SESSION VI

LONG-TERM RADIOLOGICAL IMPACT AND CONCLUSION

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LONG-TERM ENVIRONMENTAL IMPACT OF UNDERGROUND DISPOSAL OF P&T WASTE

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Abstract

The long-term radiological impact of underground disposal of vitrified HLW (high-level waste) resulting from different P&T strategies has been evaluated. The considered P&T strategies include recycling of Pu and Np in homogeneous mode and Am in heterogeneous mode in CAPRA reactors or EFRs (European Fast Reactor). The safety assessment has been performed for three different types of host rocks, i.e. hard rock, salt and clay. For comparison assessments have also been made for the direct disposal of irradiated classical PWR UOX and UOX and MOX fuel, and of CAPRA and of EFR fuels. The key radionuclides have been identified and the capabilities for their partitioning and transmutation have been assessed.

Introduction

In the framework of the EC project FI4I-CT95-0006 on P&T strategies, SCK•CEN, AEA-Technolgy and GRS have assessed the effect of applying P&T techniques on the long-term radiological impact of underground disposal of generated HLW types. They considered respectively the geological disposal in plastic clay, crystalline hard rock and salt. The considered P&T strategies include the use of CAPRA and EFR reactors. In these reactors Pu and Np are recycled in homogeneous mode (MOX with Pu and Np) while Am is burned in heterogeneous mode (Am-targets). The long-term radiological impact has been assessed for two types of radionuclide release and exposure scenarios: the first type considers the ground water pathway (often referred to as the "normal evolution scenario"); the second type is a human intrusion scenario. In the long-term radiological impact assessment we compared mainly the direct disposal of spent PWR UOX fuel (once-through cycle) with the disposal of vitrified HLW resulting from PWR UOX fuel, with the disposal of vitrified HLW resulting from PWR UOX and MOX fuel, and CAPRA and EFR MOX. Calculations have also been made for spent PWR, CAPRA and EFR fuel.

In the following sections first the three disposal concepts are briefly described. The results of the radiological impact assessments are given for the two types of radionuclide release and exposure scenarios. Then the key radionuclides are identified and the capabilities to partition and transmute them are evaluated. Finally some conclusions are drawn about the considered P&T strategies and about the required performance of P&T techniques in general.

Disposal concepts and safety assessment methodology

Disposal in crystalline hard rock

The assessment of environmental impact for the case of disposal of spent fuel and vitrified HLW to a hard rock repository has been carried out by AEA Technology in the UK. No specific disposal site has been identified in the UK, and the calculations are therefore based on a conceptual repository design in which it is envisaged that the wastes are emplaced in containers in narrow boreholes, separated by about 25 metres. Each borehole consists of a single stack of containers, 20 high, about 30 m in height and situated at a depth of around 500 m. Each stack contains the spent fuel corresponding to about 15 tonnes of uranium, or the HLW corresponding to about 27 tonnes of uranium. Each borehole, together with its backfill, can be treated as a separate sub-facility. In all calculations carried out in this work, the dose from only one borehole is considered. The results can be scaled directly according to the total amount of uranium fuel consigned for disposal or on the quantity of electricity generated from it. The modelling has addressed both cement backfill and bentonite backfill, representing high and neutral pH environments respectively.

Two types of mechanism for the release of radioactivity into the environment are considered. The first, termed the groundwater release pathway, consists of the dissolution of radionuclides in groundwater flowing through the repository followed by migration through the geosphere and biosphere. Three alternative climatic regimes were considered in the biosphere modelling: a current day climate with marine discharge; and future boreal and arctic climates with terrestrial discharge. The groundwater release pathway is modelled using the MASCOT computer program, a probabilistic safety assessment model developed by AEA Technology plc under contract to United Kingdom Nirex Ltd [1,2]. The second type of mechanism consists of human intrusion into the repository some time after closure. This is modelled by the HINDSITE (Human Intrusion into a Nuclear waste Disposal SITE) program [3].

