SCK•CEN
STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

ONDRAF/NIRAS

Common Scientific Position Paper

Partitioning & transmutation as a potential complement to geological disposal for the safe long-term management of high-level and long-lived radioactive waste

Executive summary

More than 40 years of research by ONDRAF/NIRAS and SCK•CEN has shown that geological disposal in poorly indurated clays of long-lived and high level radioactive waste (including spent nuclear fuel if declared as waste) would provide a safe, feasible long-term management solution. In the expected evolution of the disposal system, only the long-lived radionuclides could reach the biosphere. Since the diffusion of radionuclides through the geological barrier is a very slow process, even the releases of the most mobile of those long-lived radionuclides, i.e. the activation and fission products, will be spread over a long period of time, leading to radiation exposure to man and environment significantly lower than the regulatory limits. The mobility of the actinides being especially low, these would contribute only marginally to the dose, the latter being thus dominated by the long-lived activation and fission products. In the case of an intrusion scenario, corresponding to the unlikely event of human ingress into the repository after its closure, the exposure is defined by the waste's radiotoxicity. For a given host formation the disposal facility's footprint is mainly defined by the final volumes and the thermal output of the disposed waste.

Partitioning and Conditioning (P&C) and Partitioning and Transmutation (P&T) of spent nuclear fuel could provide benefits to geological disposal by reducing the radiotoxicity and/or thermal output of the spent fuel. These techniques are actually not considered to be practically applicable to already conditioned (e.g. vitrified, bituminised,…) waste.

SCK•CEN's MYRRHA-project, which the Belgian government has decided to support, will investigate the feasibility of transmutation (mainly of minor actinides) by an accelerator driven system (ADS) at pilot scale. In combination with advanced reprocessing and the re-use of plutonium, preferably in

future reactors (fast neutron systems), a fuel cycle with an ADS system could provide benefits for the geological disposal, both by reducing the radiotoxicity of the waste to be disposed of and by reducing the required footprint of such a facility. These fast neutron reactors are not planned in the current Belgian energy policy.

However, the potential application of advanced fuel cycles in the future cannot eliminate the need for a long-term solution (up to hundreds of thousands of years – due to the presence of long-lived fission products) for the safe management of the waste produced by the advanced fuel cycle, as well as the already produced high-level waste and the low and intermediate level long-lived waste. Such a safe management solution can be provided by geological disposal.

ONDRAF/NIRAS and SCK•CEN have worked closely together already for decades to demonstrate the safety and feasibility of geological disposal, and will continue to do so in the future.

Starting from their both respective legal missions, ONDRAF/NIRAS and SCK•CEN will continue to work together to develop a sustainable waste management strategy for the Belgian high-level and long-lived radioactive waste inventory, taking into consideration novel approaches that could favour its implementation.

This Scientific Position Paper is realised in context of the communication mission of ONDRAF/NIRAS and SCK•CEN.

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Common Scientific Position Paper

This document is a common Scientific Position Paper by ONDRAF/NIRAS (the Belgian National Agency for Radioactive Waste and enriched fissile materials) and SCK•CEN (the Belgian Nuclear Research Centre) regarding the research on geological disposal and on partitioning & transmutation of high-level and long-lived radioactive waste.

The document is written within the present Belgian context i.e.:

- the execution of the law on the nuclear phase-out;
- the suspension on reprocessing of spent commercial nuclear fuel, and the currently available open options for future spent fuel management;
- the future implementation of the proposed policy regarding the long term management of high-level and long-lived radioactive waste i.e. geological disposal on the Belgian territory;
- the decision of the Belgian government to support MYRRHA in which transmutation of long-lived actinides is the flagship research topic.
- the communication mission of ONDRAF/NIRAS and SCK•CEN.

