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# Energy: towards nuclear breeder installations before the end of this century?

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**Abstract**

To play an essential role in the energy crisis, the civilian nuclear fission industry urgently requires a large and safe underground deposit for irradiated materials. Breeder reactors should be required to extract more energy, not to eventually reduce the radioactivity of dangerous materials. These two aims still require much work, from the fundamental issue of the mechanics of materials, to large industrial parks for breeder development on an international basis. ***To cite this article: R. Dautray, J. Friedel, C. R. Mecanique 335 (2007).***


**Résumé**

**Énergie : vers des installations de surgénération nucléaires avant la fin de ce siècle ?** Pour jouer un rôle essentiel dans la crise de l'énergie, la fission nucléaire civile demande le développement urgent d'un aval du cycle important et sûr. On ne peut attendre la réduction éventuelle de la radioactivité des matières dangereuses, par des surgénérateurs, nécessaires en priorité pour extraire plus d'énergie. Ces deux chantiers demandent de nombreux travaux allant du fondamental sur la mécanique des matériaux à la réalisation pour la surgénération de grands parcs industriels développés de façon internationale et sûre. ***Pour citer cet article : R. Dautray, J. Friedel, C. R. Mecanique 335 (2007).***


*Keywords:* Thermonuclear reactor; Breeder reactor; Energy policy

*Mots-clés :* Réacteur thermonucléaire ; Surgénérateur ; Politique de l'énergie

## 1. Introduction

The foreseeable energy requirements at world level during the 21st century will be hard to meet without resorting to civilian nuclear energy or to coal, or, more probably, to both. France has been ahead in the first domain; it has

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recently confirmed its intention to continue with fission by replacing, in the next two decades, by EPR (Evolutionary Power Reactor[1]) those reactors presently in use and near the end of their lifetimes.

However, this major effort is not the only one to be produced during the 21st century, whatever the civilian nuclear scenario chosen:

- The storage of radioactive materials produced by nuclear activity, even before the EPR are launched, requires a fast and complete development of the back end of the fuel cycle, including geological disposal in a final repository that would be large, permanent and safe. It is an imperative priority in the next two decades; it cannot wait for the reduction in activity by fission or the neutron capture of some radioactive elements to become eventually operative.
- For the EPR, as for the present reactors, the possibilities of civilian nuclear energy using the fission of U 235 (a minor fraction of natural uranium) remain limited and can only replace petroleum and natural gas, their derivatives, and synthetic fuels for less than a century. It then becomes necessary to consider fast neutron reactors as multipliers of energy output, using U 238 or thorium. This way of breeding has been explored in France through the RAPSODIE, PHENIX and SUPERPHENIX reactors; it can multiply by several orders of magnitude the energy that fission can supply. It seems to present no insuperable difficulties, as one was able to show experimentally in the last five decades in the US, in Great Britain, in USSR then in Russia, in Japan and in France. *Only this way can justify starting a fission programme again on a world-wide scale, with a life-time of the order of a thousand years and industrial uncertainties better circumscribed than in the case of nuclear fusion.* This aim of producing energy by way of fast BREEDER nuclear reactors[2] should be a priority, compared with the eventual use of such reactors to reduce the radioactivity of REP (≡ PWR – Pressurised Water Reactor) and EPR irradiated fuels. It requires, on the other hand, the installation in the same place, of a series of operations, in a genuine nuclear park including breeder reactor, plutonium separation[3] from irradiated fuels and from the reactor blanket (and also plutonium separation from uranium), for instance, using a high temperature so-called 'pyrochemical' process to eliminate separable harmful elements, and for making new fuels and blanket elements.

*These remarks are compatible with the potential long term interest of controlled thermonuclear fusion, the only energy source presently known to be able to last more than a thousand years, but which still looks far from any industrial development.*

These various aspects are now treated in subsections 1 to 5 of Section 2.

## 2. Objectives and scenarios

### *2.1. Scientific and technical research; pilot units and engineering for the back end of the fuel cycle*

Develop a system for the back end of the fuel cycle involving:

- An appropriate geological *disposal* of high-level of radioactivity waste in a deep underground *repository*,[4] made inaccessible as fast as possible, developed by units for all radioactive materials not in use at the time, including those existing today as well as those produced until the time of the opening for operation of the storage facility, as soon as these materials can be prepared as packages in suitable canisters.
- Making glassy and ceramic matrices into which the radioactive materials can be incorporated or/and embedded [1].
- Making packages, after regrouping and preparing these radioactive materials. The packages to be put in canisters. The number of categories of packages was calculated to be 12 in 2001 (see document 15, p. 212 of [2]). It is now 16. The annual production and inventory was given in 2001 in document 16, pp. 213–215 of [3].

[1] This new appellation replaces European Pressurized Reactor.

[2] We will use the word Breeder in the following pages.

[3] These operations for the separations of diverse physical and chemical elements are the so-called 'reprocessing' or also 'partitioning' operations.

[4] In the next pages of this Note, we will only use the word '*disposal*' for the action of burying waste and the word '*repository*' for the *site*.

- The realization of canisters with resistance properties which will be one of our main degrees of freedom[5] to protect present and future populations, including, a priority today, the present and future workers involved.
- The realization of locking gears, well adapted to these canisters.[6]
- The realization of the machines to handle the packages at the bottom of the repository.
- The realization of the machines to carry the packages from the ground surface to the bottom of the repository, and eventually to take away defective packages, from the repository.
- The realization of a temporary surface storage area for these damaged packages.
- The realization of all the control and command devices of the system (including those for safety control and command).
- The realization of all the routine mining devices used by the personnel, for as long as the working and surveying of the system must be maintained.

One then can see that our only degree of freedom depends (with dilution) on science and techniques, mass production and the control of materials.[7]

Place into the storage system:

- The B waste (B = Intermediate Level Waste) as soon as this is allowed by the construction work.
- The packages containing the nuclear 'glasses' as soon as they are cool enough, with a delay to be fixed presently (between 3 and 4 decades after their casting into the specific glass), together with a suitable canister.
- The irradiated MOX, placed in suitable canisters, after cooling (6 to 10 decades, depending on its dilution).
  The other highly radioactive materials.

