Nuclear Development

Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles

A Comparative Study

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

9. ALTERNATIVE ACTINIDE TRANSMUTATION APPROACHES

9.1 Introduction

This study is essentially focused on partially or fully closed fuel cycles using solid fuel in fast spectrum reactors. There are, however, other transmutation concepts that are proposed using other fuel and reactor types, including thermal-spectrum systems. The Expert Group therefore recognised that an overview of alternative approaches to the options described in the previous chapters would be appropriate in order to complement the study.

9.2 Transmutation systems using thermal neutrons

As was mentioned in the introduction, this report aims to give a comparison of FR and ADS based on solid fuel fast spectrum devices. Despite this initial focus, this section will address those systems using other approaches, e.g. thermal spectra and liquid fuels. In particular, molten salt systems were not assessed in depth in the first phase report [2] and are not in this report, but in recent years there has been a renewed interest in those systems both conventionally critical and accelerator driven.

Accelerator-driven systems are very difficult to operate with thermal neutrons and solid fuels (as in conventional LWRs) owing to prohibitive power peaking problems. Therefore, ADS concepts based on liquid or quasi-liquid fuels offer a way round these serious constraints. Two categories of reactors and ADS capable of transmutation are under serious considerations nowadays:

- Molten salt systems.
- Particulate-fuelled gas cooled reactors or ADS.

9.2.1 ADS and reactors with liquid or quasi-liquid fuel

9.2.1.1 Molten salt reactors and ADS

One of the alternative concepts for a nuclear reactor or accelerator-driven system having a significant transmutation potential is a molten-salt system, derived from an idea of a fluid-fuel reactor extensively investigated at the dawn of the nuclear era. Indeed, it is very appealing with its potential for a very effective consumption or transmutation of nuclear fuel (most of the constraints which limit burn-up in solid fuel are relaxed), inherent safety features regarding super-criticality accidents, etc. The molten salt fuel at operating fissile concentrations provides inherent protection against criticality accidents during handling. In thermal neutron designs, the graphite moderator is required for criticality criticality which can therefore occur only in the core. In other concepts, the design would have to exclude vessels that are not safe from criticality with credible fuel mixtures.

Molten salt reactors (MSR) can be designed with or without on-line processing, or with only partial processing. With reprocessing, some of the common nuclear reactor terminology does not apply. Except for some start-up periods, molten salt reactors operate at an equilibrium steady state. The fuel concentration and content do not vary with time. Fissioned or consumed fuel is replenished by feeding or by breeding. The term “fuel burn-up”, commonly used for solid-fuel reactors, thus has not the same meaning, as there is no specific amount of energy generation related to a particular identifiable original amount of fissile material. For the same reason, there is no excess reactivity to compensate for burn-up or for the progressive poisoning caused by fission product accumulation as in solid fuel reactors. Also decay heat problems for the MSR are not as severe in comparison with traditional solid fuel because the concentration of UF_4 in the fuel salt is small (l-2 mole%).

Molten salts can operate at high temperatures and low pressures, and have favourable heat transfer properties. These properties result in high thermal efficiency for the reactor and freedom from hazards associated with high pressures. The salts are chemically stable and non-flammable, averting fire hazards, and there are no energetic chemical interactions between the salts and water.

One of the major advantages of the fluoride-based MSRs is the potential for an integrated fuel recovery capability. The processing is based on the high volatility of UF_6. By sparging the salt with fluorine, uranium can be removed as UF_6, which can then be converted back to UF_4 and recycled into a fresh batch of fuel salt. The residual salt, free from uranium, could be subjected to any of a number of processes to remove fission products and concentrate them. The carrier salt components (lithium, beryllium, fluorine) could also be isolated and recycled if that were economically desirable. All of these steps could be made independent of the reactor operation [197].

One of the first molten salt reactors was an experimental type for aircraft [198], which operated successfully in 1954 in a “proof-of principle” short-term test at a power level of 2.5 MWth and at temperatures up to 860°C. The fuel was a solution of UF_4 in other fluorides with Inconel-clad beryllium as the moderator. At a very early stage of this project it was realised that the molten-salt system was an attractive option for a power reactor. In 1957 Oak Ridge National Laboratory began work on a commercial application. A reactor experiment (MSRE) was set-up and operated successfully at a power level of 7.3 MWth until the end of 1969 [199,200], delivering data for over 17 000 hours of critical operation and confirming the feasibility of eventual power production.

Building on MSRE experience ORNL has developed further concepts, particularly of a Molten Salt Breeder Reactor (MSBR) [201] using ^{7}LiF-BeF_2-ThF_4-UF_4 as the fuel salt in the Th-^{233}U fuel cycle. It reached only a very low breeding ratio of 1.07 but with a very advantageously low nuclear fuel concentration in the salt and consequently a very low inventory of fissile materials in the core.

In the mid-1970s, MSR development in the USA had progressed far enough to justify a greatly expanded effort leading towards commercial deployment, but competed with the LMFBR programme. To avoid diverting substantial funding from the latter, the MSR programme was instead stopped [202].

Other countries such as France, Japan and Russia put some effort into developing MSR concepts in the 1970s and 1980s, recognising their potential for more effective use of nuclear fuel through a gradual introduction of Th into the cycle.

Reducing the accumulation of Pu in LWR spent fuel was initially a main driver in the development of another concept of MSR. The fluoride salts used in thermal neutron systems were not suitable for fast neutron cores because of their neutron moderating ability, so interest turned towards the use of molten chlorides. 60NaC1-37$UC1_3$-3$PuCl_3$ was suggested for the fuel and 60NaCl-40UCl_3 (% by moles) for the blanket.

There are very serious material challenges in chloride systems, particularly at high temperature. Experimental data for individual metals show very poor corrosion resistance in a molten chloride environment, several orders of magnitude worse than in fluorides. Also the chloride mixtures are less stable chemically than fluorides.

9.2.1.2 Molten salt systems for an improved nuclear fuel cycle

A few reactor and accelerator-driven systems based on molten salt fuel have been proposed since the late 1980s, addressing different objectives.

Molten salts are mostly presented as "nuclear system solutions" not only for transmutation purposes but also for a long term, synergetic development of nuclear power based on introducing Th into the fuel cycle. A few systems are focused on the specific objectives of improving the existing U-Pu fuel cycle by transmuting existing nuclear waste in dedicated accelerator-driven systems.

Furukawa *et al.* proposed a concept of THORIMS-NES ("Thorium Molten-Salt Nuclear Energy Synergetics") [203], composed of:

- A molten-salt reactor [204], without continuous chemical processing or core-graphite exchange.
- Fissile-fuel producers utilising spallation/fission reactions of 1 GeV-proton – Accelerator Molten Salt Breeder (AMSB): ASO-series [205].
- Pyro-processing plants.

CEA has put some efforts into the concept of a thorium fuelled accelerator-driven sub-critical system for both energy production and TRU- incineration, called "TASSE" (**T**horium based **A**ccelerator-driven **S**ystem with **S**implified fuel cycle for long term **E**nergy production) [206], designed for nuclear energy production with reduced radiotoxicity in the waste. Tasse takes advantage of the Th fuel as a feed (breed) material and a good neutron economy in the so-called super-thermal spectrum which is possible in molten salt cores or designs like a high temperature reactor.

Since the early 1990s, Ch. Bowman has been proposing different molten salt systems for Accelerator-driven Transmutation of Wastes (ATW) with a special emphasis on non-proliferation aspects and incinerating weapon Pu [207-214]. Bowman's 2-stage molten salt system, so-called Tier-1 and Tier-2, is aimed to simplify and improve nuclear fuel cycle with an eventual goal of sustained nuclear energy without serious proliferation concerns and without advanced reprocessing of spent fuel.