In Belgium, radioactive waste is produced by activities relating to the nuclear fuel cycle (electricity production, fuel fabrication and reprocessing[1]), to research or to medical and industrial applications of radioactivity. The Belgian radioactive waste is classified in three categories: A, B and C which can be linked to the international waste classification scheme. Category A waste corresponds to low and intermediate level short-lived waste (LILW-SL) whose final management policy consists of surface disposal in the municipality of Dessel. Category B waste is low and intermediate level long-lived waste (LILW-LL). Category B waste are conditioned in very diverse forms (cement, bitumen, glass, metals); a large fraction of the already produced B waste originates from reprocessing of spent nuclear fuel, either at the former Eurochemic plant (Dessel, Belgium), or at the Orano facility in La Hague (France). Category C waste is heat-emitting high level waste (HLW). Current category C waste consists of vitrified waste resulting from the reprocessing of spent nuclear fuel[2]. Potential future category C waste is spent nuclear fuel (mainly from the nuclear power plants at Doel and Tihange) which has currently not yet been declared as waste.

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[1] Reprocessing is the mechanical and chemical processing of spent nuclear fuel, with the aim of separating potentially useful elements (currently uranium and plutonium; maybe others in the future) from unusable material (such as (most) fission products) that is then further treated as waste.

[2] At the start of nuclear energy production in Belgium, spent nuclear fuel was sent to La Hague (France) for reprocessing. This process was halted in 1993 after a parliamentary debate and, since then, no new reprocessing contracts for spent nuclear fuel were closed. The uranium and plutonium which was recovered from reprocessing were re-introduced into the Belgian nuclear fuel cycle (either as UOX or MOX fuel). The restart of reprocessing for commercial spent fuel requires a decision by the federal government.

From radiotoxicity[3] point-of-view, category A waste is the least radiotoxic, containing mostly short-lived (half-life < 30 years) beta or gamma emitting radionuclides. These nuclides result mostly from the fissioning of uranium (U) or plutonium (Pu) isotopes in a reactor, the activation of structural materials (metals, concrete…) in nuclear installations, or other activities related to applications of radioactivity (research, medicine…). Category B waste contains also (sometimes large quantities of) alpha emitting radionuclides, and higher quantities of beta and gamma emitting radionuclides. The alpha emitters are mainly the isotopes of uranium and of transuranic elements. The transuranic elements are plutonium and the minor actinides, namely neptunium (Np), americium (Am) and curium (Cm). They are produced within the fuel of nuclear reactors. Some of those have very long half-lives and are very radiotoxic upon ingestion or inhalation. Category C waste contains the largest amounts of both alpha- and beta/gamma-emitting radionuclides, and therefore has the highest radiotoxicity and the highest emission of heat resulting from radioactive decay processes. Both category B and C waste also contain long-lived (beta or gamma emitting) activation and fission products, with half-lives exceeding 100,000 years.

The current or proposed Belgian policies for the long-term management of radioactive waste depend on the classification and type of waste.