After having built the *factories to mass produce canisters*, these should be filled, closed and tested.

Realize extra *storage places*, as *passive and protected* as possible, to be used as reserves between all the movements of radioactive materials described above.

Requirements and constraints of the system just described:

(i) Preliminaries:

- *Protection from radiation of the workers* and of all other people concerned. In particular, require proof, that any Frenchman can check, whether *iodine 129* is dangerous over a more or less long period. Many scientific publications deny this. The bibliography of [6] presents the measurable facts known today. More scientific elements are needed to progress in this field, which is not yet fully mature.
- Ensure that the canisters for all radioactive materials actually tightly preserve by themselves the materials and the glass casting they contain. This verification should be obtained for a length of time to be fixed by a criterion of radioprotection, independently of other barriers such as the geological one.

[5] Seen as the main degree of freedom in the storage conception (in Sweden and Finland, for instance), the canister is far from being a *simple* degree of freedom, when one goes into the details of the phenomena involved. In fact, the waste package tends to be viewed as the main focal point for insuring overall '*disposal and repository performance*'. This section summarizes the uncertainties inherent in *waste package design and fabrication* as well as in *the nature of the environment* it will be exposed to in the repository. It will also describe how the *environment will influence the long term degradation of the waste packages*. The aim is to demonstrate that the *waste package is one of a sequence of important barriers*, and that its performance is not merely a means to compensate for geological deficiencies (see p. 287 of [4]). Let us stress that, in the American texts, there is the question of the geological confinement of the Yucca Mountain. It happens that the *performance of the geological confinement of the argillite massif, as studied in the underground laboratory of Bure, in France, within our present knowledge, is much superior to the performance of geological confinement of the volcanic tuff of the Yucca Mountain and of the granitic Swedish massif studied in the underground laboratory of Aspö*.

[6] The soldering and welding techniques developed to produce joins for the canisters without thermal stresses require heating up to above 1150 °C. They cannot be used to close the lids, as this would act on the radioactive waste contained inside – "the high temperatures necessary for such annealing would seriously threaten the integrity of the waste form inside the package. *Consequently, the final lid closure welds must be done locally, to avoid thermal degradation, and remotely, to avoid exposure of personnel to radiation fields*" (see p. 290 of [4]). *Friction stir welding* studied in Sweden is the most suitable at present. Anyway, the join of the lid is the weak point in the canister, see [5] and p. 290 of [4].

[7] One must add radiochemistry [7], which could be developed similarly for the chemical separation (or 'partitioning' or 'reprocessing') of required elements in these materials, their use in the making of all sorts of geometries (rods, pellets of different fuels, control rods, etc.) their control, and with a very low loss – less than 0.01%, for example – of useful, or on the contrary, dangerous materials, etc.

- *Protection of radioactive materials* from various types of hostile actions: *proliferation, dissemination*, blackmail, projection of radioactive gases, etc.
- Minimize the *handling* and *transport* between the sites of the pools containing spent radioactive fuels, the factories for mechanical or chemical repairs, those producing the nuclear fuels and the installation described above.
- See to it that all these requirements and protection have a *passive* character and do not require any action from the personnel, or energy input, or artificial adding of fluids, etc.
- One must be aware that it will not be possible to prove, using the scientific methods known today, that the *natural deep geological site* is tight enough during the longest half live of radioisotopes corresponding to the least radioactive ones.

What is then to be done? A first way is to create independent barriers, at least one of them being a total degree of freedom that could be regulated by human intervention. This is the case of a glass casting. It could also be the case of an *appropriate canister*, in which to place the glass casting.

It is, however, impossible to conceive these barriers as totally independent, but they could at least act in concert. The specifications of, and conditions for making the canisters will always depend on the knowledge of the natural surroundings where one places the artificial parts, including the canisters. One will have to work by iteration: when the site is qualified, a first definition of the canisters follows a global prescription. But when years, nay decades, pass, knowledge develops, and prescriptions about the canisters will then develop.

This has happened in Sweden (as shown by the different SR-Can (Safety Reports Canister) which followed each other (see the successive versions of the KBS-1 to KBS-3 disposal concepts).

A complementary trend could be to search *outside the producing nation* for some sites that would be shown to be uninhabitable. This will indeed be necessary for the *European Union*. In tens of thousands of years, the notion of the 2006 European borders will have lost any meaning.

All this constitutes, for France, a considerable endeavour, which, in the last fifteen years, has not been considered; there have not even been published scientific articles, in professional international scientific reviews about *plutonium isotopes' and their descendants' evolution*, or discussion of the *canisters for all highly radioactive waste*. The general and substantial effort is today partly in the hands of a national organisation, while another organisation for nuclear studies covers, with excellent expertise, what concerns the 'matrices' as hosts for certain radioactive materials [8], a number of canisters for moderately radioactive materials, non-intrusive measurements, and fuel and target irradiation.[8] In France, the Haut Commissaire à l'énergie atomique is the Scientific Adviser to the General Administrator of the CEA, according to the law that defines his function.

In fact, the work of the CEA in the field of civilian nuclear energy is concentrated on the ITER program and on the realization of a GEN IV[9] for a fast breeder reactor.

Furthermore, the 'Programming Law n° 2006-739 of 28 June 2006, concerning the lasting treatment of the radioactive materials and wastes' mentions that this type of nuclear reactor must be able to transmute[10] what it defines as radioactive waste,[11] and that waste concerning plutonium and its descendants,[12] *is only mentioned under a very*

[8] Laboratories of the CNRS and Universities also produce an important contribution.

[9] GEN IV: The initiative is called *GENeration for nuclear energy system initiative*, i.e. 'GENeration IV of studies for fuel cycles and nuclear systems'. This is an international enterprise initiated by the Americans, for which the CEA (Commissariat à l'énergie atomique) is responsible for France.