Disposal in a salt dome

GRS studied the disposal of radioactive waste at more than 800 m depth in a salt dome at the Gorleben site, Germany. The repository is a combination of disposal boreholes and galleries or vaults, which are backfilled with crushed salt. There are separate disposal fields for heat emitting HLW and for MLW. Important features for the safety evaluation are diapirism and subrosion, characteristics of the dome itself, as well as the high heat conduction and creep of salt under lithostatic pressure as the most important salt characteristics. The creep of salt will cause an encapsulation of the emplaced waste, on the other hand in case of brine intrusion, the creep of salt is a powerful driving force for radionuclide transport. The creep rate of salt increases with increasing temperature and therefore the encapsulation of waste in the HLW field is faster than in the MLW field. By consequence the radionuclide release from the MLW field is in general more important than from the HLW field.

Within the EC project EVEREST [4], three main scenarios were considered, i.e. the convection diffusion scenario, the cavern-convection scenario and the uplift-erosion scenario. In the EVEREST study, a very detailed repository and aquifer model was built up. For this study, simplified models were used.

Disposal in clay

SCK•CEN assessed the performance of a repository in a deep plastic clay layer, i.e. the Boom Clay at Mol, Belgium. The Boom Clay is about 100 thick and is at Mol situated between 180 and 280 m depth. SCK•CEN considerd a gallery disposal concept in which the waste is packed in stainless steel containers disposed in the centre of a 2.5 m diameter gallery backfilled with bentonite [5]. The capacity of the repository corresponds to the electricity production of the currently installed nuclear power in Belgium (i.e. 5.5 GWe) during 40 years, which corresponds to about 1500 TWh. For the waste quantity about 7 galleries of 1200 m length are required. The simulation of the ground water pathway scenario includes the radionuclide release from the waste, the diffusion through the host clay layer, and the advective-dispersive transport through the overlying aquifers to a water well used by a self-sustaining farmer family. For the simulation of the transport in the near field and the aquifer, the finite volume code PORFLOW [6] is used.

In the human intrusion scenario it is supposed that a core-drilling crosses a disposal gallery. The consequences of core examination, including inhalation and ingestion of dust and direct irradiation are assessed

Results of the long-term safety evaluation

Ground water release pathway

Disposal in crystalline hard rock

The environmental impact of the disposal of three types of nuclear fuel (PWR, MOX and CAPRA) and of the vitrified HLW derived from them has been assessed in this study. The doses to mankind from the groundwater release pathway were in all cases very low (of the order of 10^{-7} Sv, or even lower for marine discharge situations). The following table shows examples of some of the more important results.

Table 1 **Results for the PWR Spent Fuel and the PWR, MOX and CAPRA Vitrified HLW Inventories (all data for arctic biosphere and cement backfill)**

Waste	Peak Dose (Sv)	Time of Peak (years)	Key Nuclides (in order of importance)
PWR Spent fuel	$2.49\ 10^{-7}$	$1\ 10^{5}$	^{126}Sn; ^{226}Ra; ^{129}I; ^{230}Th; ^{135}Cs; ^{79}Se
PWR HLW	$5.80\ 10^{-7}$	$8\ 10^{4}$	^{126}Sn; ^{233}U; ^{79}Se; ^{226}Ra; ^{135}Cs; ^{129}I
MOX HLW	$7.35\ 10^{-7}$	$1\ 10^{5}$	^{126}Sn; ^{226}Ra; ^{233}U; ^{79}Se; ^{135}Cs; ^{129}I
CAPRA HLW	$7.82\ 10^{-7}$	$1.6\ 10^{5}$	^{126}Sn; ^{226}Ra; ^{233}U; ^{135}Cs; ^{79}Se; ^{129}I; ^{231}Pa

The key radionuclides contributing to doses via the ground water release pathway are: ^{126}Sn; ^{226}Ra; ^{129}I; ^{230}Th; ^{135}Cs; ^{79}Se; ^{14}C; ^{36}Cl; ^{233}U.