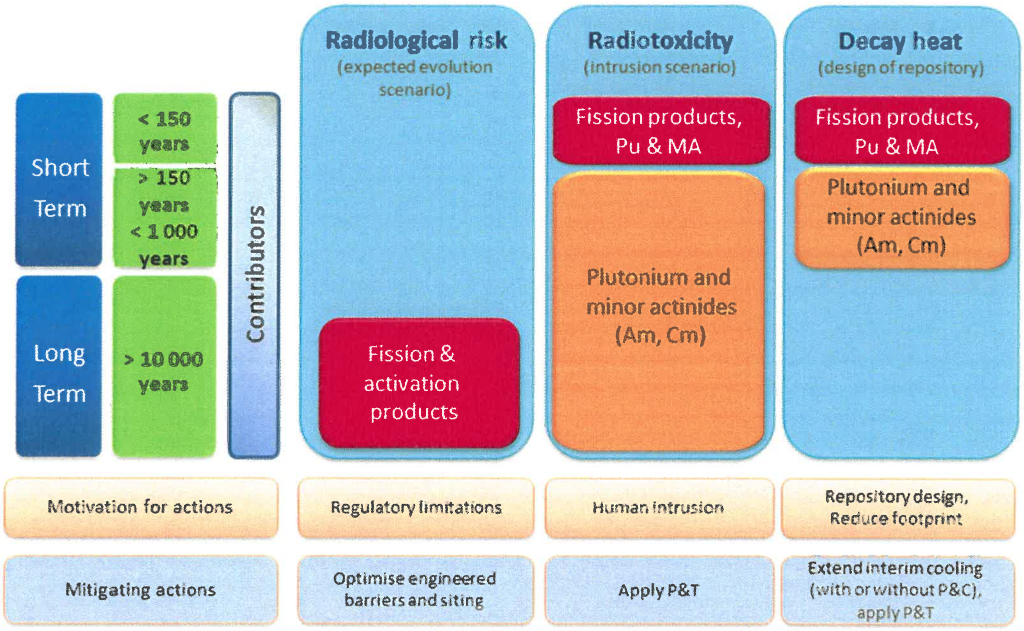
- For category A-waste, which is short-lived, a surface disposal facility will be built in Dessel (it is now in the licensing phase).
- For category B and C waste, geological disposal on the Belgian territory has been proposed by ONDRAF/NIRAS as a safe, feasible and sustainable solution; this proposal should still be formally enforced to become a National Policy. More than 40 years of research by ONDRAF/NIRAS and SCK•CEN, in line with the international recommendations, has indeed shown that geological disposal of these wastes in poorly indurated clays would provide a safe and feasible long-term management solution. Safe disposal of these wastes in a geological disposal facility (also called a geological repository), is based on the concept of containing and isolating the waste for a sufficient amount of time by a suitable choice of multiple engineered and natural barriers to allow radioactive decay to decrease the radiological hazard so that the eventual releases to the environment are well below the regulatory limits. Safety assessment calculations in the case of an expected evolution scenario have shown that only the long-lived radionuclides could reach the biosphere. Since the diffusion of radionuclides through the geological barrier is a very slow process, even the releases of the most mobile of those long-lived radionuclides, i.e. the activation and fission products, will be spread over a long period of time, leading to radiation exposure significantly lower than the regulatory limits. The mobility of the actinides being especially low, these would contribute

[3] The harmfulness of a radionuclide or a radioactive material upon ingestion or inhalation is called its radiotoxicity.

only marginally to the dose, the latter being thus dominated by the long-lived activation and fission products.

How do radionuclides impact the design and the performance of geological disposal?

There are three major ways in which radionuclides can impact the design and the performance of geological disposal: by their radiological risk, their radiotoxicity and their heat output.



The first two relate to the long-term safety of a geological disposal system. The long-term safety of a geological disposal system is ensured by a number of engineered and natural barriers. These barriers will isolate and contain the waste so that radioactive decay can decrease the radiological hazard so that the eventual releases to the environment are below the regulatory limits.

Generally speaking, there are two types of scenarios by which the radionuclides in the disposed waste can reach the biosphere and lead to radiological doses. The first one relates to the expected long-term evolution of the geological disposal system, while the other one relates to the unlikely event of human intrusion into the repository at a certain point in time after its closure.

- In the expected evolution of a geological repository, the radioactive waste and engineered barriers will slowly degrade in time and release the radionuclides. In clay-based repositories, as currently studied in the Belgian context, these radionuclides will migrate slowly, by diffusion, through the host rock and eventually parts of them could reach an aquifer (many radionuclides will have decayed to stable nuclides before reaching an aquifer). The use of water from this aquifer by the population (as drinking water, for irrigation...) finally results in a certain radiation exposure significantly lower than the regulatory limits. This is called the radiological risk and can be virtually completely ascribed to a limited number of mobile fission and activation products with a long half-life (namely Se-79, I-129, Cl-36, C-14,...).

- The second type of scenarios involves an (unlikely) involuntary human intrusion into the geological repository, e.g. by drilling a borehole into a waste package. This would result in a high radioactive dose to the intruders and/or a radioactive dose to nearby inhabitants. In this type of scenario, the risk is defined by the radiotoxicity of the waste. In spent fuel, plutonium is the major contributor to the radiotoxicity, followed by the minor actinides (neptunium, americium and curium).