[10] Article 3: Research and studies concerning waste are developed along the two following axes:

(i) Separation and transmutation of long life radioactive elements: The corresponding studies and research are conducted in relation with those leading in the new generation of nuclear reactors mentioned in article 5 of the law n° 2005-781 of the 13 July 2005 which determines energy policy as well as the nuclear reactors driven by accelerators and dedicated to the transmutation of waste, so as to be able in 2012 to evaluate the industrial outlook of these electronuclear channels and to set in operation a prototype of installation before the 31 December 2020;

(ii) Deep level reversible disposal: The request for authorization could be instructed by 2015 and, if this is obtained, the centre be put into operation by 2025.

[11] Article 5 of the law of the 28 June 2006: "The radioactive waste is radioactive substances for which no ulterior use is planned or considered. The radioactive waste is that which can no longer be treated in the technical and economical conditions of the time, in particular by extracting their usable part or by reducing their pollutant or dangerous properties."

[12] We name in the present text 'descendants' the products of disintegration ($\alpha, \beta, \gamma$, emission of neutrons, spontaneous fission, etc.) and of the different capture of neutrons $(n, \gamma)$, $(n, \alpha)$, $(n, 2n)$, etc.

*general text: this depends on the technical way these materials are used, which can change in the next decades, or half century or longer*.

It is the Agence nationale de l'état chargée de la gestion des déchets radioactifs (ANDRA) which has as its mission temporary storage, and then permanent storage, and thus final disposal of waste in a underground geological repository.

It is the AREVA company which is in charge of making fuels, reprocessing the irradiated ones, building the electronuclear power reactors REP and EPR and then, with the electricity utility Électricité de France (EDF) and ANDRA, ensuring the future of plutonium isotopes and their descendants.

This rather complex arrangement shows clearly that there should be a single and overall picture of scientific policy for the complete fuel cycle, for mechanical and chemical separation, for temporary storage, for transport, for the fabrication of all the components of the French civilian nuclear energy industry, if only for plutonium isotopes and their descendants and the making of nuclear fuel with some of their radioactive materials. Sooner or later, it will be necessary to secure *this global scientific prospect for the French nuclear effort at the highest level of the French government*.

To meet this need, the Japanese, and as in the past the USA, have an Atomic Energy Commission of four to five members. The USA has a Department of Energy (DOE), attached to the President of the country.

Sweden has the SKB (Svensk KärnBränslehantering AB) entrusted, by the firms producing nuclear electricity, of all the works, all research, conception, transport, interim storage, continual and constant social dialogue, unit pilots, industrial execution and operation, closing down, tracing of all radioactive waste of any level of activity, including plutonium isotopes and their descendants, used UOX and MOX, etc.

(ii) Experimental references:

Time scales required as a reference in order to determine the different stages (all that follows relies on good engineering of the preliminary studies and their realization, but refer also [International Panel on Climate Change 2007]).

Presently measured quantities:

- Time for cooling down nuclear glasses containing fission products (and, at least until now, so called 'minor actinides', such as americium 241, neptunium 237, curium 242 and 244, etc.) so as to be able to bury them in a dedicated repository: about three or four decades.
- Idem, but for the packages eventually containing 'unreprocessed' UOX fuel elements: three to four decades.
- Idem, but for irradiated and 'unreprocessed' MOX: six to ten decades, depending on the dilution of the fuels rods and the fissile oxides pellets they contained.
- Time to construct factories mass producing canisters and their closing mechanism: between one and two decades.
- Time to qualify an underground site for ultimate disposal in a repository: about a decade.
- Time to construct the site completely, with its means of transport, handling, supply of fluids, ventilation, installations for control and evacuation for the security of its personnel, its buffer surface storage area in the case of retrieval of radioactive packages damaged underground, etc.: about one decade of engineering work.

Duration of events in historical times and archaeological analogies:

- Knowledge of the evolution of an engineering work: some centuries, up to some thousands of years; of a mine: one to a few centuries; of so called archaeological objects: up to 5 thousand years.

Duration of geological phenomena and geological analogies:

- Numerous data of the last 8 glaciations, 840 000 years.
- Geological times since the beginning of the Cambrian period (543 million years), and more precisely of:
  - the tertiary[13] period (65 million years);
  - next glaciation: in about 50 000 years;
  - Oklo phenomena: about 2.1 billion years Before Present.

[13] Limit of Cretaceous period and the Palaeogene and Neogene periods.

Knowledge about the evolution of materials which will not be in equilibrium during the time of radioactivity: the extrapolation length of experiments must be multiplied at least by a factor 1000.[14]

Knowledge concerning the evolution of interaction phenomena between mechanics, thermics, flow of fluids, liquids and gases (and long time creep), the source of heating, of radioactivity, transport phenomena and diffusion under various gradients – all phenomena that will be in neither equilibrium nor in steady state or independent regime, during the time of notable radioactivity: observations can last at most for a century, while the site is filled; a multiplication of about 100 will then be necessary to identify the main mechanisms of these processes, in the various space scales.

We shall now consider some evaluations of the complication and extent of the work to be done. Let us first note three points that will not be developed in the following list:

(a) The amount of waste to be put in packages and not yet in that form.
(b) The required volume of underground repository depends on the future policy of breeding and on the problems of proliferation that one will observe in the future. It seems wise to store the essential part of the UOX irradiated in the REP, and afterwards in the EPR, which would not be intended for rapid reprocessing, and all the MOX irradiated in the REP and in the EPR, without waiting for an eventual industrialization of a 'reduction'[15] technique of the radioactive elements.
(c) It will, anyway, be necessary to prepare the reserves, stocks and supply of Pu 239 for the first loading of the Fast Neutron Reactors (FNR) conceived for *breeding*. The isotopes of plutonium produced in the UOX irradiated in the REP and EPR will be suitable.