The order of importance of these radionuclides varies between various fuels and release scenarios. the relative roles played by the radionuclides depends on assumptions about the rate of release from the repository and the chemical properties of the elements, but the results for the overall doses are robust against a range of assumptions. The biosphere dose factors play an important part in determining the doses from each radionuclide, and these factors are subject to considerable uncertainty. However, an essentially similar group of radionuclides, although in a different order of importance, have been identified as dominating the overall concentration of radionuclides leaving the near field.

Doses could be reduced in a roughly proportionate way by reducing the inventories of radionuclides whose release is not limited by their solubility, but by their inventories (e.g. ^{129}I, ^{135}Cs). For other cases, such as ^{126}Sn, the release is solubility controlled in some inventories; and although doses could be reduced by limiting the inventory, there would not be a simple linear relationship.

On the basis of these results for the groundwater release pathway, the radionuclides which should be removed from HLW as part of a P&T strategy, in order to reduce the environmental impact of waste disposal, are: ^{126}Sn; ^{79}Se; ^{135}Cs; ^{241}Am; ^{237}Np.

The last two of these are included because their radioactive decay chains lead to the production of ^{233}U, which is a significant contributor to the total dose. Daughters of uranium and plutonium are not included in the list because the parents are removed from spent fuel with very high efficiency during conventional reprocessing. Similarly ^{14}C and ^{129}I are not included since they are also largely separated during reprocessing. ^{36}Cl is more significant in intermediate level waste than in HLW, and it is also not addressed further here.

Disposal in a salt dome

The central feature of the applied model is the limitation of the radionuclide transport by their solubility in brine. Therefore a possible radionuclide release depends on the brine quantity coming into contact with the waste, which in turn depends on the total excavated salt volume.

As already indicated above, the main radionuclide release is expected from MLW. It is expected that the volume of MLW waste from CAPRA fuel will be much larger than from PWR UOX and MOX fuel. Therefore the excavated volume for the disposal of MLW from a fuel cycle using a

combination of PWR UOX reactors with CAPRA reactors will be about twice the excavated volume needed for a fuel cycle using only PWR's with UOX and MOX. By consequence the dose from the former fuel cycle is about twice the dose from the latter fuel cycle. In case of direct disposal of UOX fuel the expected individual dose will only be about 2% of the individual dose from the UOX/MOX PWR fuel cycle. The main radionuclides contributing to the total individual dose are ^{129}I, ^{135}Cs and U- and Np-decay chain members.

Disposal in clay

The results of the consequence analysis for geological disposal of HLW and spent fuel in clay are in line with the above results for disposal in hard rock and salt. For disposal in clay the radionuclide flux into the aquifer and thus the dose to the critical groups are determined by the solubility of the radionuclides in the near field (and required repository surface) or by their inventory if they are not solubility limited. Under the slightly alkaline and reducing conditions governing in the Boom Clay, the solubility of the actinides and of many fission products is very low. Pu and Am are strongly retarded in the clay and thanks to the relative short half-life of their isotopes compared to their transport time, their release into the aquifer is negligible. The maximum doses of Np and the U decay chain isotopes appear only on the very long-term, i.e. > 10^6 years, and are almost independent of the in this project considered P&T strategies because their inventory remains large enough to lead to saturation for a long time in the near field. The most important effect of reprocessing is the reduction of ^{129}I, which is not solubility limited, in the MLW. Most of the ^{129}I is during the reprocessing released into the sea (or ends up in the MLW). The maximum dose due to ^{129}I is reached after about 50 000 years and is about a factor 10 lower for vitrified HLW, which contains only 10 % of the iodine, than for the disposal of spent fuel. The effect of the considered P&T strategies on the other important fission products such as ^{79}Se, ^{126}Sn and ^{135}Cs is negligible. The dose due to the disposal of the different HLW types is very similar

The dose due to the disposal of spent fuel is higher than the dose due to the disposal of vitrified HLW. The dose due to CAPRA fuel is higher than the dose due to EFR fuel, which on its turn is higher than the dose due to PWR MOX or UOX.