The heat output of category C waste determines the distance between the disposal galleries and has therefore a significant effect on the cost and footprint of the repository. The thermal output is initially determined by fission products Cs-137 and Sr-90, and to a minor extent by Am-241 and some Pu isotopes. After a few hundred years of decay, americium and plutonium are the major contributors to the thermal output.

Category B waste does not emit enough heat to impact the layout.

Up to now, the fuel assemblies which have been irradiated in nuclear power reactors (referred to as spent fuel) are not yet declared as radioactive waste and are stored at the sites of the Belgian nuclear power plants.

In a comparative study by the Federal Public Service Economy[4], 6 scenarios for the management of Belgian spent fuel[5] were determined considering the phase out law (law of 31 January 2003) and a number of other hypotheses. Five scenarios are described hereunder[6].

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[4] FOD Economie, K.M.O., Middenstand en Energie - Algemene Directie Energie - Afdeling nucleaire toepassingen (2014). Vergelijkende studie van de beheerstrategieën van de Belgische splijtstof - Deel 2.

[5] The Belgian spent fuel (current and foreseen) inventory contains UOX (uranium oxide) fuel and a small amount of MOX (mixed (= uranium + plutonium) oxide) fuel. The UOX fuel is mainly produced from enriched natural uranium, but can also be produced from reprocessed uranium.

A. Direct disposal of spent fuel (no further reprocessing)

In this scenario, the spent nuclear fuel (UOX and MOX) is not reprocessed. It will be stored for multiple decades (at least 50 years) in order to cool down, after which it will be conditioned and disposed of in a geological repository (as C-waste).

B. Full reprocessing of spent nuclear fuel

In this scenario, all spent nuclear fuel (UOX and MOX) is reprocessed. The recovered uranium can be re-enriched to be used in Belgian or foreign nuclear reactors. The recovered plutonium would be sold to other (foreign) parties to be used in their (civil) nuclear reactors. The reprocessing produces vitrified high-level waste (C-waste) and compacted metallic intermediate-level waste (B-waste).

C. Partial reprocessing of spent nuclear fuel

In this scenario, only a part of the spent nuclear fuel (part of UOX and all of MOX) is reprocessed. Here, it is considered that both the recovered uranium and plutonium would be re-used in the Belgian nuclear power plants until the last one is shut-down in 2025. The reprocessing produces vitrified high-level waste (C-waste) and compacted metallic intermediate-level waste (B-waste). The spent fuel that is not reprocessed is furthermore declared as waste.

D. Partitioning and conditioning (P&C)

In this scenario, the spent nuclear fuel (UOX and MOX) would be separated by advanced reprocessing techniques into multiple material streams. Some of them could contain re-usable materials (currently U and Pu; potentially other radionuclides), while the others would have to be conditioned as waste. Specific conditioning techniques could be applied depending on the chemical nature of the waste streams. The idea is to optimise the conditioning matrix and/or management strategy (e.g. longer cooling time before disposal for heat emitting radionuclides) based on the specific properties of the waste stream. Advanced separation techniques are not yet applied at an industrial scale, although a number are already used at a small scale. This scenario thus starts with an R&D phase to study and select a strategy for the optimal partitioning and conditioning of spent fuel, and develop it at industrial scale.

E. Partitioning and transmutation (P&T)

This scenario builds further on the previous one (P&C), but additionally considers an industrial scale transmutation facility that is capable of transmuting actinides into shorter-lived radionuclides. For Belgium, this scenario focuses on an Accelerator Driven System (ADS) whose goal is to treat effectively the waste while it considers production of electricity as a by-product

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[6] The sixth one, additional research to explore other management options, is not discussed here.

(in contrast with other Generation IV-type fast reactors[7]). A P&T fuel cycle is expected to produce vitrified high-level waste (C-waste) and compacted metallic intermediate level waste (B-waste) from the reprocessing of spent UOX and MOX fuel, vitrified high-level waste (C-waste) and possibly some B-waste from the reprocessing of spent ADS fuel and new, secondary waste streams from the ADS spallation target. This scenario requires R&D on different distinct steps in the fuel cycle, among which, partitioning of the spent fuel and selection of an appropriate minor actinides extraction process, fabrication of dedicated fuels and/or targets, demonstration of transmutation of minor actinides in an accelerator-driven system and conditioning of waste produced in the various steps of this fuel cycle. These various steps would not necessarily be implemented on the Belgian territory.

