotope, Pu-239, increases during the first part of the TRU feeding period and stays almost constant in the second part, as shown in Figure 7. The Pu-239 content decreases after the feeding to an asymptotic value, the Pu-240 isotopic content in the fuel salt increases to an asymptotic value almost twice as high as the initial value.

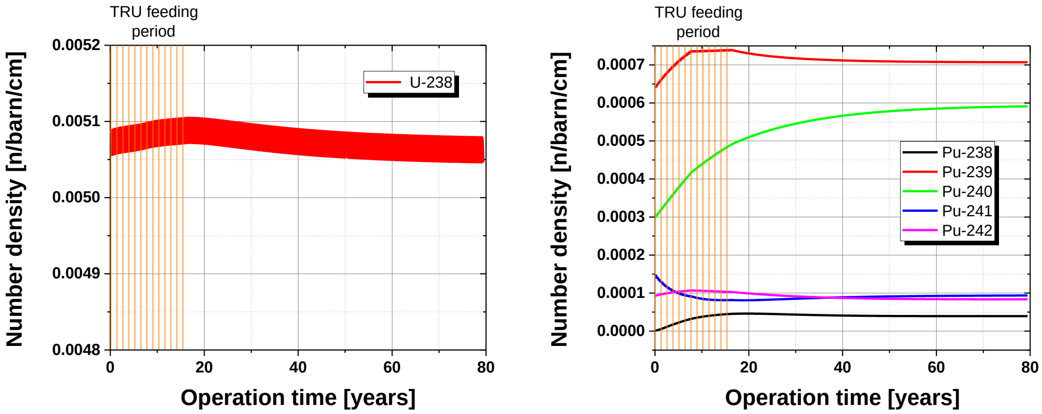

Figure 7: Number density of U-238 (left) and Pu (right) particles in the fuel salt over the observed operational period

Isotopic vector of the Pu compositions at the end of the observed operational time shows dramatic changes, when compared to the initial TRU feed, see TABLE 1. The share of LWR fissile Pu isotopes, decreases from the already low content in the TRU which is caused by high burnup of the LWR fuel. The major cause is strong build-up of Pu-240 and the observed decrease of Pu-241 and 242.

TABLE 1: COMPARISON OF THE PLUTONIUM VECTOR AFTER LONG TERM OPERATION WITH THE FEED

	Unload	Load
Pu-238	2.6%	3.1%
Pu-239	46.7%	52.5%
Pu-240	39.0%	24.6%
Pu-241	6.2%	12.2%
Pu-242	5.5%	7.7%
Pu_{fiss}	52.9%	64.7%

4. Discussion

The results shown here demonstrate that a molten salt fast reactor could be made operating on pure LWR spent nuclear fuel (SNF) with some limited feed of fissile material for the start-up phase, see Figure 6. Once this transition has occurred, the feeding can be solely SNF. Over all, ~90 tons of heavy metal are inserted within 80 years of operation. A hand calculation leads 88 tons of HM burnt, with the characteristic burning rate of 42 kg/TWh describing a fertile free system where no fissile and fertile material leaves the reactor [8].

The proposed reactor system has the potential to limit the misuse of plutonium this can be seen TABLE 1. On the one hand, the system plutonium is low in Pu-239 but high in Pu-240. On the other hand, the Pu staying within the reactor until it undergoes fission and no separation occurs in the fuel cycle. This fact has already been highlighted in 1978 by Engel et al, as one of the most attractive features of liquid fuelled reactors. [15]. Additionally, the system provides no immediate possibility for inserting pure fertile material since all fuel components are mixed immediately.

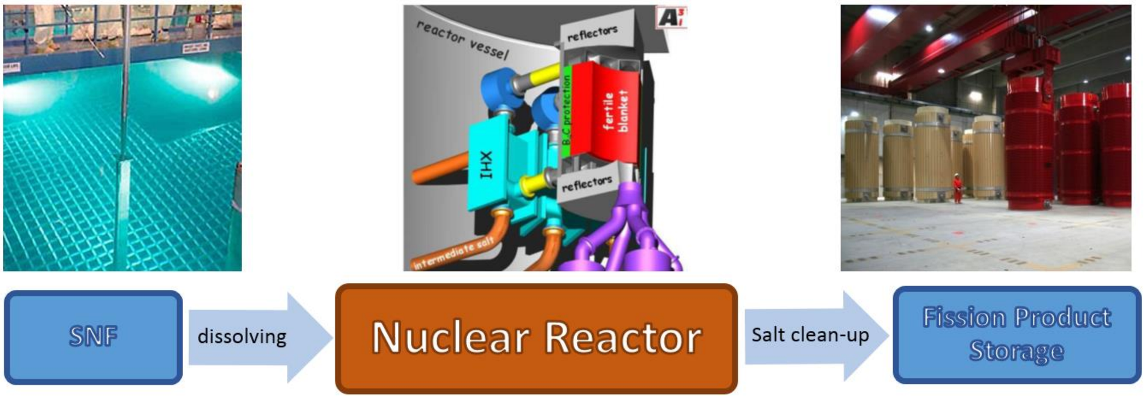

Figure 8: The reduced fuel cycle like it would be given using MSFR fed with SNF

Pictures: left http://www.wikiwand.com/en/Spent_fuel_pool, credit DOE License: Public domain, middle: EVOL benchmark configuration, right: Gorleben storage hall, Origin: GNS Gesellschaft für Nuklear-Service mbH