Human intrusion scenario

Disposal in crystalline hard rock

AEA-Technology has considered the drilling for e.g. exploratory purposes of a borehole through the repository. It is supposed that the drilling crosses a waste disposal borehole over its full length. Two exposure scenarios have been considered: one in which a geotechnical worker examines the core and one in which residence occurs (site occupation) on land contaminated by the spail from the drilling operations. While in the first scenario only direct irradiation, inhalation and injection of dust needs to be considered, also ingestion of locally grown vegetables is considered in the occupation scenario. Calculations were made for vitrified HLW and spent UOX fuel.

The results show that on long term the key contributors to the dose are ^{241}Am and the Pu isotopes for the core examination scenario and ^{99}Tc for the residence scenario. On short term i.e. less than 100 years the contributions of ^{137}Cs and ^{90}Sr are very important. The doses are general very high and remain above the deterministic effect level for more than 10^5 years. Reprocessing reduces the dose from the core examination scenario but has almost no effect on the dose from the residence scenario as the key contributors for this scenario are not affected by reprocessing.

Disposal in salt

GRS considers, as human intrusion scenario, a solution mining scenario. In this scenario salt is mined by injecting water through a borehole in the salt dome and by pumping up the brine. In this human intrusion scenario, medium active waste canisters are assumed to fall into the mined cavern. The number of canisters falling into the cavern is defined by the scenario. The release rates for the human intrusion scenario show a strong dependency on the scenario definition and a low dependency of radionuclide content. Because of large scenario uncertainty of the human intrusion scenario for a salt repository, a comparison of fuel cycle scenarios is not meaningful. For example the UOX fuel cycle (usage of UOX fuel only) takes no medium active waste into consideration. The calculated release rates for this human intrusion scenario are zero.

Disposal in clay

SCK has considered a core examination scenario similar to the one considered by AEA. The scenario is based on the borehole model developed by NRPB [7]. We only considered the routine inspection of a drill core. Calculations have been made for vitrified HLW and spent fuel from PWR UOX and MOX and from CAPRA and EFR reactors. The results are similar to those from AEA-Technology. In Figure 1 the dose from the core examination scenario is shown for vitrified HLW resulting from the reprocessing of CAPRA fuel. At early times the dose is dominated by ^{137}Cs and ^{90}Sr while at the long term the dose is dominated by the actinides, mainly ^{241}Am, Pu isotopes and ^{245}Cm. The doses are larger for CAPRA waste than for EFR waste, then for PWR UOX waste, because of the higher burn-up and actinide inventory. As the considered P&T strategies were concentrated on the actinides and as it was not defined what was done with Cm, SCK made a comparison of the dose due to the actinides between the disposal of spent fuel and vitrified HLW with the Cm added to it or without Cm. This comparison for CAPRA waste is shown in Figure 2. The effect of reprocessing on the dose due to the actinides is important, however if Cm is not burned the dose remains above the deterministic effect level for about 10^4 years.

Figure 1. **Dose from the core examination scenario for vitrified HLW from reprocessing of CAPRA fuel**