AMSTER – Actinides Molten Salt TransmutER concept – a continually reprocessed molten salt critical reactor, moderated with graphite and burning TRUs on a support of uranium or Th with various levels of enrichment, has been proposed by Vergnes *et al.* [23]. The AMSTER reactor design has benefited from a detailed analysis of transmutation performance in different systems and especially in a conventional type of molten salt reactor.

Finally, the Kurchatov Institute in Moscow developed a concept of the cascade sub-critical molten salt reactor (CSMSR) to optimise the nuclear fuel cycle [215]. The cascade, divided into fast and thermal neutron zones, allows the accelerator power to be ten times less than in other ADS-concepts.

9.2.1.3 Thorium molten-salt nuclear energy synergetics – THORIMS-NES

Furukawa *et al.* have worked for over 20 years and published a number of papers [203-205] on a molten salt reactor concept and its capabilities for sustainable nuclear energy generation with a significant potential for Pu-incineration. The principles of THORIMS-NES concept were:

- Using thorium as a breeding nuclear fuel.
- Application of molten fluoride fuel technology.
- Separation of breeder reactor plants (MSB, based on advanced technologies for external neutron sources like spallation or even fusion) from power generating reactors (MSR).

The first stage of the THORIMS-NES concept has produced pre-conceptual designs of Small Molten Salt Reactors, as shown in Table 9.1.

Fuji Molten Salt Reactor concepts are based on the MSRE design, having molten salt circulating through a graphite moderator enclosed in a Hastelloy reactor vessel. The design was focused on achieving graphite lifetime of 30 years, and on-line chemical reprocessing has been limited to removal of volatile fission products.

An Accelerator Molten Salt Breeder AMSB concept presented in 1981 was a rather naive design with an integrated target-fuel molten-salt pool. The main objective for this system was breeding ^{233}U (up to 800 kg/year), incurring a high cost in electric power for the rather unrealistic 1 GeV, 300 mA accelerator.

1. The preliminary assessments of the transmutation potential of Molten Salt Systems indicate that fission products like ^{129}I, ^{135}Cs, ^{151}Sm and possibly ^{90}Sr, ^{93}Zr, ^{126}Sn and ^{137}Cs could be transmuted in the form of molten fluorides. However, there are no really reliable data on effective transmutation rates and requirements for isotopic separations.

2. The potential for TRU-transmutation in the modified FUJI-reactor, called FUJI-IV, in which 1/3 of the ^{233}U inventory is replaced by TRU (up to 200 kg in the core) is claimed to be about 50 kg/yr for a reactor or molten salt ADS of about 300 MWe. A so called Fuji-Pu small molten salt reactor of 250 MWth, specially designed to incinerate Pu, would incinerate about 100 kg Pu per year.

All the data quoted above are based mainly on relatively simple calculations and should be considered rather as an assessment of the potential of a Th-based nuclear fuel cycle phased into the existing LWR fuel cycle. The chemistry of TRU-fluorides is not really well known and all the neutronic calculations must be thoroughly verified with experimental results on solubility limits for Pu and MA-fluorides and their chemical stability.

Table 9.1. **Some characteristics of small molten salt reactors in comparison to MSRE-ORNL**

	Fuji-II	**Mini Fuji-II**	**MSRE-ORNL**
Thermal power (MWth)	350	16.7	7.3
Net electric generation (MWe)	155	7	–
Thermal efficiency (%)	44.3	42.	–
Reactor vessel inner diameter × height (m)	5.5 × 4.1	1.8 × 2.1	1.45 × 2.2
Maximum core diameter (m)	1.4, 3.4 (II zone)	0.6	1.14
Core graphite fraction (vol%)	93, 90 (II zone)	90	77.5
Core-Blanket power density – average/peak (kWth/l)	9.5/17.5	16.4/24.9	2.9/6.6
Neutron flux (10^{14} n/cm^2·s) Max. thermal Max. over 50 keV (damage to graphite)	 8.3 0.8	 0.58 0.75	 0.5 0.3

9.2.1.4 Tier-1 – Tier-2 Molten Salt Transmutation Systems

C. Bowman's concepts depart partially from the Li-Be based fluoride salts in favour of NaF-ZrF_4. A molten salt system called Tier-1 is a once-through accelerator-driven transmuter (see Figure 9.1), while a system called Tier-2 becomes a final burner of actinide remnants from Tier 1 operation. Unlike the Tier-1 system, Tier-2 is based on "conventional" Li-Be fluoride salts.

The goal of Tier-1 is to reach a single-pass remnant near the 10 % level without recycling or back-end chemical reprocessing (see Figure 9.2). In Tier-1 (see Figure 9.3), spent fuel assemblies from an LWR are first converted to fluorides to remove the uranium as UF_6, and facilitate the removal of fission products, and to prepare the rest of the waste for insertion into the transmuter. The primary constituent of the input to the transmuter from the spent fuel is the cladding as ZrF_4. NaF is added to the mixture and the NaF-ZrF_4 becomes a carrier for the actinides and the fission products. The waste flows through the transmuter continuously spending about five years on average in an effective flux of 2-4 × 10^{14} n/cm^2·s. Most of the actinides are burned away and the remnant isotopic composition is uninteresting as weapons material and incapable of supporting a thermal spectrum chain reaction. Without fission product removal at the input, the burn-up factor is 0.33; with fission product removal the burn-up factor improves to 0.217. If this once-through remnant from Tier-1 is geologically stored, there is greatly reduced concern for the repository as a plutonium mine for weapons material or for unused and concentrated nuclear fuel. The risk of spontaneous criticality is much reduced or eliminated and indefinite supervision becomes unnecessary.

Figure 9.1. **Tier-1 molten salt transmuter [214]**

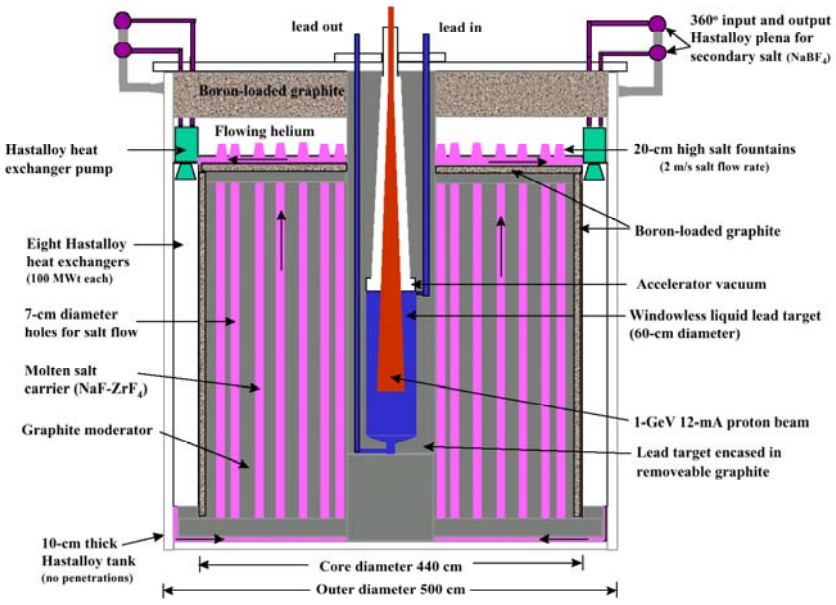
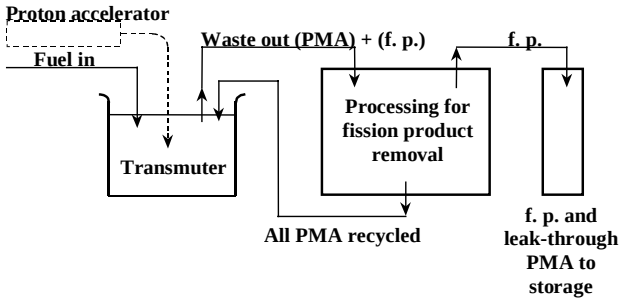


Figure 9.2. **Key feature of the Tier-1 approach [214]**

The conventional transmuter shown in the upper part of the figure requires chemical processing on the back-end to remove fission products and return the TRU for further burning. In the Tier-1 approach, the neutron economy enhancement by the accelerator and the burn-out of the fission products in the thermal spectrum allows a high burn-up without back-end chemical separations.

