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Perspectives on the closed fuel cycle – Implications for high-level waste matrices

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Abstract

Nuclear energy accounts for 80% of electricity production in France, generating approximately 1150 t of spent fuel for an electrical output of 420 TWh. Based on a reprocessing–conditioning–recycling strategy, the orientations taken by Électricité de France (EDF) for the mid-term and the far-future are to keep the fleet performances at the highest level, and to maintain the nuclear option fully open by the replacement of present pressurized water reactor (PWR) by new light water reactor (LWR), such as the evolutionary pressurized reactor (EPR) and future Generation IV designs. Adaptations of waste materials to new requirements will come with these orientations in order to meet long-term energy sustainability. In particular, waste materials and spent fuels are expected to meet increased requirements in comparison with the present situation. So the treatment of higher burn-up UO_2 spent fuel and MOX fuel requires determining the performances of glass and other matrices according to several criteria: chemical 'digestibility' (i.e. capacity of glass to incorporate fission products and minor actinides without loss of quality), resistance to alpha self-irradiation, residual power in view of disposal. Considering the long-term evolution of spent MOX fuel in storage, the helium production, the influence of irradiation damages accumulation and the evolution of the microstructure of the fuel pellet need to be known, as well as for the future fuels. Further, the eventual transmutation of minor actinides in fast neutron reactors (FR) of Generation IV, if its interest in optimising high-level waste management is proven, may also raise new challenges about the materials and fuel design. Some major questions in terms of waste materials and spent fuel are discussed in this paper.
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1. Introduction

Nuclear energy, thanks to its ability to produce a cost-competitive, safe, reliable and independent energy source, without CO_2 emission, is a prominent contributor to energy sustainability for long-term electricity production. World energy prospective

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studies forecast, in most scenarios, a significant increase in energy needs, despite the improvement in energy efficiency and the efforts for an improved mastery of energy demand. Therefore, nuclear energy will continue to play a major and long lasting role for baseload generation in the future, with an extension in lifetime, when possible, of the existing nuclear power plants (NPP) and/or their replacement by advanced evolutionary LWR reactors. Moreover, considering the expected increase of world nuclear installed capacity, Generation IV nuclear systems, breeder reactors associated with a closed uranium–plutonium cycle, capable of using more completely the value of uranium resource and multirecycling plutonium, could emerge technically at a time around 2040.

The economic and social stakes represented by the mastery of the implementation of these development programmes involve crucial scientific and technical challenges concerning the material used in the nuclear power plants, in particular the fuel materials and those used in the containment matrices for highly radioactive and long-lived waste. The perspectives opened up with the future fuel management techniques (e.g. an increase in the burn-up) and the development of the nuclear systems of the future will lead these materials to becoming subject to more severe conditions than those prevailing at the present time. With reference to the present situation, the issues faced by and the needs of the French power utility and its partners for the coming years are examined.

1.1. Current nuclear industry in France

Nuclear energy currently accounts for 80% of electricity production in France thanks to 58 PWR representing an electrical capacity of 63 GW. Designed on standardised PWR series, most of them were commissioned in the eighties. They produced 429 TWh in 2005 with a high-level of reliability and safety.

A major economic challenge for the near-term is to improve nuclear fuel energy efficiency, in order to have longer operating cycles and to reduce the amount of nuclear fuel for the same energy output. The goal is to increase competitiveness both by optimising operating cycle length and flexibility of reloads and by improving nuclear fuel efficiency and so to evolve from a current average of 44 GWd/t up to 56 GWd/t in average in the next decade. This endeavour will procure both an economic benefit and a reduction in spent fuel inventory.

In parallel with the development of nuclear electricity production in France in the seventies, and in order to preserve energy options and possible fast neutron reactors (FR) development, the choice has been to develop a closed fuel cycle based on reprocessing of spent nuclear fuel (SNF), and plutonium and uranium partitioning. Assemblies are sheared and dissolved in order to separate high-level waste using the PUREX process. These elements are confined in glass ingots, and valuable nuclear material – plutonium and uranium – is recovered as a potential energy resource to be recycled. The separated plutonium is recycled into MOX fuel, according to plutonium flux adequacy policy, which ensures that no separated plutonium in excess above industrial needs will accumulate.