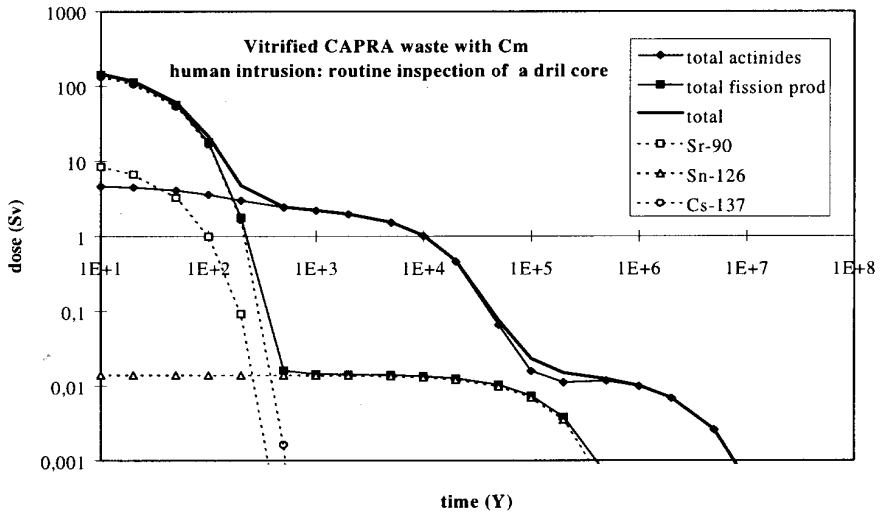
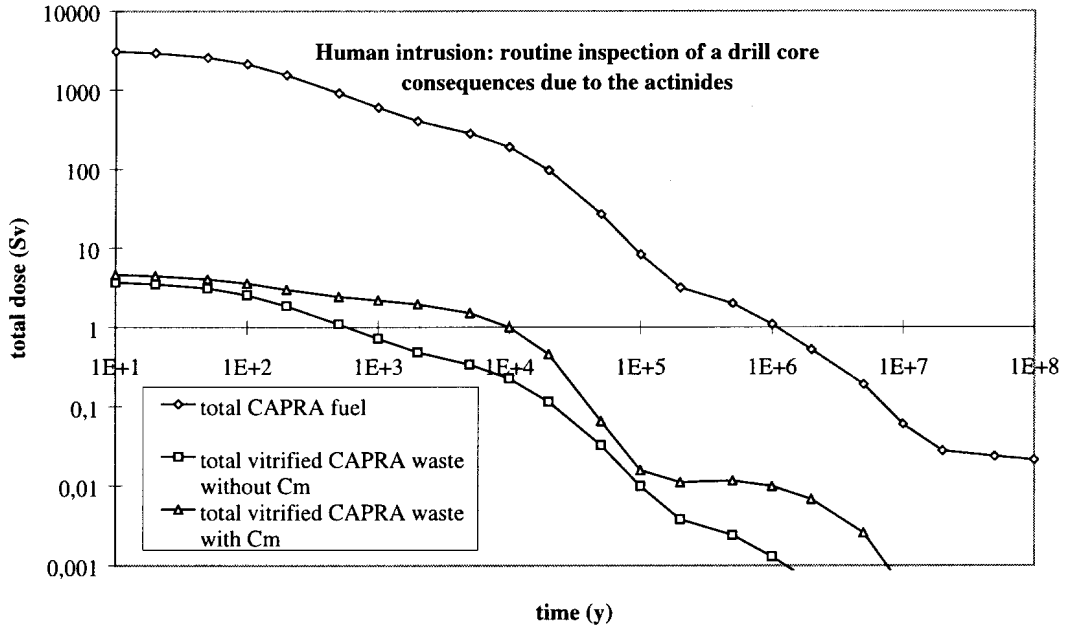


Figure 2. **Comparison of the dose due to the actinides between the disposal of spent fuel and vitrified HLW with the Cm added to it or without Cm**



Assessments of the possible partitioning and transmutation of some key radionuclides

Assessments have been made of the capability of existing and foreseen P&T technologies to separate and transmute the radionuclides identified as important. The Table 2 summarises the results of this analysis.

Conclusion

For the ground water release scenario, the dose from the different vitrified HLW types or spent fuel types is very similar. The dose is dominated by the following radionuclides: ^{79}Se, ^{126}Sn, ^{129}I, ^{135}Cs and U-decay chains. Their order of importance depends on the type of host rock and considered biosphere.