Conventional approach



New approach

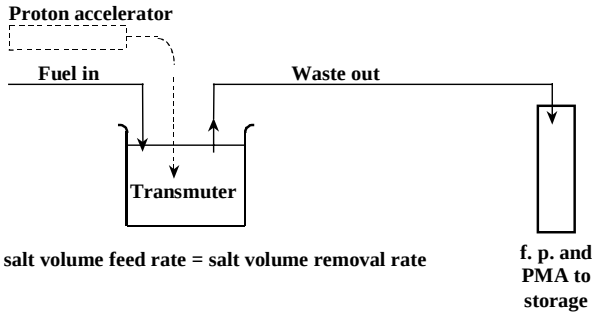


Figure 9.3. **Implementation of the once-through transmuter**

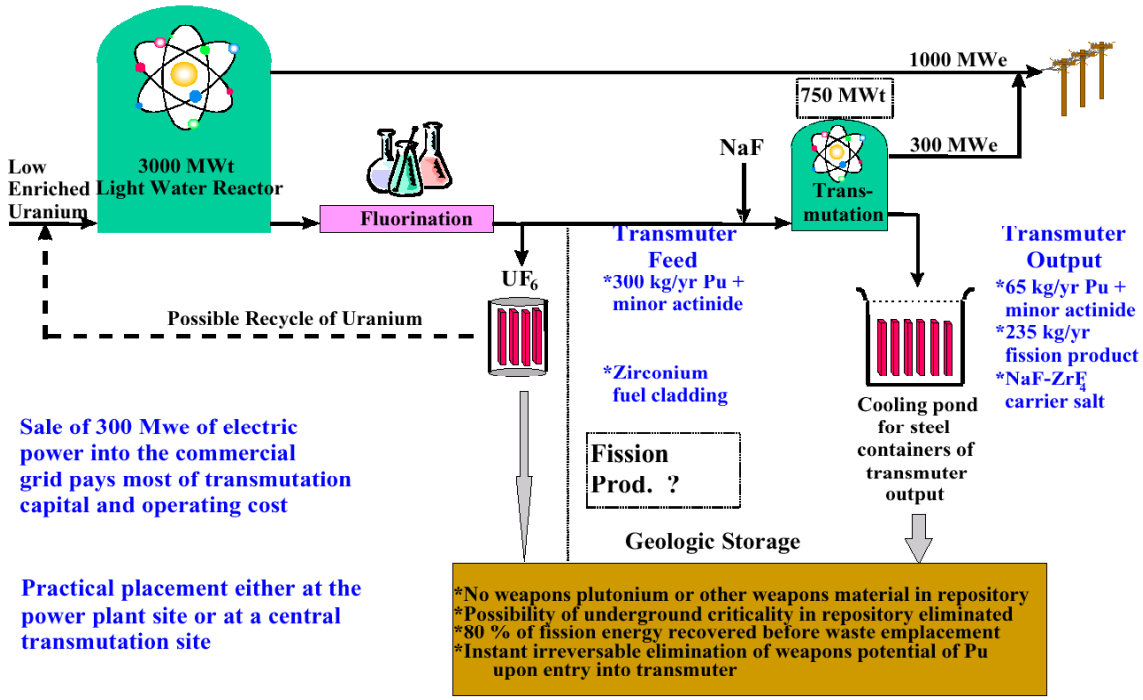
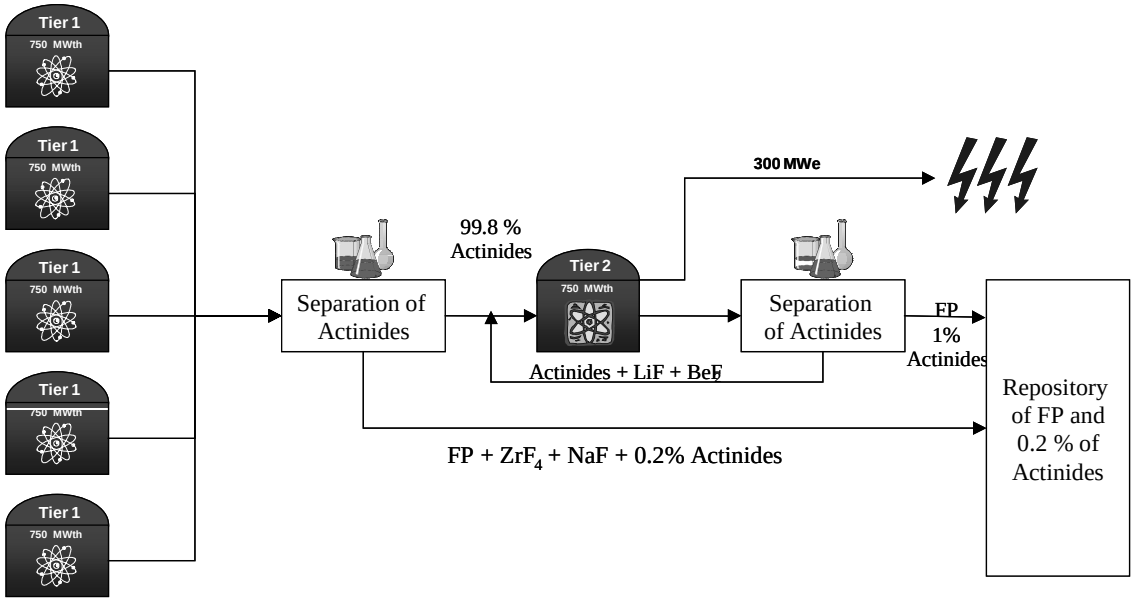


Figure 9.4. **The concept of Tier-2 systems [216]**
For a nuclear park corresponding to French nuclear power, 60 Tier-1 and 12 Tier-2 should be deployed, i.e. Tier-2 can support 5 Tier-1 facilities.



If there is further concern about the remnant actinides from Tier-1, they may be fed into Tier-2 to reduce these isotopes and their radioactivity perhaps by a factor of 1 000 or more. However the Tier 2 (see Figure 9.4) system follows the conventional approach shown at the top of Figure 9.2 and the performance depends on the efficiency of recovery in the chemical separation, which might be 99.9%.

This efficiency is degraded in the recycle process since each time the separation is performed some of the actinide is lost to the fission product waste stream. If the burn-up remnant is 90%, the proportion lost is increased by a factor of nine from 0.001 to 0.009. Bowman claims that the build up of curium and higher elements in his thermal spectrum system is not evident. Results presented in [216] show that Tier-1 may well be an effective burner of ^{239}Pu in a single pass. Although there is a some build up of the higher isotopes of ^{242}Pu and ^{244}Cm, ^{242}Pu is the most stable of the common plutonium isotopes and ^{244}Cm decays with a half-life of 18 years to ^{240}Pu.

The Tier-2 system requires a back-end separations facility and the elimination of fission products from the Tier-1 waste stream before feeding to Tier-2.

The total actinide inventory for the Tier-1 system operating at 750 MW thermal fission power and at a flux of 4×10^{14} n/cm^2·s is 193 kg. The actinide inventory for Tier-2 is 1 126 kg for a unit of 750 MWth power level. Since the fuel dwell time is 3.2 years, the back-end separations rate should be 350 kg/year.