An additional, important gain is the significant reduction of the fuel cycle (see Figure 8), especially when compared to current closed fuel cycle proposal for fast reactor operation and the planned operational schemes in GenIV systems. No new, fresh resources are required which makes mining obsolete which causes the major contribution to eco and the human toxicity [17]. Thus the fuel cycle is reduced as well as the environmental impact of nuclear energy production. For this kind of reactors the salt cleanup will be demand driven, too. The task is to reduce the isotopes which prevent the reactor from long term operation instead of the classical approach of separating the fissile and fertile isotopes. In addition, this approach has the potential to keep the waste stream free of TRUs which will fulfil the requests of P&T as a side effect without transports to reprocessing, fuel production, and back to the reactor. Since no fissile material is separated, no ‘Plutonium economy’ is formed like it would be required a closed fuel cycle in traditional fast reactors which requires the separation of actinides.

The most challenging design parameter is a nuclear reactor which will not produce additional waste which is physically impossible. Operating on SNF, such as proposed here, will keep the waste mass on the same level as before but with increased short term activity, but ~ 20 times more energy is produced out of the material. However, this doesn’t relief the nuclear industry from developing a strategy for handling and storage of the separated fission products coming from the clean-up system and the off gas treatment. Regarding more than 320 000 tons of spent fuel in storage expected by 2020 (*IAEA https://www.iaea.org/sites/default/files/costingfuel0909.pdf)* leads to the possibility of operating more than 2000 reactors (~1.4 GWe) for ~80 years when the feed of fissile material can be provided from stockpiles like it is available in UK.

The final challenge, i.e. highly economic, reliable, secure, and safe, as well as sustainable in construction with a limited financial risk, will be an important challenge engineers. In order for a MSFR design to be realized, and to be manufactured and operated several multi-disciplinary scientific and technological challenges have to be solved, see Table 2. Besides operational experience has to be gained which will require a process in a manner similar to the historic development of reactors, starting with a small, low power experimental machine. This first machine has to be planned, built, and financed [8].

Engineering:

- Optimisation of burning used LWR fuel , i.e. demonstration of principles with validation
- Optimisation of reactor design, i.e. better than EVOL
    - Fully understanding the fluid dynamics under both normal and accidental conditions
    - Componentry for molten salt system
- Development of materials capable of operating under such extreme conditions, i.e. temperature, induced radiation damage, corrosion

Chemistry and Thermodynamics:

- Optimisation of salt purification, i.e. removal of fission products from within the liquid phase
- Design, implementation and capture of volatile fission products, helping to keep the liquid phase pure
- Preparing the fuel – choosing the optimal method for converting used LWR fuel in to a form sufficient for use within the reactor
- Appreciation of the chemical thermodynamics, and limitations in using molten salts with high levels of actinide loading

Safety:

- Implementation protocols for such a novel liquid reactor design
- Assurance in safety of such a co-located site, i.e. both reactor and reprocessing

Surrounding:

- Economic viability, ensuring the reactor is economically viable and competitive with current technology
- Ensure the public acceptance of such a new technology, without which the reactor is unviable

Table 2: Overview on the major challenges for the development of a MSFR following [3]

5. Conclusions

Invention as well as innovation in nuclear system development can be described with the concept of developments in S-curves. The identified key points to drive invention and innovation are the changed boundary conditions as well as the evolvable objectives. These new objectives require in our view the change to a new S-curve for nuclear development. The updated vision ideally coincides with the ultimate, universal vision for energy production which is characterized by minimal use of resources and production of waste, while being economically affordable and safe, secure, and reliable in operation.

Following these updated vision, a new innovative proposal is shown, which is based on the utilization of SNF from LWRs as the main fuel. This offers a way to fulfil the sustainability goals of the UN for innovative electric energy production. A proof of the feasibility is given from neutronics point of view to demonstrate the establishment of sufficient breeding long term operation. For the initial transition phase a support of fissile material is required.

The proposed system opens the way to a nuclear system which does neither require new resources since it uses the already existing ones nor does it produce additional waste since there is only more energy produces out of the already accumulated waste. This innovation provides a new option for a more sustainable future nuclear system and thus the ground for the long term success of nuclear reactors to act as a major contributor for the production of reliable carbon free, sustainable electric energy. In addition, the requirements for P&T will be fulfilled as an advantageous side effect. Furthermore the system provides enhanced resistance against misuse of Pu. The sustainability is supported by the significantly reduced fuel cycle, which consists of dissolving of SNF, reactor operation, storage of fission products only. The elimination of mining, conversion, and enrichment results in eliminating the major source of toxicity.

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