FRENCH WASTE MANAGEMENT STRATEGY FOR A SUSTAINABLE DEVELOPMENT OF NUCLEAR ENERGY

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Abstract

The production of nuclear energy in France has been associated, since its inception, with the optimisation of radioactive waste management, comprising the partitioning and the recycling of recoverable energetic materials, volume reduction, the conditioning, and the disposal of the end waste.

The public's concern regarding its long-term management led the French Government in 1990 to delay the implementation of the geological disposal and to prepare a law passed on December 30, 1991, requesting in particular the study of solutions and processes for:

- minimising the quantity and the hazardousness of waste, via partitioning and transmutation;
- disposing the waste either reversibly or irreversibly in deep geological formations;
- waste conditioning and long-term interim storage.

The law stipulates that, by 2006, the research results obtained should be documented in the form of a general report to be submitted to members of parliament. The law also created the National Evaluation Commission to supervise the research work on an annual basis.

At the time of the implementation of measures comprised in the Law, the Government commissioned CEA to conduct research in the area of Line 1 (partitioning and transmutation) and Line 3 (conditioning and interim storage) and Andra on Line 2 (geological disposal).

The feasibility of partitioning did not appear easily accessible at the time the research began. Its feasibility was demonstrated in 2001 following a series of tests conducted on actual solutions of dissolved spent fuel, in the CEA's Atalante installation at Marcoule. The 2002-2005 programme encompasses technological demonstrations, with representative process equipment, and an economic evaluation of the industrial implementation of partitioning.

Studies on transmutation, which were initiated before the Law of 1991, rapidly led to concluding that transmutation of minor actinides (Americium, Curium, and Neptunium) was feasible in particular in fast neutron systems.

The work on transmutation is now focusing on a demonstration of technology feasibility and this is considered in relation with the development of future nuclear energy systems capable of minimising their production of long-lived radioactive waste, within the framework of the Generation IV International Forum.

For supporting the objective of a sustainable nuclear energy, such Generation IV systems are envisioned with fast neutrons and a global recycling of all actinides from the spent fuel to be capable of burning fissile materials, breeding fertile materials and transmuting minor actinides. Furthermore, a group management of actinides is aimed at achieving an appropriate resistance to risks of proliferation.

Interim storage is a mode of package management ensuring, by design, the protection of waste package and their recovery at a later date, under safe and technically established conditions. It is a factor of flexibility for all considered waste management strategies, in the medium or the longer term. The work done in the context of Line 3 of the 1991 Law has comprised the identification and the development of long-term interim storage concepts. The 2003-2005 programmes are targeted at the technological development of canisters and specific interim storage components, using demonstrators and qualification programmes.

Important results are now available, on the one hand concerning the possibility of significantly reducing the quantity and the radio-toxicity of long-lived waste, and also relating to the modes of waste conditioning and long-term interim storage facilities.

With these results combined with those obtained by the Andra on geological disposal, technical solutions that can be implemented progressively, will be presented for decision in 2006 to manage the high level and long-lived radioactive waste generated by the LWRs. Most of these solutions are seeds for the advanced fuel cycle processes needed to manage the group actinides streams and the waste forms generated by vision of sustainable nuclear energy that Generation IV systems constitute. These scientific achievements concerning waste management must be accompanied by a democratic and political debate at the French parliament.

Introduction

The production of nuclear energy in France has been associated, since its inception, with the optimization of radioactive waste management, comprising the partitioning and the recycling of recoverable energetic materials, volume reduction, the conditioning, and the disposal of the end waste. ¹

Low-activity and short-lived waste (approximately 15 000 m³/year) mainly originates from the operation of nuclear generating plants, is managed by the Andra (the French National Agency for Radioactive Waste Management) and disposed on surface at the Centre de l'Aube (and formerly at the Centre de La Manche, closed down since 1997).