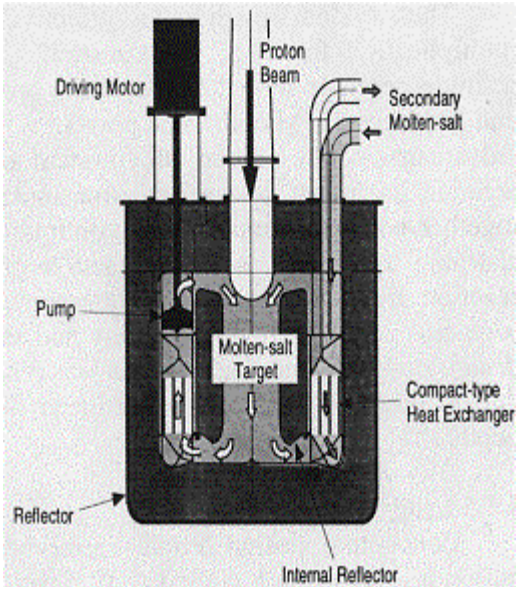
For Tier-2, the remnant waste is mainly ^{244Cm} and ^{246}Cm, but the actinide content of the Tier 2 waste is smaller by a factor of about 400 than in the Tier-1 waste stream. After 1000 years the ^{244}Cm has decayed to ^{240}Pu; most of the ^{246}Cm remains and decays with approximately the same half-life as ^{240}Pu. The alpha decay rate from Tier 2 at 1000 years is 1/1400 of that for unprocessed waste stored for the same period.

9.2.1.5 JAERI molten-salt and molten-alloy ADS concepts

JAERI has conducted conceptual design studies on an 800-MWth molten-salt target/blanket system for a dedicated accelerator-driven nuclear waste transmutation system. Figure 9.5 shows it schematically. A mixture of 64NaCl-5$PuCl_3$-31$MACl_3$ (where MA represents Np, Am, and Cm) has been chosen for the molten-salt system based mainly on the consideration of actinide solubility. Molten chloride is an attractive option since it has high Pu solubility and the mass number of Cl is about twice of that of F. The NaCl-$PuCl_3$ system has an eutectic temperature of 726 K for the composition 64NaCl-36$PuCl_3$. The solubilities of MAs in the salt are not known, but Pu in the chloride salt may be replaced by any minor actinide.

The molten-salt acts at once as fuel-target material and coolant. This significantly simplifies the target/blanket system configuration eliminating the physical and functional separation of target and core. One of the disadvantages of the fast neutron molten salt system is a large actinide fuel inventory. To reduce the primary molten-salt inventory, main pumps and heat exchangers are contained within the primary vessel. In the 800 MW molten salt system with an effective multiplication factor of 0.92 and a 1.5 GeV, 25 mA proton beam, the transmutation rate is approximately 250 kg/y, or 4.6% of the inventory per year assuming a load factor of 80%.

Figure 9.5. **Concept of JAERI's molten-chloride ADS [217]**



Another candidate for a fast-spectrum molten salt system is $PbCl_2$-$AnCl_3$ (where An refers to an actinide). Table 9.2 compares the parameters of the both systems. The difference seems not to be large, but the results are not conclusive owing to the lack of reliable property data for these salts.

The major issue of the molten-salt concept is the compatibility of structural materials exposed to a high temperature flowing chloride salt.

Table 9.2. **Comparison of sodium-based salt and lead based salt systems**

Target/Coolant	**Molten-chloride salt**	
Salt	$64NaCl$-$36AnCl_3$	$70PbCl_2$-$30AnCl_3$
k_{eff}	0.93	0.88
Proton Beam (GeV)	1.5	1.5
Neutrons per proton	37	40
Average neutron energy (keV)	800	768
Power density (keV/cm^3/p) max/ave	66/27	54/16
Power peaking	2.5	3.5
Primary system volume (m^3)	2.7	3.2
Molten-salt/Actinide inventory (kg)	10 000/5 400	17 000/4 100

* Averaged over target/core region, excluding IHX region.

Molten alloy ADS concept

Molten actinide alloy could be a possible alternative to molten salt as a liquid target/fuel for a fast neutron transmutation system. An evolutionary concept was studied by Katsuta *et al.* [218] in order to achieve a minimum inventory of fissile material by this means. Figure 9.6 shows a concept of an ADS

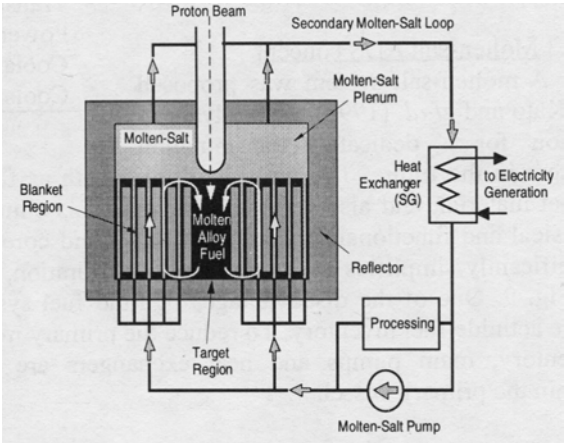
with a molten-alloy target/fuel, a graphite blanket with vertical coolant channels, and an upper plenum of molten fluoride salt. The preliminary study was performed on the alloy with the composition (11-32.5)Np-(4-12.5)Pu-24Co-(60-30)Ce-Tc.

The secondary molten fluoride salt (Li_2BeF_4) directly contacts with the molten-alloy through the vertical channels in the blanket. Efficient heat, mass and momentum transfer is expected at the contact interface between two co-current fluids. This eliminates the need of primary molten-alloy pumps and heat exchanger hardware.

The system with the effective neutron multiplication factor of 0.9 transmutes 145 kg of actinide per year with 1.5 GeV-16 mA proton beam and produces 455 MW thermal power.

Advantages of the molten-alloy system are a small actinide inventory and a high transmutation rate, together with the possibility of continuous on-line charging of minor actinides and removal of reaction products. The system, however, poses problems of material compatibility and safety. The design study of fluid-fuel systems was halted owing to safety concerns of about reduced defence-in-depth.

Figure 9.6. **JAERI's concept of molten salt ADS [217]**



9.2.1.6 TASSE-concept

The TASSE concept, a sub-critical nuclear system with simplified front and back end fuel cycles based on Th and with mobile fuel, pursues the following goals [206]:

- Simplifying the fuel cycle through:
  - Elimination of fuel enrichment.
  - Elimination or significant reduction of recycling through "on-line" technology without separation of TRU and fission products.
- Significant reduction of long-lived nuclear wastes due to both the negligible TRU-production and the high fuel burn-up.
- Burning out TRU from the current Nuclear Power park (PWR's) without toxic long-lived wastes on the way to the complete replacement of PWRs by TASSEs.

- Practically inexhaustible fuel resources for future NP:
  - Significant potential enhancement of deterministic safety due to stable reactivity and sub-criticality.

TASSE is not yet specified in technical details and following engineering solutions are under consideration:

- For the fast neutron spectrum option:
  - Molten salt compositions 30ThF4 + 25NaF + 45PbF2 with fuel density 7 t/m3 at 600°C.
  - Fuel burn-up: ~25-30% h.a. in the equilibrium once-through cycle.
  - A constant level of sub-criticality during core life due to the continuous feed-discharge regime of the fuel.
- In the super-thermal spectrum option:
  - Molten salt (32ThF4 + 14NaF + 54LiF) or HTR-type fuels (with a graphite to fuel proportion 4000/1 as optimal for HTR fuel).
  - Once-through fuel cycle.
  - Constant level of sub-criticality taking advantage of quasi-continuous feed-reloading regime for HTR-type fuel.

Figure 9.7 [219] presents the conceptual scheme for transition from present nuclear system to a TASSE system, using TRU to start the new TASSE system.

Two fuel cycles are possible for the TASSE concept, once-through with no processing of irradiated fuel and a version with on-line separation of fission products (see Figure 9.8).