What is transmutation and what role can MYRRHA play in its development?

In general terms, the goal of transmutation is to convert a hazardous radiotoxic material into a material that is less radiotoxic, by bombardment with fast neutrons. The aim of the process is to transmute heavy nuclei (that typically decay by alpha decay with long half-lives) into lighter nuclei (which typically decay by beta decay with shorter half-lives). In this way, the resulting waste disposed of in a geological repository is less hazardous in the case of a human intrusion. Additionally, the thermal output of the waste can be reduced, which is beneficial for the footprint and cost of the repository.

The application of partitioning and transmutation is limited (in practice) to the actinides contained in spent nuclear fuel. For other nuclides or other waste types, partitioning and transmutation is presently not expected to be practically feasible.

The scientific feasibility of P&T has been demonstrated. However, significant R&D efforts and commissioning of demonstration facilities at sufficient scale are still required to achieve viable industrial P&T processes and to improve the reliability of the estimations on ecological, social and economic impacts. Europe has identified four building blocks to be developed at engineering scale: 1) advanced reprocessing of spent light-water reactor fuels; 2) fabrication of dedicated transmutation fuels; 3) transmutation in a pre-industrial scale transmuter and 4) reprocessing of the transmutation fuel.

MYRRHA is a planned prototype demonstration facility responding to the 3rd building block listed above for, amongst other applications, studying transmutation of minor actinides (primarily, Am) from spent nuclear fuel by operating as a testing facility for various materials of the fuel matrix and compositions in minor actinides.

The Belgian government has decided to support the realisation of the MYRRHA research infrastructure on SCK•CEN's site. This project investigates the feasibility of transmutation by an ADS at pilot scale.

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[7] Generation IV reactors are a set of innovative reactor designs that are currently being researched (for example by the Generation IV International Forum (www.gen-4.org)).

Given this decision, it is important to highlight the main impacts of a P&C/P&T scenario on the proposed long-term waste management strategy for the B&C waste (geological disposal) for Belgium.

- Impact of P&T on the radiotoxicity of spent fuel

A few hundred years after unloading the spent nuclear fuel from the reactor, plutonium becomes the major contributor to its radiotoxicity, followed by the minor actinides. The radiotoxicity of the fission and activation products is initially higher, but decreases more rapidly. In a P&T scenario the minor actinides could be recycled in the ADS system. The ultimate goal here is to produce waste of which the radiotoxicity decreases faster than the initial spent fuel. The degree by which partitioning and transmutation will have an impact on the radiotoxicity of the spent fuel strongly depends on the efficiency of the P&T fuel cycle. A separation of Pu together with its complete fissioning (preferably in Generation-IV fast reactors, or in an ADS), could reduce the spent fuel's radiotoxicity to 10% of its initial value. If additionally a complete transmutation of americium would be achieved (preferably in an ADS), this would lead to a further reduction of the radiotoxicity to about 1% of its initial value. Such a radiotoxicity decrease would reduce the radiological risk associated with human intrusion in a repository.

- Impact of P&T on the thermal output of spent fuel

Initially, the thermal output of spent nuclear fuel is determined by the activity of short-lived fission products Sr-90 and Cs-137. After a few hundred years of decay, the transuranic elements (mainly Am-241, but also some Pu isotopes) are the major contributors to the thermal output. An advanced separation process, removing these nuclides from the spent fuel to reuse them as fuel in an ADS system (in case of Am and Pu), or by adopting a separate waste management strategy (in case of Sr and Cs – with prolonged interim storage), would thus reduce the thermal output of the waste to be disposed of. A reduction in thermal output significantly impacts the footprint of a geological repository, thereby significantly reducing its costs.