The actual industrial reprocessing operations remove and recycle the plutonium, which is a highly energetic material and also the primary contributor to long-term radiotoxicity, and condition the long-lived waste in a safe and durable manner, and in a very small volume (130 m³/year for HLW glasses and 320 m³/year for medium-activity wastes).

Nevertheless the decay period of the waste remains long, and the public's concern regarding the long-term management of high level and intermediate level waste made the French Government in 1990 to delay the implementation of the geological disposal and to prepare a law, passed on December 30, 1991, requesting in particular the study of solutions and processes for:

- minimising the quantity and the hazardousness of waste, via partitioning and transmutation;
- either reversibly or irreversibly disposing the waste in deep geological formations;
- waste conditioning and long-term interim storage.

At the time of the implementation of measures comprised in the Law, the Government commissioned CEA to conduct research in the area of Line 1 (partitioning and transmutation) and Line 3 (conditioning and interim storage) and Andra on Line 2 (geological disposal). We will present here an update on the progress made by the research. Research has been conducted in a very sustained way since 1992 and benefits from major co-operations in France (EDF, AREVA, Andra, CNRS, universities...), Europe, and internationally.

For line 1: partitioning and transmutation, the actual industrial reprocessing operations remove and recycle the plutonium, so the reprocessing greatly reduces both the waste radiotoxicity and its lifetime (from a few hundred thousand years in an open cycle to a few tens of thousand years in a closed cycle). In the most advanced fuel cycles using fast-spectrum reactors and extensive recycling, it may be possible to reduce the radiotoxicity of all wastes such that the isolation requirements can be reduced by several orders of magnitude (e.g., for a time as low as 1 000 years) after discharge from the reactor. This would have a beneficial impact on the design of future repositories and disposal facilities worldwide.

Line 1 researches concerns future wastes which could be produced by enhanced reprocessing mainly in the perspective of sustainable development of nuclear energy.

1. France produces, each year and per capita, approximately 1 kg of waste originating from the production of nuclear energy, compared to 2 500 kg of industrial waste, which includes 100 kg of toxic chemical waste.

The management of nuclear waste and the access to cost effective natural uranium resources will assuredly be major challenges for nuclear energy systems over the 21st century, taking into account the rapid growth of primary energy demand and the increased share of nuclear power to meet both needs of electricity generation and hydrogen production for the transports, while limiting greenhouse gas emissions.

Legacy wastes and wastes which are actually produced in France by reprocessing are ultimate wastes non relevant of line 1 operations, in fact researches have shown that nuclides retrieval from actual conditioned wastes would be technically very difficult and non interesting from wastes volume and economics points of view.

These wastes are concerned by studies relevant of line 2: Disposal (Andra) and line 3: Conditioning and interim storage.

Legacy and actual wastes

The R&D work in line 3 comprises two chapters:

- conditioning, which entails the making of and the knowledge base about the waste package: development of radioactive material conditioning processes, canisters, package characterization and long-term behaviour studies;
- interim storage, focusing on the definition and the qualification of long-term interim storage concept designs, on surface or subsurface.

Conditioning

Treatment and conditioning processes

Developments made in the area of waste treatment and conditioning were targeted at ensuring the availability of qualified processes that could be applied to historic waste to be recovered or to improve (volume reduction) a number of existing treatment processes: waste vitrification in cold-crucible, bituminization of treatment effluent residues currently stored.

Overall, these developments have reached their objectives and are, for the most part, complete.

Canisters

A major effort is being made in the area of development and qualification of canisters for long-term interim storage or disposal. The canister is an external envelope around the internal primary package, providing an additional barrier (useful for handling, recovery and reopening [reversibility], tightness, durability, etc.).