The essential feature of the TASSE concept is the choice of the appropriate burn-up. The main idea is to choose the burn-up for the once-through cycle, tailored to an “optimal” value of the core reactivity, kept approximately constant over all the cycle. This can be achieved through an optimised choice of the fuel salt avoiding too large epithermal component or through an appropriate choice of the flux level.

In a sense the sub-criticality level in TASSE-system is adjustable to fuel enrichment. TASSE gives significant gains in terms of radiotoxicity reduction compared to PWR fuel cycle, up to an order of magnitude for the long term [219].

9.2.1.7 AMSTER

AMSTER is a new design of a molten salt critical reactor intended to burn TRUs with a uranium or Th support. AMSTER is based on the same salt composition as MSBR, i.e. 61LiF-21BeF_2+18AcF_4 (Ac stands for actinides). The concept is now under continuous development and can be considered as revisiting the principles of ORNL molten salt reactors but focused on incineration of TRU instead of breeding.

Figure 9.9 shows the conceptual layout of the AMSTER reactor with a working temperature range of 550°C-800°C and pumps placed within the graphite core.

Figure 9.7. **TASSE system in transition from LWRs and U-fuel cycle**

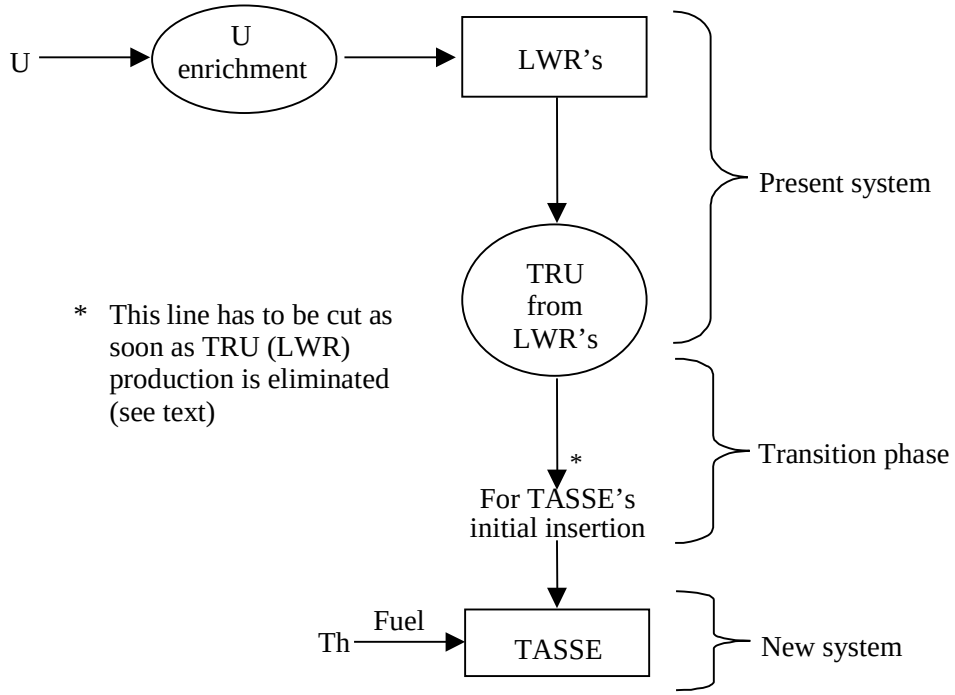


Figure 9.8. **Fuel cycle options for TASSE at equilibrium stage**

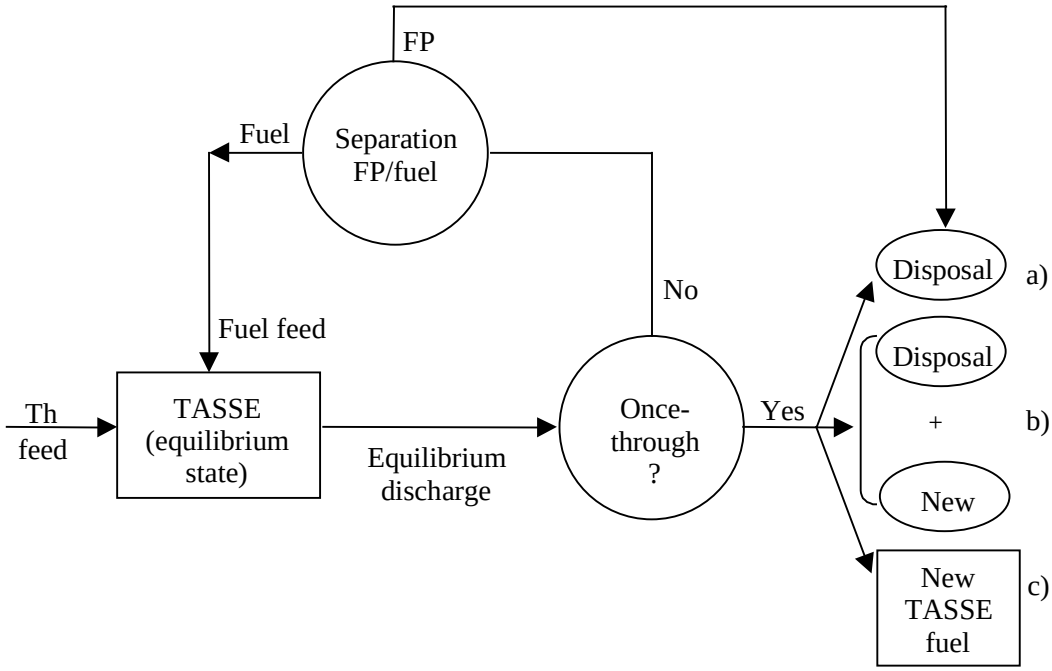
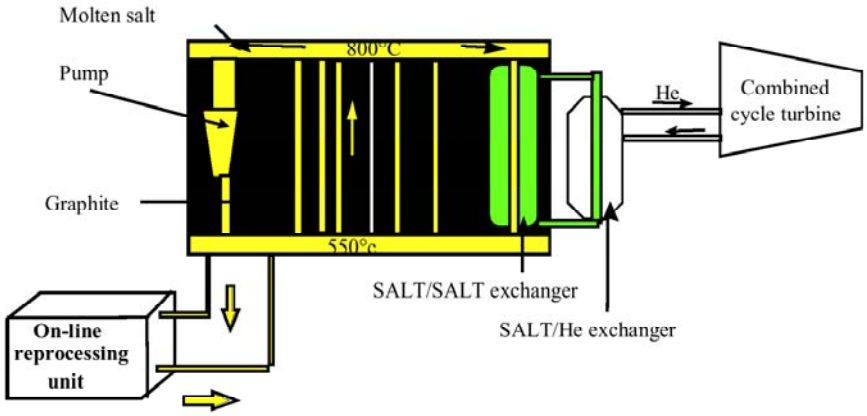


Figure 9.9. **A concept of the AMSTER reactor [220]**



Several fuelling options are envisaged for AMSTER:

- Core at equilibrium: based on spent PWR fuel with continuous addition of $^{235/238}$U or Th.
- TRU incinerator: core in which the TRU inventory is higher than that of the LWR spent fuel and kept constant by continually adding ^{235}U/Th and TRUs.

Table 9.3 presents the main characteristics of AMSTER with enriched U fuelling. Table 9.4 shows characteristics of the more advanced AMSTER reactor with two zones and Th feed.

There are two efficiency criteria for a TRU incineration strategy, namely:

- Minimising TRU losses per TWhe.
- Minimising the residual inventory in the cycle after e.g. 60 year decay.

AMSTER shows a very good performance compared to other transmutation systems [23]. It may be concluded that AMSTER promises a low TRU inventory with relatively small losses, owing to less degradation of the isotopic quality of the TRUs and above all to the contribution of highly fissile ^{235}U (in a system with U-feed) allowing a deep burnup of the molten salt fuel.

AMSTER may be considered as an effective critical transmutation system.