The P&T strategies considered in this study have only a minor effect on the calculated doses for the following reasons: the release of most radionuclides, especially the actinides, is strongly limited by their low solubility under deep geological conditions; the release of soluble fission products is generally proportional to burn-up and thus higher for CAPRA or EFR wastes. It is important to note that for all host rock and waste types, both vitrified HLW, and spent fuels, the doses were well below the internationally recommended limit.

For human intrusion scenarios with direct exposure to waste, the doses are much higher than those for the ground water release pathway, but the magnitude of the risk depends also on the probability of intrusion, which is difficult to assess. In case of human intrusion for hard rock and clay, the dose remains over long time periods above the deterministic effect level.

Table 2. **The partition and transmutation of key radionuclides**

Radionuclide	Environmental impact	Partitioning technology	Transmutation technology
^{126}Sn	Important in groundwater release pathway	Probably goes mainly to dissolver insolubles in reprocessing. Difficult to separate. Possible methods have not been studied.	Half times of transmutation very long even in high neutron fluxes. May be better to segregate insoluble residues
^{79}Se	Important in groundwater release pathway	Probably difficult to separate from HLW. Possible methods have not been studied.	Long transmutation half times for ^{79}Se. More ^{79}Se would be formed from ^{78}Se
^{135}Cs	Important in groundwater release pathway. ^{137}Cs also important in early human intrusion situation	Methods for separation of caesium from HLW have been developed .	Not feasible because of transmutation of other caesium isotopes, forming more ^{135}Cs
^{241}Am	Important in groundwater release pathway as parent of ^{233}U. Very important in human intrusion scenarios and on radiotoxicity grounds	Much research in progress on separation from HLW by aqueous methods. Separation from curium and lanthanides requires further development.	Could be incinerated, preferably in fast reactors. Multiple cycling & intermediate reprocessing needed. Curium would be generated.
^{237}Np	Important in groundwater release pathway as parent of ^{233}U.	Probably could be separated during conventional reprocessing	Could be incinerated, but significant amounts of ^{238}Pu would be formed
^{99}Tc	Important in human intrusion scenarios	Divided between dissolver insolubles and dissolver liquor. Could be separated from the latter during reprocessing	Could be transmuted. Several cycles with intermediate processing probably needed
^{129}I	Important in groundwater release pathway	Mainly diverted into ILW streams in reprocessing. Could be largely separated if required	Could be transmuted. Several cycles with intermediate processing probably needed. Corrosion and gaseous product could be problems

The identified key radionuclides are in general radionuclides for which partitioning and transmutation is difficult. It is clear that to obtain important dose reductions by P&T techniques, a very high efficiency of both the partitioning and transmutation is required. The partitioning efficiency needed for a consequent reduction of dose must be much higher than for today's plants.

In this study, activation products and generation of secondary MLW have not been fully studied and they might further reduce the effect of P&T. The disposal of spent Am-targets has also not been studied. Preliminary calculations have shown that their high heat output can be a problem for their disposal.

It is also clear that once a P&T strategy including CAPRA reactors is applied, one needs to pursue it till the end, which can require more than a century, because otherwise one might need to dispose off important quantities of CAPRA spent fuel.

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REFERENCES

[1] Sinclair J.E., Robinson P.C., Cooper N.S., Worgan K.J. and Williams M.J. (1994a)**,** *MASCOT and MOP Programs for Probabilistic Safety Assessment Part A: MASCOT (Version 3C)User Guide*, UK Nirex Ltd, NSS/R336, Part D 1994.

[2] Sinclair J.E., Robinson P.C., Cooper N.S., Worgan K.J., and Cliffe K.A., (1994b*) MASCOT and MOP Programs for Probabilistic Safety Assessment Part B: MASCOT (Version 3C) Technical Details*, 1994.

[3] Nirex (1995b) Post Closure Performance Assessment, *Human Intrusion and Natural Disruptive Events*, United Kingdom Nirex Limited, Science Report no. S/95/004, July 1995.