Regarding spent fuel and back-end, the nuclear power plants generate each year approximately 1150 t of spent fuel for an output of ~420 TWh. A quantity of about 850 t of UO_2 spent fuel is reprocessed, the choice depending on maintaining global optimisation for plutonium recycling and energy efficiency. It is then about 100 t of MOX fuel which is fabricated and loaded into twenty 900 MW licensed reactors (30% of core), which contributes to 8–10% of nuclear production.

1.2. Current materials of the back-end of fuel cycle

1.2.1. High-level waste (HLW)

High-level waste (containing fission products (FP) and minor actinides (MA): neptunium, americium, curium) produced by reprocessing SNF has been vitrified on an industrial scale in France since 1978. The advantage of this process is that it considerably reduces the waste volume, in comparison to a direct disposal of spent fuel, by concentrating it in a chemically durable matrix. Due to their amorphous structure, glasses appeared to be a category of materials capable of incorporating most of the fission product oxides in their vitreous network. Research performed by the French atomic energy commission (CEA) in the 1950's led to the selection of borosilicate glass as the most suitable containment matrix for waste from SNF. Borosilicate glasses appear as a very good compromise in terms of containment (leach resistance, thermal stability, resistance to irradiation), technological stability, and cost (via the volume reduction factor). Today, borosilicate glasses have become a worldwide standard and have

been chosen for nearly all vitrification processes of HLW solutions [1].

The R7/T7 glass formulation (Table 1) implemented by Areva NC was designed to hold a maximum of 18.5 wt% of radioactive waste oxides (fission products, actinides, noble metals and Zr fines), the total waste loading factor being above 25 wt%. This limit is related to chemical criteria associated to the glass manufacturing temperature. The glass product has a high activity (predominantly ^{137}Cs, ^{90}Sr) and significant amounts of noble metals (3 wt% max.).

Industrial glass samples produced by Areva NC have been characterised [2]. Satisfactory quality of the glass has been demonstrated: glasses were homogeneous with no undissolved feed and their characteristics were in full agreement with the expected values.

Scientific models describing the possibilities of vitrified waste alteration in the very long-term have been developed to predict the behaviour of such waste on these time scales, under different conditions. These models predict that the vitrified waste package is a major contributor in the safety assessment of the disposal [3,4].

The global volume of HLW conditioned and stored reached 1710 m^3 (i.e. 0.13 m^3 of vitrified waste per ton of reprocessed SNF) at the end of 2005. The inventory grows at a rate of 120 m^3 a year and would rise to ~3600 m^3 in 2020 [5]. The inert glass canisters are intended to be ultimately disposed of in an optimised package.

1.2.2. Spent nuclear fuel (SNF)

Concerning recycling of plutonium, the fabrication and use of MOX fuel (around 100 t per year, with 7 wt% Pu content) enable to maintain the amount of separated plutonium at a limited level (plutonium flux adequacy strategy). Future progress is planned to improve the efficiency of MOX fuel (with 8.65 wt% Pu content) and energy equivalence with enriched UO_2 fuel (3.7 wt%).

Monorecycling of plutonium allows today to substantially reduce the amount of spent fuel assemblies to be stored (factor 7). It leads to a limited and acceptable degradation of plutonium stock, compatible with the future energy needs (cf. Section 2.2). The spent MOX fuel destiny is to be reprocessed in the future. As such, it constitutes a long-term potential energy resource option, which could be treated and reused according to future energy needs, especially to fuel future fast reactors, or stored for another period of time, or other options also studied (transmutation, geological disposal).

2. French strategy for the nuclear fleet renewal

2.1. For the mid-term

The first goal is, of course, to maintain the nuclear fleet performances at a high-level, taking advantage of mature industrial capacity in France and abroad, in order to meet the economic challenge of electricity market opening and to go on with long-term and safe operation of existing NPPs.

The preparation of the future of the French nuclear fleet relies on the two following strategic lines which are complementary:

- extending the existing reactors lifetime beyond 40 years, within the framework of the periodic safety reassessment process. EDF's current studies show it is technically possible to reach an average lifetime of 50 years;
- preparing the fleet renewal, with the launching of a first-of-a-kind EPR, scheduled for commissioning in 2012.

The launching of the first-of-a-kind EPR would contribute, first to meeting the rising consumption and, second to preparing for renewal of the nuclear fleet in optimal conditions beyond 2020. The new fleet will be operating during the 21st century: the uncertainties are high on this time frame and obviously progress in the technologies should continue.