Developments include in particular:

- Canisters for dry interim storage of spent fuel: ² in order to guarantee the next recovery step under safe conditions, each fuel assembly is placed within an individual stainless-steel case kept under inert (helium) atmosphere; cases are then placed within a canister developed for interim storage. The technological canister demonstrators will be available at Marcoule in 2004. CEA, EDF, and Andra are also developing, following similar principles, a disposal canister compatible with interim storage. The demonstrator will also be available at Marcoule in 2004 for qualification.
- Canisters for interim storage of medium-activity long-lived waste:
 - Concrete canisters with a holding capacity of one to several primary packages, these canisters conceived with Andra are common for storage and disposal;
 - Primary canisters ensuring physico-chemical compatibility between the waste and the surrounding canister.

The technological demonstrators will be available at Marcoule in 2004.

Package long-term behaviour studies

The objective of the conditioning being to ensure durable confinement for all the steps in the package management, it is necessary to establish scientific and technical ground for the prediction of the package long-term behaviour, and to confirm that the relevant functions are provided, in particular confinement, handling, and recovery.

The work done in recent years has provided the scientific base of a true science on long-term behaviour, with detailed modeling and experimental validation of the principal phenomena at work considering all the different types of packages (glass, concrete, bituminized, compacted waste, spent fuel), and of the evaluation of their performance with regard to the durability of confinement in interim storage (physico-chemical evolution of the materials and prediction of their state at the time of recovery and handling operations) or in disposal (in particular under typical conditions of alteration by water in the very long term). The long durability of glass, in particular, has been established.

All the phenomena affecting long-term behaviour for each type of packages are integrated into qualified "operational models" which are used in global studies looking into the long term waste management facilities operation and behaviour over time (interim storage or disposal). These programmes benefit from the expertise of international experts in the fields.

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^{2.} The French downstream strategy does not consider spent fuel to be a waste form, since this would be contradictory to the goal of minimising its long-term hazardousness; nevertheless, in order to explore all the various options, the research conducted under Line 3 on conditioning and interim storage considers long-lived radioactive waste (vitrified high-activity long-lived waste, all medium-activity waste), and also spent fuel, as a whole. In 1992, the CEA undertook, in cooperation with EDF, COGEMA and Andra, a study of conditioning, interim storage, and disposal of spent fuel, which resulted in a report in 1996.

Package characterization and inspection

Research conducted has resulted in the development of very thorough methods and systems of package characterization and inspection, via non-destructive measurements or via samplings and radio-chemical analyses, in order to confirm that any package can meet the acceptance criteria developed for the interim storage or disposal facilities.

Interim storage

Interim storage is a mode of package management ensuring, by design, the protection of waste package and their recovery at a later date, under safe and technically established conditions.

By design, the interim storages can be constructed and operated to accommodate the long term ("aptitude séculaire") interim storage of waste package. The feasibility of such interim storage facilities is demonstrated, and site characteristics being used are sufficiently broad for conducting the research in a generic way.

Preliminary design studies of two facility concepts, on surface and subsurface, will be achieved at the end of 2004.

Geological Disposal

Researches conduct by Andra must enable the evaluation of disposal feasibility in clay. These programmes include study and presentation of a solution for geological disposal with scientific questions which have been addressed.

The feasibility evaluation is based upon a characterization of geological medium with global models founded on results obtained in Bure underground laboratory. Main questions which will be addressed are absence of fracturation, comprehension of the behaviour of altered zone and confirmation of favorable hydrological and geochemical properties. A drilling programme achieved in 2004 brings data on these points in order to demonstrate the geological disposal feasibility.

Future Wastes

Studies on partitioning and transmutation aim at isolating the most radiotoxic long-lived elements present in the waste (primarily minor actinides, which are vitrified with the fission products), then at transmuting them through recycling in nuclear reactors, in order to change them into non-radioactive or shorter-lived elements.

Commercial processing-recycling of plutonium is the first necessary step, the partitioning of minor actinides (americium, curium and neptunium), followed by their transmutation, would reduce to a few hundred years the time necessary for the radiotoxicity of the vitrified waste to become similar to that contained in the natural uranium ore originally used.