However, the isotopes of the MOX irradiated in the REP, and even more in the EPR (with a higher *neutron fluence* [neutrons/cm$^2$ during the whole irradiation] or the same with other units, *burn up* [GigaWatts day/ton of initial heavy metal]), will contain amounts of Pu 238 and Pu 241 (thus producing americium 241), too high to be merely handled.[16]

(iii) The tasks to be carried out in France:

A few *orders of magnitude can be mentioned concerning packages to be buried permanently, for ever, in a repository. 16 sorts of packages of waste*, according to a classification of the state agency responsible for dealing with the radioactive waste[17]:

– type B: B1 to B8;
– type C: C0 to C4.

[14] The casing material for canisters of highly radioactive elements in the US (at Yucca Mountain, if the site is confirmed and qualified). The DOE has selected a material called Alloy 22, a chromium, nickel, molybdenum alloy to form the outside layer of the waste canister. Data from DOE laboratory tests of 6 months to 5 years length have been extrapolated to 600 000 years, and suggest that the waste packages will begin to fail at 50 000 years (changed recently to 100 000 years); see DOE [9] and Chapter 18, p. 30 of [10] and in Chapter 18, p. 301–316 in [4].

[15] '*Reduction', but of what*? Volume? Radioactivity? Inhalation radiotoxicity? Ingestion radiotoxicity? Migrating aptitude? Chemical forms and evolution during migration, in what surroundings? Aptitude to form colloids? solutes? Aptitude to be adsorbed? On what? Aptitude to enter various chemical reactions? *In what conditions*? *Reducing*? *Oxidizing*? Relation to confinement through chemical reactions with bacterial environment? Organic? Radiotoxicity by ingestion? By inhalation? By whom? Who will ingest or inhale 500 meters underground? In what circumstances? *Chemical toxicity of all this material*? In what conditions? By what channels into the human body? Through what intermediary? etc. All this is to show that the word *reduction* cannot be used alone: one could say the same about the word '*transmutation*'; it is too ill-defined, as it concerns all phenomena of disintegration and nuclear reactions.

[16] As for their descendants, their fission threshold is too high (except for neptunium 237) and their half life too short, so their radioactivity too high to be made and handled and produce fissions, without producing, by neutron capture, elements extending to the region of high spontaneous fission rates, thus copious neutron emissions. The good news is that all these isotopes cannot be used for explosive nuclear weapons. What will one do with all this material? The other MOX irradiators (in Belgium, Germany, USA with a very different isotope composition of plutonium of military origin because composed of Pu 239 at more 90–94%, Sweden, Finland in the future, etc.) do not have this problem, as they intend to bury all the irradiated MOX fuel elements in their final disposal into the repository. It would be useful for the solution for burying suggested by [2,3], and that developed in France, to be published by the competent organisms concerned, in professional scientific journals, with a comparison with those developed in other countries.

[17] Without counting the waste eventually extracted from irradiated UOX, either to make provision of plutonium isotopes and perhaps some of their descendants, or the same but from irradiated MOX.

- Number of waste packages of B (ANDRA denomination) to be buried: about 160 000 in 2010 (about 2.4% of the total civilian radioactivity in France (see [3] and ANDRA scenario S2[18] of document [11]).
- Number of *vitrified* C packages (ANDRA denomination) to be buried underground: about 14 000 in 2010 (about 72% of the total civilian radioactivity in France [1,3], increasing by 500 packages per year).
- Number of *fuel assemblies REP* irradiated until 2010, following Scenario S2 CU1: 54 000.
- Eventual number of CU2 packages of *irradiated MOX* in 2010: 4000 packages, increasing by about 150 per year (about 25% of the total radioactivity [3]).
- Number of CU3 packages (other than UOX or MOX) of fuel assemblies in 2010 (Scenario S2[19]): 5800 packages.

Other methods of estimation, following 4 categories of packages, made by ANDRA on the 31 December 2004:

- Type of package: CSD-V,[20] total number stored: 7866 packages; to be produced in supplement with the waste not yet conditioned: 1700 packages – total 9566 packages.
- Idem for CSD-C: 2079 packages and 15 700 packages; total 17 779 packages.
- Idem bituminous barrels: 10 328 packages, 40 000 packages; total 50 328 packages.
- Idem cemented waste barrels; 5170 packages, 1800 packages; total 6970 packages.

Used fuel elements – in reactors: 4155 tons, including:

- UOX: 4569 tons;
- URE[21]: 74 tons;
- MOX: 312 tons.

In the water pools of electronuclear plants and reprocessing factories, etc.: 11 600 tons, including:

- UOX: 10 700 tons;
- URE: 200 tons;
- MOX: 700 tons;
- SUPERPHENIX: 115 tons.

One should add to all this about 40 000 tons of high activity nuclear waste to be produced and placed in packages until the (eventual) burying of the fuels elements or their remains in the repository.

The total radioactivity present in France in 2006: in radioactive waste and plutonium isotopes and their descendants, including those intended for making fuels elements for existing REP or future EPR, thus certainly in the coming half century or more: about 5 to $6 \times 10^9$ Ci,[22] thus 5 to 6 billion Ci.

[18] Scenario S2 "assumes that part of the UOX fuel is reprocessed until 2010, 8000 tons of heavy metal from UOX1 and 8000 tons of heavy metal from UOX2, then this stops. This is why there are no C3 and C4 wastes, which concerns only the waste of the future". This use of Scenario S2 is not made by preference but because it estimates correctly the approximate number of packages to put in the repository, assuming that waste which is not yet conditioned is put into packages.

[19] In Sweden, all irradiated fuels make up a single category of packages, in Finland too. Consequently, there is only *one type of canister for the high level radioactivity packages*. Thus, one can concentrate efforts on obtaining high quality confinement of radioactivity and also on conceiving the final repository of these canisters.

[20] CSD-V Conteneur de Stockage de Déchets Vitrifiés (storage container for vitrified waste). CSD-C: Conteneur de Stockage de Déchets Compactés (storage container for compacted waste).