2.2. For the far-future

In many relevant scenarios, world energy prospective studies forecast a significant increase in

Table 1
Chemical composition of Areva NC R7/T7 reference glass (FP: fission products, An: actinides – including U and Pu rests, and MP: metallic particles) [1]

Component	SiO_2	B_2O_3	Al_2O_3	Na_2O	Fe_2O_3	NiO	Cr_2O_3	(FP) Ox	An Ox	MP
Fraction (wt%)	45.1	13.9	4.9	10.0	2.9	0.4	0.5	12.4	0.37	1.6

energy needs, despite the improvement in energy efficiency and the efforts in order to master energy demand. Due to the environmental constraints (e.g. on CO_2 emissions) on one hand and to the rarefaction of fossil resources (oil and gas) on the other hand, nuclear energy appears to have to play a significant role in any clean and efficient energy mix, together with clean coal (with carbon capture and sequestration) and renewable energies. For the most of the relevant energy scenarios, Generation IV nuclear systems, breeder reactors, capable of using the natural resource more completely, could prove necessary as early as 2040 or 2050, and deployed during the second half of this century, if the proposed scarce energy scenario effectively takes place. Besides, the Generation IV systems would offer a possibility of optimising high-level waste management (by transmutation of minor actinides), if this option is judged efficient, economical and feasible.

Thus EDF has built a two-step strategy for the renewal of its nuclear fleet: a first step from 2020 on, with the commissioning of Generation III reactors like EPR, and a second step, with the launching of Generation IV systems, around 2040 or later, depending on the increase of the world installed nuclear capacity. A possible scenario for the renewal is presented in Fig. 1.

EDF considers that the priorities in R&D efforts must be given to: (i) the sodium-cooled fast reactor which is the most mature system, as feedback from real industrial experience is available, and (ii) the gas-cooled fast reactor, more innovative but also more challenging than the former.

Regarding the consequences on the fuel management, the reprocessing and recycling strategy developed since the early times of the French nuclear programme, based on a uranium–plutonium cycle, allows the optimised management of fissile and fertile material. It opens the possibility of using the resource constituted by the plutonium concentrated in MOX spent fuel, which is currently stored in cooling pools, to allow the start of future Generation IV reactors. In this way, deploying FR will be possible while maintaining the reprocessing flux at a level consistent with the existing industrial capacities and managing HLW in continuity. In this prospect, EDF studies have shown that the plutonium inventory contained in the spent fuel to be reprocessed (UO_2 and MOX fuels) would be just sufficient to start a FR fleet equivalent to the current PWR fleet, in the five to ten decades to come.

3. Consequences on the needs for waste materials

3.1. Industrial continuity with glasses incorporating more PF and MA

Ensuring the continuation of the treatment of the spent fuel from the PWR requires that the performances of the waste conditioned in compliance with industrial standards be adapted to the foreseeable needs of the power utility, i.e.

- first of all, an increase in the combustion rate up to 60 GWd t^{-1} for the future UO_2 fuels,
- the treatment of the MOX when it will be necessary to recover the plutonium they contain to deploy the Generation IV FR's (see Section 2.2). In comparison with the UO_2 fuel, the MOX fuel features a significantly increased content of minor actinides (Am, Cm) and, therefore, a higher thermal output in the long range.

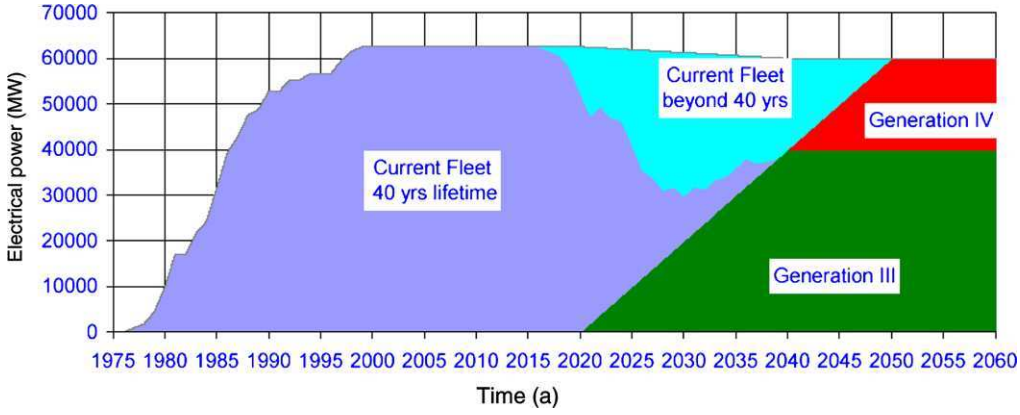

Fig. 1. An example of renewal scheme of EDF's French fleet at an electrical power level of 60 000 MW [6].