Table 9.3. **Main characteristics of AMSTER with U fuelling**

Load	**Core at equilibrium**	**TRU-incinerator**
^{235}U enrichment (%)	2.45	3.3
TRU content	2.6	4.7
TRU inventory in active core per GWe(t)	1.19	2.1
TRU inventory in reactor per GWe(t)	1.9	3.3
TRU consumption per TWhe (kg)	0	12
^{235}U consumption per TWhe (kg)	65	60

Table 9.4. **Characteristic of the 2-zone AMSTER with a Th-feed**

Ratio of volumes salt fertile/salt fissile	**1.5**	**2**	**2.5**
Inventory in the core (kg/GWe)			
^{232}Th	138 800	153 580	167 360
Uranium	3 354	3 650	3 900
Transuranium	53	54	54
Consumption of ^{233}U (kg/TWhe)	2.11	-0.12	+1.91
Core radius (m)	4.95	5.04	5,11
Volume of the salt in the reactor (m^3)	82	91	99
Power of the fissile zone (MWe)	1 895	1 800	1 714

9.2.1.8 Cascade Sub-critical Molten Salt Reactors (CSMSR)

The CSMSR concept is based on four main ideas [221]:

- Molten-salt fuel.
- Accelerator-driven sub-critical reactor.
- Cascade scheme in the sub-critical reactor.
- Non-aqueous methods of reprocessing the nuclear fuel.

CSMSR is envisaged to close the fuel cycle of LWRs.

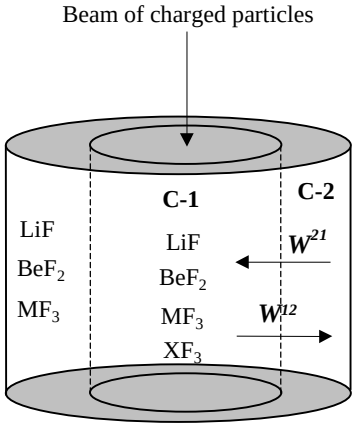
For the operation of a standard sub-critical reactor with $k_{eff} = 0.95$ and thermal power 1 GW, an external neutron source is required with an intensity of $\sim 10^{17}$ n/s.

The advantages of the sub-critical systems in combination with MSR are stated for CSMSR as:

- Reactivity accidents are excluded.
- Absence of instabilities, typical for critical MSR.
- Effective control of the reactor.
- Surplus neutrons that can be used for transmuting fission products and actinides.

Figure 9.10 shows a very schematic layout of the cascade sub-critical reactor with molten salt fuel. A beam of charged particles from the accelerator-driver spalls primary neutrons in C-1. These neutrons are multiplied by a factor $n \sim 1/\Delta k_{eff} \sim 20$ at sub-criticality $\Delta k_{eff} = 0.05$. In C-2 the number of such neutrons is amplified to $N \sim n^2 W_{12}$, where $W_{12} \sim 0.4$ is the probability to initiate fission in C-2 by neutrons from C-1. The number of fissions in C-2 per primary neutron in C-1 is $N_f \sim (n^2/\nu) W_{12} \sim 100$ at $k_{eff} = 0.95$. Besides the salt composition (LiF, BeF_2, MF_3), where M are minor actinides (Cm, Np, Am, etc., plus U, Pu and Th) C-1 contains also salts XF_3 of thermal neutron absorbers X (Gd, Sm) which depress the probability W21 of neutron penetration from C-2 to C-1.

Figure 9.10. **A very schematic layout of the CSMSR. C-1: a fast core, $k_{1\infty}$>1 – first cascade; C-2: a thermal core, transmutation zone $k_{2\infty}$ <1 – second cascade; M-actinide; X-thermal neutron's absorber.**



To provide the condition W21~0 in the considered scheme of CSMSR it is enough to introduce Gd at a proportion of 10^{-3} into the molten salt of C-1. Such admixture provides the necessary value W_{21}~10^{-2} - 10^{-3} and decreases the number of neutrons by only ~0.5%. In the traditional scheme of the sub-critical reactor with k_{eff} = 0.95 and thermal power 1 GW the accelerator is required to have a beam power of 50 MW. The cascade scheme allows the accelerator power to be reduced by a factor of 10 without loss in safety, and electron accelerators to be used, instead of proton accelerators. In the first case the accelerator consumes ~1%, and in the second ~10% of the electric power generated by CSMSR.

Table 9.5 presents data for different TRU in the equilibrium state of Nuclear Power Parks (NPP) of various structures with recycling of all actinides. It is assumed that these systems are fed with 1 000 kg/year of heavy atoms (^{238}U, ^{235}U) to give a power of 1 GWe. In the ideal case only fission products would be discharged and buried – about 1 000 kg/year.

Table 9.5. **Amount of TRU in equilibrium closed fuel cycles for thermal (LWR) and molten salt reactors (MSR), tons/GWe**

	LWR	**75% LWR + 25% MSR**
Neutron flux – average over cycle, n/cm^2·s	10^{14}	10^{14} (LWR), $5\cdot10^{15}$ (MSR)
^{237}Np	0.72	0.12
Pu (total)	5.8	3.1
^{241}Am + ^{243}Am	0.76	0.08
Cm (total)	1.38	0.09
TRU (total)	8.7	3.4
TRU without Pu	2.88	0.30
Heavy nuclides (total)	265	283

Table 9.6. **Amount of TRU in equilibrium closed fuel cycles for thermal (LWR), fast (FR) and molten salt reactors (MSR), t/GWe**

	FR	**89% FR + 11% MSR**	**51% LWR + 38% FR + 11% MSR**
Neutron flux – average over cycle, n cm^{-2} s^{-1}	10^{15}	10^{15} (FR), 5×10^{15} (MSR)	10^{14} (LWR), 10^{15} (FR), $5 \cdot 10^{15}$ (MSR)
^{237}Np	0.11	0.02	0.02
Pu (total)	21.1	18.0	10.4
^{241}Am + ^{243}Am	0.77	0.10	0.19
Cm (total)	0.19	0.04	0.11
TRU (total)	22.2	18.2	10.7
TRU without Pu	1.10	0.17	0.3
Heavy nuclides (total)	121	117	221

Having considered an overall neutron economy for the fuel cycle one can conclude that for a NPP of LWR's (with recycling of all actinides), the feed consists of 250 kg/year of ^{235}U and 750 kg/year of ^{238}U. For this system the equilibrium amount of MA reaches 2.88 t/GWe. All MA should be recycled in MSR. In this case the equilibrium amount of MA is reduced to 0.3 t/GW(e) and the ^{235}U feed to 200 kg/year, but the total amount of heavy nuclides in the fuel cycle is increased.

For 3-component NP ("triple strata") system (LWR+FR+MSR) one can really close the fuel cycle with ^{238}U feed only (1 000 kg/GWe per year) – see Table 9.6. In this self-sustaining NP system all surplus neutrons will be spent on actinide transmutation or on incinerating the long-living fission products – ^{129}I and ^{99}Tc, and the equilibrium amounts of MA and Pu are minimal.

The equilibrium amount of TRU can be further reduced only by feeding with ^{232}Th (about 80%) and ^{238}U (about 20%). It is possible to reduce the amount by a factor of 5 and maintain the necessary neutron balance without an external neutron source as has been shown for the AMSTER case.

Table 9.7 presents the various TRU inventories in a quasi-equilibrium fuel cycle for a MSR-burner taking all Pu and MA unloaded from LWRs of VVER type after the first fuel irradiation cycle.

Two cases have been considered:

- Spent fuel is unloaded and immediately after reprocessing TRU comes to the MSR.
- Unloaded fuel is stored for 20 years, then after reprocessing TRU come to MSR.