[21] URE: Uranium RE-enriched, refers to uranium extracted from irradiated UOX (with roughly 1% of U 235 [2,3]) reprocessed (so-called URT: Uranium de ReTraitement), and 're-enriched' (between 3.5 and 4% [2,3]), so as to be used again in the fuel cycle of REP.

[22] To understand this number, note:

(i) the radioactive waste to be put in the Yucca Mountain deposit will be of the order of 10 billion Ci;

(ii) the radioactivity of all American high activity nuclear waste from Defence (i.e. military) installations is 460 million Ci, reduced to 230 millions Ci at burying time (Macfarlane et al., Table 21.1, p. 354 of [4]). This amount will be transformed into nuclear glasses, following the method developed and put onto the market by France (the addition of borosilicate with $SiO_2$ and $B_2O_3$), giving about 50 000 packages with 60 cm diameter and 3 meters height at Savannah River (a height of 4.5 meters at Hanford). The fabrication will last for about 2 to 3 decades.

The *time required* for the scientific and technical work to present an exhaustive proposal of *Qualification* of a site for final disposal into an underground geological repository and all its internal engineering[23]: at least, one decade.

Duration of verification and control works for examination by the administrations concerned: one half to one decade.

The *duration of filling the underground parts*[24] *of the repository*: about 3 to 5 decades. This means that one can take out damaged packages during this time.

(iv) Lapse of time during which risks for human should be contemplated:

The various institutions concerned in countries which have studied repository sites fix this time at around 10 000 years for a quantitative evaluation and up to 100 000 years for an estimation of trends.[25]

In Sweden,[26] it is the canister and its artificial surroundings which provide the main part of confinement for packages of radioactive waste.

In the USA, it is the same criterion of the canister and its artificial surrounding, in the present practice as planned for the Yucca Mountain geological final repository.[27]

In France, the legal texts existing today give this role of confinement, for canisters of 'radioactive waste', to the natural geological barriers.[28]

These uncertainties and thus the working methods which must be related to theory, make up the fundamental message of these few pages of Section 2.1. These difficulties of fabrication have stimulated research in alternative directions, postponing the realization of the final underground geological repository of undesirable radioactive materials. These alternatives essentially push back the date when one will have to consider squarely the problem. However, every day of delay adds new radioactive materials and the degradation of temporary devices adds to the risks that already exist for public health.[29]

Half would be buried at Yucca Mountain, being marginal compared with the irradiated fuels from civilian electronuclear power reactors. The proportion of radioactivity of military origin which would be placed at Yucca Mountain would be of the order of 10% [4]. A rough estimate by proportion would give for an eventual French disposal into the repository of military fuel a proportion smaller by several orders of magnitude.

[23] 'All' means: the canisters, their sealing, the artificial filling (the so-called 'engineered barriers').

[24] These parts of the repository may be independent.

[25] The uncertainty increases with the lapse of time of this prevision; this leads to standards of health radioprotection that are much stricter in the far away future, after 10 000 years, than before that time. These are the standards imposed presently for ultimate disposal in the repository.

[26] In Sweden, Finland, and the USA, as in more than 2/3 of the world production of irradiated fuels which are not reprocessed, the radioactivity of the in situ radioactive waste has such a composition as to be dominated during 300 years by the radioactivity of cesium 137 (the direct fission product is iodine 137 – 24 seconds → 92% xenon 137 – 3.9 minutes → cesium 137 – 29 years 92% → barium 137 – 2.57 minutes → barium 137 – stable) and strontium 90 (the direct fission product is krypton 90 – 33 seconds → rubidium 90 – 2.7 minutes → strontium 90 – 28 years → yttrium 90 – 64 h → zirconium 90 – stable). These will have decreased by a factor 500 after 300 years. The radioactivity is then dominated until a little more than a thousand years by the radioactivity of americium 241, descendant of plutonium 241. After about 1000 years, the radioactivity is dominated by plutonium 239 (90% of the radioactivity), the remaining radioactivity being due to plutonium 240, with 100 times less for technetium 99 and 100 000 times less for iodine 129, this until several tens of thousands of years; after 100 000 years, the radioactivity of neptunium 237, a descendant of americium 241, dominates (documents 53 to 63, pp. 240–250 of [3]). Thus, not to reprocess fuel elements is to create *artificial mines of plutonium 239* with a mean composition of 1%!

[27] However, this practice of the American Administration is strongly contested by some experts.

[28] We must add that all the High French Authorities concerned agree with the notion of *reversible* underground storage. Reversibility is a matter of course during the period of operation in the disposal of packages into the repository (filling up, checking the packages and the artificial structures installed) and during the time of control and check of the reachable properties of the geological site. However, beyond that period, is reversibility realistic in argillite? In any case, it has a cost, not only for the budget, but above all in the *danger for the workforce, the employees, the personnel concerned* and the neighbouring population. Can one estimate this? To see whether it is, while it lasts, very onerous and more or less dangerous for the workers, depending on the duration, nature and level of reversibility one aims at?

[29] Leaving this Section 2.1, the reader can observe that the problems of the radioactive waste related to the *dismantling* of categories of nuclear installations have not been considered. One example will show their relative dimension compared with the problems treated in this section: the dismantling of the installations producing the central parts (so called 'pits') of the triggers of thermonuclear bombs made with plutonium, which contain many other elements difficult to handle, such as beryllium (high *chemical toxicity by inhalation or/and ingestion*), etc., was made in the USA at Rocky Flats Nuclear Weapons Plant. With the help of in situ scientific experimental studies, the methods foreseen have been radically altered and the dismantling time reduced by one year [19]. *This site is now given back to public life*. However the radioactive waste of this dismantled material must, of course, find an ultimate outlet, and this bring us back to Section 2.1. A second example is the irradiated structures of SUPERPHENIX that have been dismantled without any difficulty concerning the radioactivity of its components.