Such "light glass" packages will be the future ultimate waste form destined to the geological repository.

Partitioning

The on-going programme since 1992 comprises two chapters: *PURETEX* (which was completed in 1998) and *ACTINEX* (partitioning).

The PURETEX programme goal was the minimization of the waste stream originating from the spent-fuel processing operations in the UP3 plant at La Hague, commissioned in 1989, the aim being to reduce the volume of medium-activity waste and diminish the activity released in liquid and gaseous effluents.

Overall, results obtained entail a threefold reduction in the volume of solid waste and a tenfold reduction in the activity of liquid effluents since the start of the reprocessing plant.

The *ACTINEX* programme deals specifically with the partitioning of the long-lived radionuclides contained in the waste.

The feasibility of partitioning did not appear easily accessible at the time the research began because chemical species to separate have very similar properties. Therefore a new chemistry for partitioning elements had to be developed. One hundred very selective new molecules have been tried for minor actinides (the feasibility of separation of neptunium, iodine and technetium has been established; it is based on adjustments of the reprocessing process). Three molecules (two molecules of the family of the diamides and one organic acid) were identified to be used in the partitioning process for americium and curium. Its feasibility was demonstrated in 2001 following a series of tests conducted on actual solutions of dissolved spent fuel, in the CEA's Atalante installation at Marcoule. The process partitioning performances are very satisfactory (~99.9% of the minor actinides recovered). Specific molecules (of the calixarene family) for cesium extraction have also been developed, and the feasibility of its partitioning has also been established, with similar performances.

In 2005, the programme encompasses technological demonstration with representative process equipment, that is to say real testing of these processes involving fifteen kilogrammes of spent fuel in hot cell in Atalante at Marcoule, and economic evaluation of industrial implementation of partitioning.

Specific conditioning processes for the partitioned elements which could wait for transmutation are also being developed (reversible conditioning for interim storage).

Transmutation

Studies on transmutation, which were initiated before the 1991 Law, rapidly led to concluding that transmutation of minor actinides (americium, curium, and neptunium) was feasible in particular in fast neutron spectra. This is linked to the thorough knowledge of transmutation yields resulting from developments in reactor physics and from qualification in this field. However, the transmutation of long-lived fission products (such as iodine 129, cesium 135 and technetium 99) appears less attractive (marginal reduction of radiotoxicity compared to that achieved by transmutation of minor actinides) and not very promising (low transmutation yields).

Results obtained show that the feasibility of transmutation is demonstrated, in pressurised-water reactors (recycling and transmutation of plutonium), in advanced systems of nuclear-energy production (fourth-generation fast-spectrum reactors, with recycling and transmutation of all heavy nuclides, uranium, plutonium, the minor actinides) and in dedicated subcritical incinerator reactors. The best conditions for transmutation are obtained in fast neutron spectra.

Work on transmutation is now focusing on technical elements necessary for the demonstration of its technological feasibility.

Conceptual studies of subcritical systems dedicated to transmutation (ADS Accelerator Driven Systems consisting of a nuclear reactor operating subcritically and fed by an external source of neutrons produced by spallation reactions generated by a proton accelerator) are being conducted in collaboration with CNRS and European laboratories, among others, as part of the 5th, followed by the 6th European Programme for Research and Development initiatives. The results of the international programme TRADE (TRIGA Accelerator Driven Experiment) expected in 2008 will confirm the technological feasibility of the coupling of an accelerator and a subcritical reactor operating at power.

For transmutation studies in reactors able to produce electricity, in 1998, after the decision to stop the Super-Phénix breeder reactor was made, the experimental transmutation demonstration programme conducted in this reactor was redeployed to the Phénix reactor, which was bring it back up to power level on June 15, 2003.