Preliminary calculations for a MSR of power 1 GWe show that neutron economy in both cases is sufficient not only for stable operation in critical conditions but for incineration of some long-lived fission products (^{129}I and ^{99}Tc). Actinide amounts in both cases are not large, but in intermediate storage the total reaches 20 t/GWe.

In the second case neutron economy is decreased by 0.1 n/fission but remains sufficient for MSR operation without an external neutron source.

Table 9.7. **Amount of TRU in quasi-equilibrium fuel cycle for molten salt reactor (MSR) fed with MA and Pu from LWR with or without intermediate storage (IS), t/GWe**

	MSR fed directly from LWR, without IS	20 years in IS	MSR fed from IS
^{237}Np	0.015	0.81	0.016
Pu (total)	0.81	17.7	0.83
^{241}Am +^{243}Am	0.062	1.42	0.084
Cm (total)	0.22	0.03	0.24
TRU (total)	1.11	20.0	1.18
TRU without Pu	0.30	2.26	0.35
Heavy nuclides (total)	1.11	20.0	1.18

9.2.2 Modular helium reactor and accelerator-driven transmuter with a particle bed fuel – MHR and MHA

A modular helium reactor and accelerator-driven transmuter with TRISO coated ceramic particle fuel has been proposed by General Atomics and is under serious investigations for a simplified, effective scheme of nuclear waste transmutation. In this transmutation scheme, the thermally fissile isotopes are destroyed in a critical but passively safe Gas-turbine Modular Helium Reactor, or GT-MHR, followed by a deep burn-up phase in an accelerator-driven GT-MHA [222,223].

The main idea is to take advantage of the unique feature of TRISO fuel particles, namely:

- Possibly very large burn-ups over 60%.
- Flexible design of TRISO particles for diverse waste destruction requirements (variation of kernel diameter).
- Slow, progressive burn.
- Fast destruction.
- As a final waste form, spent TRISO fuel in graphite blocks can be permanently stored without further processing.

The MHR/A based transmuter system concept has few important components necessary for high transmutation performance. The transmutation system consists of 2 strata: reactor stratum (MHR, so-called Tier-1) and accelerator-driven stratum (MHA Tier-2). Both MHR and MHA have heterogeneous cores with two distinct regions: driver region and transmutation region (see Figures 9.11 and 9.12). Driver regions are loaded with “driver” fuel (DF), containing mainly Pu + Np coming from UREX processes. ^{240}Pu and ^{237}Np in this fuel provide negative prompt feedback. TRISO-coated DF is engineered for 70% burn-up. Dimensions of kernel and fuel block are chosen to provide steady reactivity for 240-days. DF fuel is irradiated for 3 × 240-day periods in the MHR.

Transmutation regions are loaded with “transmutation” fuel containing Am + Cm coming from the UREX process, and TRU coming from a single reprocessing step of spent DF.

Fresh TF is neutronically positive, i.e. it acts as fuel not poison. TRISO-coated transmutation fuel (TF) is engineered to withstand 100% burn-up, with TRISO particles sized so as to provide high destruction rates (no resonance self-shielding). TF fuel is irradiated for 3×240-day periods in the MHR (thermal spectrum with neutrons of 0.01-1 eV) and then up to 6×240-day periods in the MHA (epithermal neutron spectrum of 1-10 eV).

Transmutation is performed in few steps with only two reprocessing phases, the front end UREX reprocessing of LWR spent fuel, and one single reprocessing of TRISO particles to recover TRU-elements and convert them into transmutation fuel – TF (see Figure 9.13). The LWR spent fuel is processed by the UREX process. Pu and Np are formed into "Driver" fuel elements". Higher Actinides (Am, Cm) are formed into "Transmutation" fuel elements. Transmutation fuel fabrication is highly challenging because of the higher actinides are volatile.

Long-lived fission products, Tc and I, are isolated and fabricated into special transmutation elements. It is suggested that Tc and I can be irradiated and transmuted in existing LWRs with minimal impact on operations.

Short-lived fission products, especially Sr and Cs, are isolated and stored in special waste-forms designed to deal with the intense short-term heat generation of the short-lived isotopes. Uranium goes to low level waste.

Driver and transmutation fuel elements are loaded into the MHR-bT critical core (the first stratum – Tier-1). The MHR core is divided into three Regions: Reflector, Driver and Transmutation Regions. The Driver Region (annulus) is divided into three zones of equal volume, and driver fuel (Np, Pu) is loaded into the driver region starting from the innermost zone (see Figure 9.11). The Transmutation Region (inner core) is also divided in three zones of equal volume and transmutation fuel (MA) is loaded in the transmutation region, starting from the outermost zone (see Figure 9.12). Residence of driver and transmutation fuel in the MHR core is three 240-day periods. In the driver region, after each period of operation, the driver fuel is moved outwards to the next zone and new fuel is added in the innermost zone; axial shuffling is also performed, to improve burn-up uniformity. The driver fuel is discharged after it has reached the outermost driver zone and processed to extract the fission products and remaining Pu, Np, MA are added to the transmutation fuel mix.

In the transmutation region, after each period of operation, the transmutation fuel is moved inwards to the next zone and new transmutation fuel added in the outermost zone. After reaching the innermost transmutation zone, the transmutation fuel is discharged. It is suitable for further irradiation in the sub-critical MHA (the second stratum – Tier-2) and no further reprocessing of the TF particles is needed.

The MHA fuel blocks are different in c/f ratio from the MHR fuel, assuring operation in the epithermal neutron spectrum. The MHA core is sub-critical, operating at k_{eff} = 0.7 - 0.8, and a power multiplication of ~10. Like the MHR core it is also divided into three zones of equal volume. Fuel is loaded in the core, starting from the outermost zone, and fuel shuffling is as in the MHR. After irradiation, fuel is sent for final packaging and repository storage.

In the transmutation region, after each period of operation, the transmutation fuel is moved inwards to the next zone and new transmutation fuel added in the outermost zone. After reaching the innermost transmutation zone, the transmutation fuel is discharged. It is suitable for further irradiation in the sub-critical MHA (the second stratum – Tier-2) and no further reprocessing of the TF particles is needed.

Figure 9.11. **Core layout of the MHR-system – Tier-1 Strata.**
Neutrons generated in the MHR Driver (outer) Region from "fresh" spent fuel, rich in Pu, drive the transmutation of the waste actinides in the Transmutation (inner) Region. In the Driver Region, the fuel to graphite ratio and particle size are engineered to provide slowly changing reactivity. In the Transmutation Region, the fuel to graphite ratio and particle size are engineered to provide fast burn rates [222].

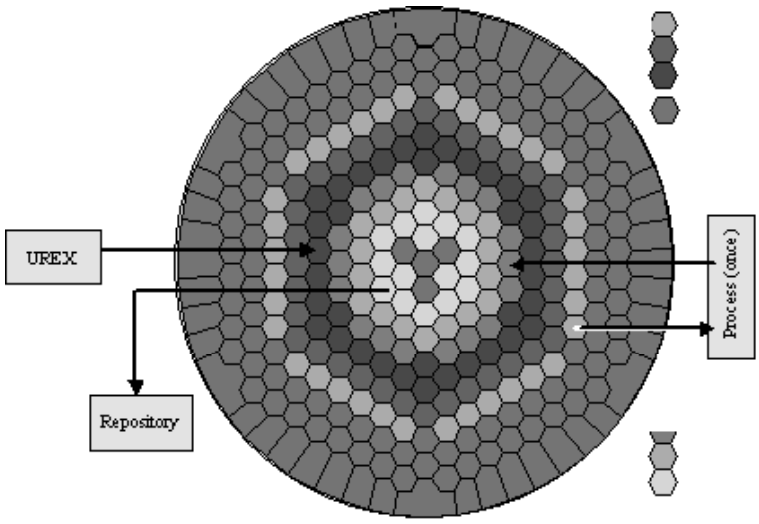


Figure 9.12. **Core layout of the MHA-System – Tier 2 strata.**
Neutrons generated in the spallation target drive the final destruction of the waste actinides in MHA through a progressively harder neutron spectrum (1-10 eV range). In the Outbound Track, going towards softer spectrum, remaining fissile isotopes are reduced. In the Inbound Track, going towards harder spectrum and target neutrons, remaining TRU is deep burned.