Balance sheet of Section 2.1:
The main problem of nuclear waste and of plutonium isotopes and their descendants is that the scientific, chemical, industrial, administrative communities and the leaders of most of the countries concerned have seriously underestimated the difficulties of the problem posed by the material aspects of burying nuclear waste underground in a geological site. More precisely, to characterize what are the properties of the site, including both the natural and artificial parts, to predict its behaviour during at least ten thousand years and even during hundreds of thousands of years, would require an effort much larger than what it is presently made by the organisations concerned in France. Moreover, this work does not depend only on deterministic or even probabilistic predictions, but more simply on a fair part of uncertainty.[30] At most, one can try and come to terms with these uncertainties by reducing them with the help of what we are best at mastering, by redundancies and by managing in such ways that the positive aspects work together. Of all these uncertainties, those relatives to *hydrology* (with all related phenomena) and its various consequences are dominant.

### 2.2. *On the electronuclear power plants to be launched to replace the present 'park' during the next two decades*

The aims are:

(1) To progressively replace the existing electronuclear reactors REP by *EPR* (*Evolutionary Power Reactor*).
(2) To adapt all the installations for reprocessing, making fuels, interim storage, buffer storage, final repository, etc., to the new conditions of irradiation with a higher neutron 'fluence' [Neutrons/cm$^2$ during the total irradiation of the fuel element or of the target or the fertile element of the blanket] and thus an increased proportion of isotopes (non-fissile, in a classical spectrum of fast reactors) of plutonium, of americium 241, of neptunium 237, of isotopes 242 and 244 of curium (document 104, p. 280 of [3]) and eventually beyond. This holds a fortiori in a more stringent way for irradiated MOX in these *EPR*.
(3) The resulting scenarios do not create problems, except for the back end of the fuel cycle[31] where the increase of irradiation of fuel elements increases the amount of isotopes 238, 240, 241, 242 of plutonium and their descendants, but making them still more unfit for nuclear explosives. *It is a substantial progress against proliferation*. For the rest, *the progress brought by EPR in safety for the workers and neighbouring populations and from the radioprotection of workers is considerable*. It is likely to be the best project in the world for nuclear power plants which have reached the industrial stage. EPR offers a reference of excellence to the whole world for all the scientific, technical and industrial aspects, for safety, radioprotection and the fight against proliferation.

**Remark.** In developing countries, these nuclear parks are not sufficient. For it is assumed, in the above list, that *there exists a substructure of crafts, factories, services, trained and experienced personnel outside the nuclear parks* and other eventual parks.

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This ultimate and final burying of waste and other radioactive materials without present assignment is, once more, the key to the cleaning up of nuclear installations.

[30] To summarize, it is not possible to validate or verify [12] mathematical models of the evolution of the system, whether partial or more complete, of the filled up repository because they are open, dynamic systems, able, as is also each of their components, to exchange matter and energy of all sorts. Furthermore, even these component systems are too complex to be known completely, and their evolution cannot be described and simulated numerically [13]. This system of a whole repository site (even when it will be completely sealed down) is permanently under many gradients of temperature, chemical composition, diffusion, pressure (including those due to gas emissions and their evolutions) (p. 263 of [14]), of pH, redox conditions (a redox reaction is a reaction in which there is a transfer of electrons from one species to another) tensions, composition of water, type of existing colloids, [15] etc. which induce transport, kinetics, deformation, irradiation, etc. A whole scientific literature exists to choose, depending on the criteria of preference (clear, simple and already explicitly stated, for example in [2]), the possible associated optimal solutions. This is already using the terms of optimal control [16,17]. There does not exist one unique model of evolution of the phenomena, but families of models of evolution, for instance those that put more weight on some knowledge at some period. In one word, as in many engineering works, one can at best have *indicators* [18].

[31] After the exit of irradiation.

*2.3. Carrying out of the research necessary to realize nuclear installations for energy production, using isotope 238 of natural uranium*[32] *(99.2745% of natural uranium), by transforming it into useful plutonium isotopes, especially the isotope 239*

This future eventual industrial installation for energy production would involve the following system:

- The making of a prototype reactor, prototype in the sense of first in a series. And thus, this comes the realization of several intermediary size reactors and material testing reactors.
- The factories producing the various types of fuels starting from radioactive materials, $\alpha$ emitters as in the MELOX factory,[33] or using other types of disintegration, which would present more problems (including powerful emission of fast neutrons by spontaneous fission).
- The behaviour of these fuels in irradiation, including the accumulation of gases (helium, gaseous fission products, etc.)
- The factories for mechanical, physical and chemical separations of the combination of useful plutonium isotopes of the fuel from radioactive materials to be buried.
- The handling devices.
- The devices for carrying new kinds of radioactive materials.
- The preparation of all waste before burying it in the underground geological final repository and *all that comes with it*, inside and outside, as described in Section 2.1, including factories for *making and handling canisters, filling* them and *closing them* with a *tightly leak proof*, adapted and sealed[34] lid, and for the control all these processes and their behaviour.
- The factory to separate uranium isotopes by centrifugation and its destiny in case of new orientations of the country with respect to *proliferation*.
- All this should be, as much as possible, concentrated on *one single site* to avoid handling, changes into transport canisters and the transport of these.
- The *final repository* has so many constraints *that it will have to be separated from these other functions and also to serve several nuclear parks*.

International scientific, technical and industrial research (pilot factories) will have to be carried out, together with developing new tools. To carry out this research, these tools are of first necessity, especially used those to study materials:

- Testing reactors for materials and components of the system.
- A laboratory to examine radioactive materials and components.
- Manufacture of targets out of highly radioactive elements.

Aims and constraints for a system of this kind that would be studied under the direct responsibility of, and perhaps principally in, France:

[32] As well as isotopes 232 of natural thorium (100% of isotope 232) which one could wish to transform by neutron capture into protactinium 233 and then, by $\beta$ disintegration – half life 27 days – into isotope 233 of uranium (half life: $1.59 \times 10^5$ years) which is suitable for breeding by fast neutrons, but also by thermal neutrons. U 233 is also an excellent material for the nuclear weapons. Some mock up of this pure material U 233 exists in India (Purnima 1, Purnima 2, Purnima 3).