Following results already obtained with Super-Phénix and Phénix (definition of families of new types of fuel suited for transmutation, development of americium targets in concentrated form), irradiations for transmutation studies will continue in Phénix in a much sustained way until 2008. The programme is intended to complete the qualification of the neutron data, test optimized irradiation matrices, study the behaviour under irradiation of specific fuel based on americium or other long-lived elements, and test advanced fuel concepts for the fourth-generation reactors. The first experiment MATINA 1 for testing of -material matrices get out of Phénix on February 2004.

Studies on fuel and transmutation targets also benefit from European and international cooperation. For example, in the HFR reactor at the European Joint Research Center in Petten, the full amount of americium contained in a target consisting of a magnesium oxide and aluminum was transmuted out.

Studies of scenarios continue in order to demonstrate the transmutation of actinides, in power reactor concepts of the fourth generation.

The fourth generation fast neutron systems have unique assets to recycle globally the Plutonium and the Minor Actinides, beyond the first recycles of Plutonium achievable in 2nd and 3rd generation PWRs.

Several scenarios of global management of the Plutonium and Minor Actinides produced in the PWRs have been analysed, depending on the date of deployment of the 4th generation systems.

These scenarios permit to cover reasonably the range of possibilities over the 21st century, while proposing a scenario consistent with the French generating fleet renewal strategy, and considering various alternatives corresponding to different options for the interim storage or the recycling of the Plutonium and Minor Actinides. The transition from today's fuel cycle in France to the deployment of Generation IV fast nuclear systems together with the continuing use of Generation III reactors lead to a mixed fleet of thermal and fast spectrum reactors to be optimised for managing globally the Actinides of both nuclear systems, and making best use of the available natural resources.

Conclusion

Advanced waste management strategies include the transmutation of selected nuclides, cost-effective decay-heat management, flexible interim storage, and customized waste forms for specific geologic repository environments. These strategies hold the promise to reduce the long-lived radiotoxicity of waste destined for geological repositories by at least an order of magnitude. This is accomplished by recovering most of the heavy long-lived radioactive elements. These reductions and the ability to optimally condition the residual wastes and manage their heat loads permit far more efficient use of limited repository capacity and enhances the overall safety of the final disposal of radioactive wastes.

Important results are now available, concerning the possibility of significantly reducing the quantity and the radiotoxicity of long-lived waste, concerning the feasibility of geological disposal and also relating to the modes of waste conditioning and long-term interim storage facilities. Complementary R&D work to be conducted in the coming years will enrich and consolidate these results and, in particular, will detail factors related to the industrial feasibility (costs, tentative schedule for implementation, etc.). On this basis, it can be assumed that sufficient technical and economic data will be available for decisions to be made in 2006 on the possible modes of long-lived waste management.

In this perspective, it is important to underline that:

- Waste which has been produced in the past, nowadays has to be disposed (Andra conducts the French research on this aspect).
- Plutonium control, recycling and management are the first priority in any strategy to minimize waste radiotoxicity.
- Processing and recycling are already significantly reducing the radiotoxicity of the ultimate
 radioactive waste. Furthermore, the further partitioning of minor actinides should reduce to a
 few hundred years the time necessary for the radiotoxicity level of vitrified waste to reach
 that of the natural uranium ore used to originally generate the waste.
- Key design criteria (closed cycle, recycling and integral consumption of actinides, fast spectrum) for the fourth generation of nuclear energy systems are set so that they can transmute their own long-lived waste and contribute to the transmutation of long-lived radionuclides (after their partitioning) produced in existing reactors.
- Despite this prospect of a very large reduction in the quantity and radiotoxicity of waste produced in nuclear power reactors, it will not be possible to get rid of ultimate waste which will have to be disposed.
- Last, the possibility of storing radioactive materials in a safe and robust fashion throughout the long-term is now confirmed. This provides a useful flexible tool for the implementation of back-end fuel cycle management strategies.
- These scientific achievements must be accompanied by a democratic and political debate under the care of government.