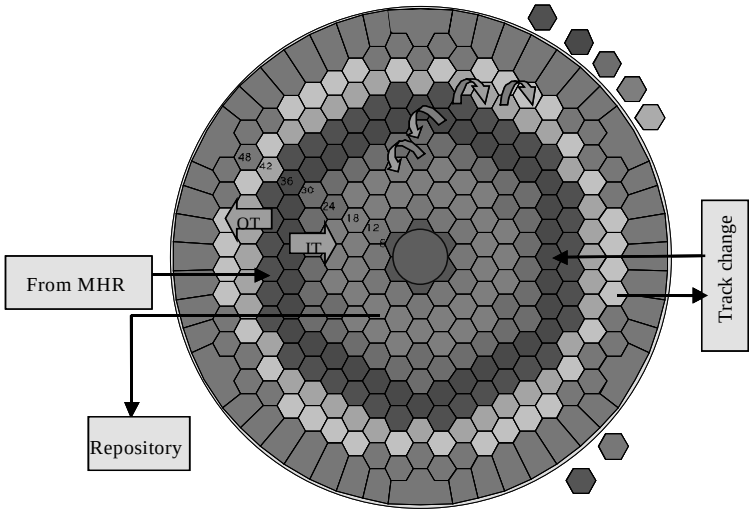
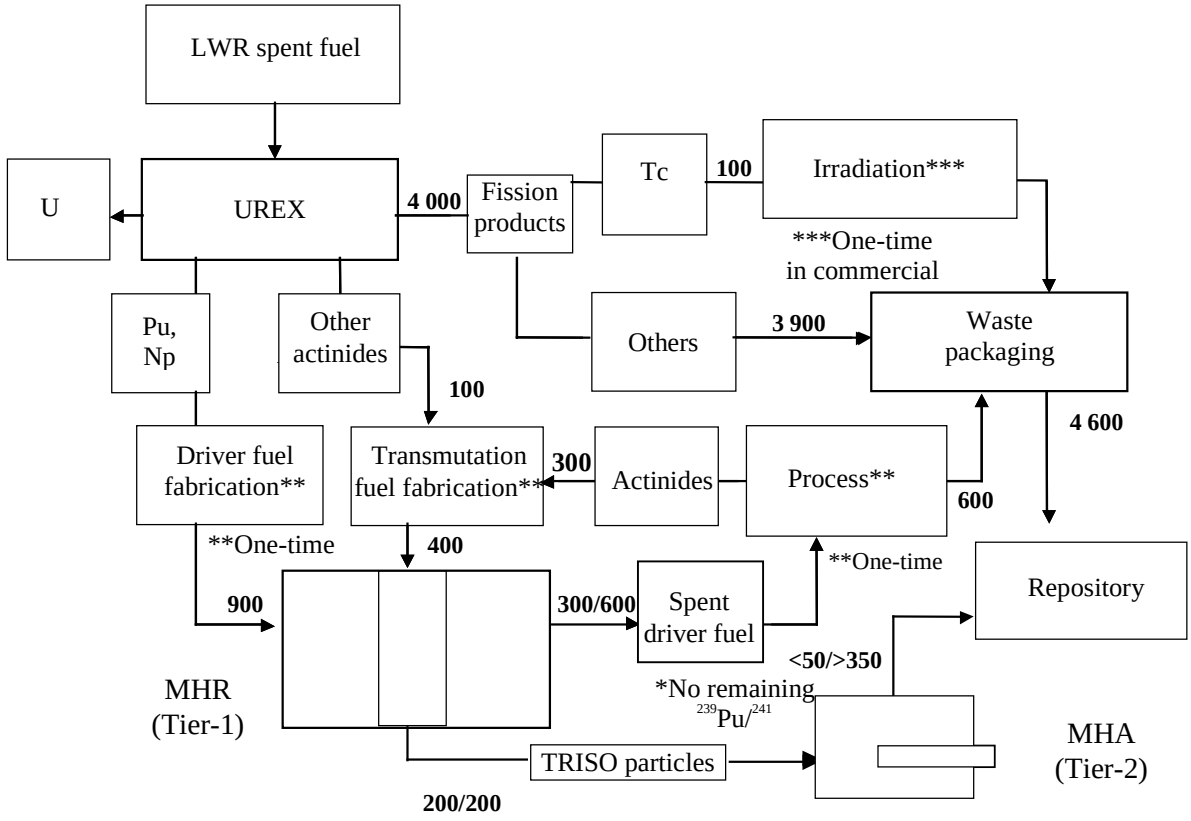


Figure 9.13. **A schematic fuel cycle for MHR/A transmutation system. Material balance in kg/y. One MHA would serve 4-5 MHRs [222] (flows in kg/y)**



The MHA fuel blocks are different in c/f ratio from the MHR fuel, assuring operation in the epithermal neutron spectrum. The MHA core is sub-critical, operating at k_{eff} = 0.7 - 0.8, and a power multiplication of ~10. Like the MHR core it is also divided into three zones of equal volume. Fuel is loaded in the core, starting from the outermost zone, and fuel shuffling is as in the MHR. After irradiation, fuel is sent for final packaging and repository storage.

In the latest General Atomics concept a unit park of the MHR/A system comprises 4 × 600 MWth MHRs and one 600 MWth MHA, all designed on the basis of a commercial HTGR reactor with competitive cost/performance features. Table 9.8 gives a preliminary comparison of ALMR and MHR/A performance.

In conclusion it can be said that the MHR/A transmutation concept has very attractive features. In the first stratum 75% TRU transmutation can be achieved and then after a single reprocessing an accelerator-driven MHA increases burn-up to 95% of the initial TRU inventory. These deep burn-up levels are achieved with no plutonium reprocessing and at a much higher rate than in a corresponding fast reactor ALMR based system. This is made possible by encapsulating the waste to be transmuted in coated ceramic microspheres that accommodate large amounts of fission products in spherical expansion volumes.

It remains, however, to be shown that spent TRISO particles are really suitable as a final waste form and that burn-up performance can really reach the limits assumed in the calculations.

Table 9.8. **Comparison of transmutation performances of ALMR and MHR/A systems**

	ALMR* (3 000-MWth parks) each park is 6 modules	**MHR/A (4 × 600-MWth R-units + 1 × 600-MWt A-unit)**
TRU initial	700 t	700 t
TRU inventory (per unit or park, Core + Process)	12 000 + 4 000 = 16 000 kg	3 000 + 200 = 3 200 kg
Breeding ratio	0.76	0.0
TRU destruction rate (75% avail.)	200 kg/y	1 000 kg/y
TRU destruction ratio (rate/inventory)	1.5 %/y	30 %/y
Decontamination factor	200	200
TRU loss rate to processing/ fuel fabrication	40 kg/y	1 kg/y
Campaign time to destroy 700 t	~3 500 park-y	~700 park-y
Residual TRU-inventory:		
– TRU waste to repository	3.5 t	35 t
– TRU lost in processing	~100 t	~1 t
– TRU in last unit	12 t	~0 t
Total	**115.5 t**	**36 t**

9.3 Conclusions

Attention should be paid to a category of actinide transmutation systems using alternative technologies, e.g. thermal neutrons and liquid fuels. However, most of these should be seen in a very long-term perspective as the respective fuel and reprocessing technologies as well as the systems themselves are essentially based on even more advanced concepts, compared with the solid-fuel/fast-spectrum systems on which the present study is focussed. This applies especially to the molten salt systems which, from a sustainability viewpoint, appear to be the most interesting systems in this category.