[33] The factory MELOX, at Marcoule, fabricates the MOX fuel elements with the isotopes of plutonium (roughly 8.5 tons/year of isotopes of plutonium) separated from reprocessed irradiated UOX and with uranium – natural or depleted – roughly 0.3% of uranium 235 produced by the enrichment factory by diffusion of Tricastin. Every year, to fabricate the 1000 tons of uranium enriched to 3.5–4% of uranium 235, the French producers use roughly 8100 tons of natural uranium. There remains roughly 7080 tons of depleted uranium (roughly 0.3% of U 235). 92 tons/year of this depleted uranium is used for the fabrication of the actual 100 tons/year of MOX fuel elements. The duration of this process of cycles of fabrication of MOX is around 4 years of mining of the rocks containing natural uranium in the mine, 3 years of conversion to natural uranium separated from rocks and its own descendants – radium, radon, polonium, etc., – 2 years of enrichment of uranium, 1 year of fabrication of the metal, 2 years of transport of irradiated UOX to the reprocessing plant, 8 years of reprocessing, 9 to 10 years of fabrication of MOX. So, *the duration of the sum of all the processes included to use one MOX fuel assembly is, roughly, 12 years* (document 17, p. 216; document 27, p. 222, documents 88, 89, 90, 91, pp. 270–273 of [3]).

[34] This sealing is made in Sweden by friction stir welding. Cf. note 6.

- The only aim of this system must be to produce plentiful and economical energy[35] in a directly usable form. This goes through the creation of plutonium 239 through the capture of a neutron by a nucleus of uranium 238[36], and this with any drawbacks and disadvantages made as low, or as manageable, as possible.
- To try and add another objective, for instance the destruction of some radioactive elements such as isotopes of plutonium other than 239, and their descendants accumulated by irradiating MOX, would lead to incompatible constraints, including in time scales.

*Proliferation*: The uranium of the blanket, whether natural or depleted in U 235 (from the enrichment plant waste), leads by capture of neutrons escaping from the core of the reactor, to a plutonium nearly all made of isotope 239, thus well adapted for military uses, as the successive ratios of capture of neutron cross section (so called $(n, \gamma)$) divided by their fission cross section (so called $(n, f)$) for the capture of a neutron are relatively low for fast neutrons escaping the core of the reactor.

*2.3.1. The necessary basic knowledge*

The materials and their treatment: As for previous types of nuclear reactors, the behaviour of the materials used as fuel or structural elements must be fully mastered. This includes the reaction to high temperatures, stresses, irradiation and chemical attacks. It involves an understanding and mastery of creep, swelling, embrittlement and possible anisotropic growth under irradiation, working from a detailed knowledge of the defects at atomic scale to the consequences for the macroscopic behaviour of materials. If an obvious progress has been made since Fermi's first reactor in Chicago, their micro–macro progression still poses essential questions, even the simplest questions of crystal plasticity.

However, the back end of the fuel cycle will also pose new and serious questions. One must then separate mechanically, physically and chemically the isotopes of plutonium and their descendants from targets in uranium 238. The proportion of uranium 238 transformed into plutonium 239 will depend on the *neutron spectrum* (density of number of neutrons of kinetic energy $E$, a function of the kinetic energy $E$ versus its kinetic energy [Millions electron Volt: MeV ], on the *flux* [Number of neutrons/cm$^2$ second] and most important, the neutron *fluence*[37] that this material containing uranium can support. This property of the material under irradiation will then determine the number of times the same uranium will go through the reactor, hence the *necessary number of operations of reprocessing and refabrication* of fuels and of control rods also containing highly radioactive elements. *One is brought back, once more, to specifications, articles and conditions*[38], *which would define the conditions to be endured by the materials to be adapted and, conversely, to the studies of materials which would delimit the ambition one could have for such prescriptions.*

Radiochemistry: A review of the works to be done is in a special report of the French Academy of Sciences [7].

For all this, tools are first necessary: a reactor for material testing with fast neutrons beams of adjustable average kinetic energy; a laboratory for testing and examination, etc., but not a prototype (a prototype of what?). Reactors such as PHENIX are needed in priority.

Starting fuels for breeding of the GEN IV in France: At the start of these prototypes, and after of these parks of nuclear installations, reserves, stocks and a supply of plutonium 239 will be needed. In what proportion will one be able to accept Pu 238, Pu 240, and Pu 241 for making the fuels, as well as for irradiating them in the reactor, with a required stability of the neutron flux in each part of its core and also a combination of isotopes varying at each cycle of irradiation? Are these successive combinations of plutonium isotopes, due to the chemical separation of MOX, acceptable for the spectrum of fast neutrons in the core of the future breeder of the GEN IV series developed by the competent French organization? What quantities will be required? What will we do with their descendants of various thresholds of energy for fission? On the other hand, one has seen above that the blanket of U 238 will give Pu 239 with cross sections for successive captures of neutrons favourable to obtaining Pu of military 'quality'. This plutonium

[35] See [20,21].

[36] U 238 + neutron → uranium 239 → $\beta$ (23.5 minutes) neptunium 239 → $\beta$ (2.35 days) Pu 239.

[37] Number of neutrons/cm$^2$ during the whole irradiation or energy created/duration of the irradiation of the fuel: GigaWatt days divided by the Mass of Initial Heavy Metal contained in the fuel: GWD/ton MLI.

[38] *Depending* on the kind of component using the materials, one can use also the terms 'list of duties prescription', or 'the terms of reference'.

will be well adapted to making fuels for fast neutron reactors. However, will the problems of *proliferation*, in foreign countries, or terrorist's networks be treated?[39]

This contradiction comes from the physics of plutonium production in the blanket of the breeder reactor (by the different rates of successive capture cross sections multiplied by neutrons flux [Neutrons/cm$^2$ second] and half lives of successive disintegrations). It gives a plutonium rich in isotope 239, so excellent for breeders, but also well adapted for the nuclear components of thermonuclear weapons. It emphasizes the necessity of an international control of the fuel cycle[40] for breeders and also accentuates the *crucial need for 'an international data bank' of known nuclear explosive materials* where France can aid in this vitally important progress [22].