[4] Becker A**.**, Fischer H., Hofer E., Kloos M., Krzykacz B., Martens K.-H., Röhlig K.-J**.** (1997), *Evaluation of Elements Responsible for the effective Engaged dose rates associated with the final STorage of radioactive waste: EVEREST project, Volume 3a: Salt formation, site in Germany*, Commission of the European Communities, Luxembourg, EUR 17449/3a EN, 1997.

[5] Marivoet. J., Volckaert G., Wemaere I., Wibin J.(1997), *Evaluation of Elements Responsible for the effective Engaged dose rates associated with the final STorage of radioactive waste: EVEREST project, Volume 2a: Clay formation, site in Belgium, Commission of the European Communities*, Luxembourg, EUR 17449/2a EN, 1997.

[6] ACRI (1997**)**, *PORFLOW A software tool for multiphase fluid flow, heat and mass transport in porous media*, User's manual, Version 3.07, March 1997.

[7] Mobbs S.F**.**, Klos, R.A., Martin J.-S., Laurens J.-M., Winters K.H., Beally J.M., Dalrymple G., *PACOMA: Performance assessment of the confinement of medium-active and alpha-bearing wastes. Assessment of disposal in clay formation in the United Kingdom*, Commission of the European Communities, Luxembourg, EUR 13143 EN, 1991.

THORIUM CYCLE AS A WASTE MANAGEMENT OPTION

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Abstract

As part of the European Union Fourth Framework Programme on “New Concepts” an investigation is made on the thorium cycle from the perspective of nuclear waste minimisation. The objective of the work is a re-assessment of thorium cycles in the context of limitation of nuclear waste production and prospects for waste burning. The aim is to obtain a review of the major steps of the fuel cycle, focusing to the waste aspect. A restriction is made to European reactor types: PWR and FBR and the Fast Energy Amplifier (FEA).

A number of six working packages has been defined, including: 1. Mining, 2. Fuel fabrication, 3. Reactor Assessments, 4. Reprocessing, 5. Residual risks of disposal, 6. Non-proliferation aspects. This 3-years programme is in the final stage.

The results of the above-mentioned working packages are discussed. A synthesis of the results shows that there are important advantages of thorium cycles with respect to the waste issue. Long-lived radiotoxicity of mining waste is expected to be relatively small, which leads to more manageable waste. Fabrication of Th fuels is comparable with MOX fabrication methods as long as fresh Th, U and recycled Pu are used. Recycling of U, however, needs remote handling. Open cycles are possible in PWRs, but require make-up fuel. To reduce the radiotoxicity of PWR waste, make-up fuel like ^{233}U or highly-enriched ^{235}U should be added to Th. Advantages are seen during the first 10,000 years of storage. The long-term risk of directly stored fuel in a thorium matrix is still not known very well, but there are speculations on improved performance. Further experimental work is needed to clarify this point. Recycling gives a further reduction of radiotoxicity up to 10,000 to 50,000 years of storage.

Th-assisted Pu burning in a PWR is an attractive option with respect to mass reduction of Pu, which could be twice that of U/Pu MOX in a 100% core loading. In open cycles the produced ^{233}U is somewhat disturbing from the point of view of proliferation; spiking with ^{238}U could help. For all scenarios a quantitative non-proliferation metric is developed.

Fast reactors and accelerator-driven systems, like FEA, offer the possibility of a closed Th cycle without make-up fuel, reducing mining needs and risks. Full recycling of actinides gives impressively low radiotoxicity results. Initially, these systems could be used for Th-assisted Pu-burning and simultaneous ^{233}U breeding, providing fuel for a new generation of low-actinide-waste producing energy systems.

It is expected and recommended that the introduction of the thorium cycle will be realised in steps. A recommended first step could be once-through Pu-burning in LWRs with a mixture of thorium and plutonium oxide. To this end an irradiation experiment has been proposed and further work on nuclear and thermodynamic data.