- Impact of P&T on the long-term radiological exposure

In the expected evolution of a geological disposal system, the radiological exposure can be virtually completely ascribed to a limited number of fission and activation products with a long half-life (namely Se-79, I-129, Cl-36, C-14…). Despite their high radiotoxicity, the actinides have only a limited impact in terms of radiological exposure, thanks to their immobility in the engineered barrier and the clay host rock environment. A P&T scenario leads to the production of additional fission and activation products and thus to an increase of their inventory. The transmutation of fission and activation products itself is actually not considered to be practically feasible. Therefore, irrespective of a P&C/P&T scenario, a long-term management strategy for (the waste containing) these nuclides remains necessary. Given the long half-life of these nuclides, geological disposal is the only internationally accepted management solution.

- Impact on the already produced category C and B waste

In vitrified waste, the radionuclides consist mainly of fission products and all of the minor actinides (in minor amounts compared to the FPs) that are homogeneously dispersed in a stable glass matrix. Moreover, these wastes produce very high radiation levels. It is therefore actually not considered to be practically feasible, nor recommendable, to recover the radionuclides (i.e. reverse-engineer the vitrification process) from the waste matrix to apply P&C or P&T. The same argumentation applies for the already produced category B waste (such as bituminised waste), which contains even less actinides. Therefore, also for the already produced conditioned B&C waste, geological disposal is necessary as a safe waste management solution.

- New waste forms produced in a P&C/P&T scenario

Next to high-level and long-lived waste comparable to existing waste, an advanced fuel cycle in which P&C/P&T is applied will also produce new types of category B waste, thereby increasing the amount of this waste type in the Belgian inventory. Within a P&C scenario specific tailored matrices can be developed in which specific waste streams could be immobilised. Apart from the already mentioned Cs/Sr waste stream, one could also consider a specific waste matrix targeting (e.g.) the long-lived fission products which would be the most mobile nuclides within a geological repository, thereby also contributing to the long-term safety of such a repository. Of course, this means that matrices should be developed that have a similar performance as the UOX matrix.

Conclusion

Independently of the chosen scenario of the management of spent nuclear fuel, a long-term solution (up to hundreds of thousands of years – due to the presence of long-lived fission products) is in any case needed for the safe management of radioactive waste resulting from the nuclear fuel cycle, as well as of the already produced high-level waste and the low and intermediate level long-lived waste. The potential application of advanced fuel cycles in the future cannot eliminate this need. Such a safe management solution can be provided by a geological repository. ONDRAF/NIRAS and SCK•CEN have worked closely together for decades already to demonstrate the safety and feasibility of such a solution, and will continue to do so in the future.

An advanced fuel cycle which includes P&C/P&T in addition to the re-use of U/Pu, e.g., in Generation-IV fast reactors or ADS systems, could provide benefits for the geological disposal, both by reducing the radiotoxicity of the waste to be disposed of, by proposing tailored waste management strategies for specific nuclides and waste streams, and by positively impacting the required footprint of such a repository. These fast neutron reactors are not planned in the current Belgian energy policy.

However, since such a fuel cycle is still in the R&D phase, the actual benefits for both geological disposal in particular and for the whole fuel cycle in general are nowadays difficult to estimate exactly. In order to have a complete picture of the pros and cons of an advanced fuel cycle with partitioning and transmutation, a thorough and realistic full lifecycle analysis is necessary. Such a lifecycle analysis should also take into account aspects of radiation protection, economics, use of natural resources, planning and societal acceptability. The MYRRHA project will contribute greatly to increase the knowledge on transmutation within Europe.

Starting from their both respective legal missions, ONDRAF/NIRAS and SCK•CEN will continue to work together to develop a sustainable waste management strategy for the Belgian high-level and long-lived radioactive waste inventory, taking into consideration novel approaches that could favour its implementation.

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The Belgian Nuclear Research Centre (SCK•CEN) devotes itself every day to developing peaceful applications of nuclear energy. The SCK•CEN aims to develop innovative technologies that provide solutions for the social issues and requirements in the field of nuclear energy and ionising radiation.

ONDRAF/NIRAS, the Belgian Agency for Radioactive Waste and Enriched Fissile Materials, serves the community. It manages all radioactive waste in Belgium, now and in the future, by developing and implementing solutions with due respect for society and the environment.