### *2.4. Research in controlled thermonuclear fusion by magnetic confinement in a tokomak*

Nuclear fusion is still at the stage of feasibility studies, far from possible industrial exploitation. As a result, fusion cannot reasonably take the place of coal or fission breeders during this century. The experimental study of plasma physics is a matter of course, as the research machine ITER is planned for progressing for in this field.

As a complement to the ITER program, it would be good if French research people took an intensive part in the international research, especially with Japanese and American researchers, on the following topics:

(1) The *materials* [23] for the future thermonuclear machine with magnetic confinement. This is a vital activity for the future of thermonuclear fusion. It requires experimental means (most of them still to be conceived – for instance for microscopic examination – but bound to be beneficial to other activities) of the same order of magnitude as the ITER machine and with important teams of fundamental research.
(2) One of the open questions is the behaviour, both in volume and surface, of materials submitted to very high energy neutrons developed in thermonuclear Tokamaks with a continuous power (the generation of reactors after ITER), as there is so far practically no experiment in this field. Another general question is the possible use of nanotechnology to develop the blankets discussed below or any other material that should be active through its surface.
(3) The *physical 'modelling'* and its *mathematical and numerical simulation* of all significant phenomena needed to conceive and realize such machines, and already for the ITER. French mathematicians could play a fundamental role in conceiving the mathematical models that could be numerically studied on computers.
(4) The *neutron balance* sheet of these machines and, going necessarily together, the concepts of blankets[41] to regenerate the tritium,[42] to extract and recover the energy from these very fast neutrons,[43] of nuclear reactions in the blankets such as:
   – very fast neutron + beryllium 9 → 2 neutrons + 2 helium 4,
   – Li 6 + neutron → tritium + helium 4,
   as well as the energy inside the plasma torus[44] from the $\alpha$ particles produced in fusion and the heating of the plasma they carry along.

[39] This question has already been asked (and a reply given) by the French Government in the past.
[40] Including the back end of the fuel cycle.
[41] These blankets are in a high magnetic field, which induces powerful Magneto Hydro Dynamic (MHD) effects.
[42] One needs to produce a slightly higher number of neutrons in the blanket of the torus than the tritium used in the Torus thermonuclear reactions because the extraction of tritium from the blanket and the purification of this recovered tritium will never be 'perfect', without losses of neutrons (neutron capture losses and neutron leakage, for example) and of tritium produced in the blanket by the nuclear reaction neutron from the thermonuclear fusion of T + D → He 4 + neutron; neutron + lithium 6 → tritium + helium 4 + helium 4, so with a yield of one for the neutron and also for the tritium, when neglecting some other minor fusion and capture nuclear reactions. It is why one introduces beryllium at the frontier between the torus and the blanket in order to use the reaction $(n, 2n)$ on beryllium 9. This cross-section is practically zero for a kinetic energy of the incident neutron under 1.9 MeV, approximately 0.5 barn between 4 and 14 MeV (1 barn = $10^{-24}$ cm$^2$).
[43] 'Virgin' neutrons from the fusion reaction or scattered neutrons on their way.
[44] The extraction of power from the torus is made, together with the ions of so called 'ashes' by a device called a *divertor*, which has to be studied and proved on the scale of a thermonuclear plasma in ITER.

*2.5. The global time scales, evolution of the technical system and content of resources, prices and 'global change' due to human activity*

- The theme of Section 2.1 should be the priority in the coming decade [24–26] and thus start being realized in the decade after. One could aim, from the mere technical and industrial points of view, to qualify a site and all its future contents, as described in 2.1, for 2015.
- Any delay [27] is a supplementary threat to public health. At the close of these sections 2.1 to 2.5, one must underline, accentuate and emphasize the importance in these new problems, concerning our knowledge and safeguards from the effects of radiations by radioactive materials. They are cited in [6].

## 3. Conclusions

- Together with the important launching of EPR reactors (and of considerable importance for safety and radioprotection), the next two decades should in priority finalize the back end of the fuel cycle of the thermal neutron power reactors, the REP, with a final disposal into a underground geological repository (for the radioactive products generated in the past and future nuclear activity of this country). It is an illusion to count on a notable reduction of the fission products by using fast neutrons reactors: to face the long term world requirements, their essential task, necessary before the end of the century, will be to make energy from fission competitive with that of coal or eventually with other types of energy from fusion.
- To fulfil these indispensable and urgent tasks, one must return again to the long term policy which has made possible the success of our country in this field. Such a policy requires a realistic analysis of the stakes, which would enlighten the national representation and the population, as well as the Government, on the possible choices (breeding processes in nuclear installations) and on the urgency. It requires a realization in the long term by a personnel strongly motivated and with a solid scientific and technical competence, with clearly defined tasks, driven by leaders of exceptional merit and sustained by the general scientific and technical activity of this country.
- Nothing valid will be made in France if our country does not, in this field, take its future in hand. However, international contacts and collaborations are also essential, on the methods to be used and the dangers to be avoided, as well as for the management at world level of this energy which, as Aesop's tongue, can be good or evil, according to the way it is operated and used. It is indeed clear that a world extension of fission (with the possibilities of diverse processes of proliferation), requires a fundamental reform of international relations in this field, in which France must take part.

The aim of this Note has been to start a reflection which seems necessary and urgent.

## 4. Supplementary information

Further information can be found on the following sites:

- AREVA: www.cogema.fr – www.areva.com – www.framatome.com,
- Autorité de sûreté nucléaire: www.asn.gouv.fr,
- CEA (including the GEN IV initiative): www.cea.fr,
- CNRS: www.cnrs.fr,
- EDF: www.edf.fr,
- Institut de radioprotection et sûreté nucléaire: www.irsn.fr.

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