For the industrialist, what is at stake therefore consists in adapting the current matrices – the glass – to these new needs, through the continuation of the progress already achieved in the reduction of the volumes, and by striving to define the best technical and economic choices compatible with the long-term management of the waste. In this respect, Section 4 provides a description of some of the research work achieved in France by Areva NC and CEA.

New glass formulations might be envisaged, which would require about ten years of R&D.

3.2. Hypothetical contribution of partitioning and transmutation in the far-future

Another aspect is that pursuant to the logic of ongoing enhancement and follow-up of nuclear power technology, the research projects undertaken within the framework of the 1991 and the 2006 Acts on the MA partitioning and transmutation (P & T) have to be considered as complementary ways of dealing with the HLW. Perhaps they will be able to bring in solutions for the optimisation of the design and utilisation of geological disposal facilities, which will to be validated within the framework of technical and economic optimisation, both of the fuel cycle and the management of the final waste, with respect to the current industrial generation systems. At the present time, disposal costs reduction due to P & T are difficult to estimate, since optimisation of disposal is still to be done. P & T costs are difficult to estimate as well.

This logic of ongoing enhancement is based on the MA transmutation in the FR's that could be achieved in the middle of the century. Research on waste conducted in the framework of the Act of December 30, 1991 has shown that recycling of minor actinides in the present LWR fleet would present important drawbacks, and that new generation FR (or accelerator driven systems) would be much more suitable to do that.

Then three kinds of motivations can be envisaged for P & T [7]:

- to reduce the radiotoxic inventory of the waste or its period,
- to reduce the thermal load of waste on the repository,
- to help in the social acceptation of the disposal, and of the waste itself, by reducing its lifetime.

Hence, in our concern to optimise the waste disposal concepts and space requirements, implementing of MA transmutation would be expected to be an applicable process to reduce the waste's radiotoxic inventory and the thermal load associated with the long-term activity of the vitrified waste canisters disposed of in the repository (Fig. 2). Thus the waste material would be a so-called 'light glass', i.e. mainly containing fission products only, the minor actinides obtained by enhanced separation being intended for incineration in the appropriate reactors. Prior to incurring high expenses for the industrial implementation, a comprehensive and precise cost-benefits balance will have to back up the possible contribution of such a strategy.

Considered under the aspect of the materials, the scientific and technical challenges actually reside less in the sole encapsulating matrix of the fission products than in the fuel itself, i.e. the materials containing the MA's, and suitable – in an appropriate packaging – to be irradiated in a reactor. Such issues as the production of helium through α-disintegration raise questions extremely difficult to answer. The

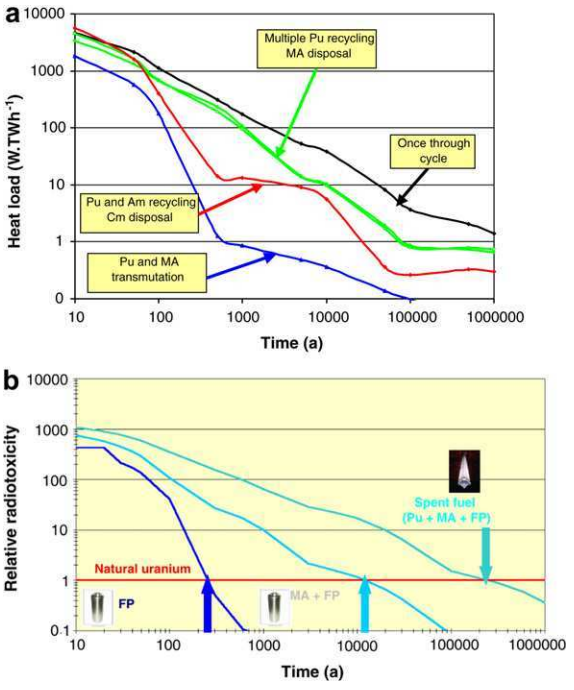

Fig. 2. (a) Reduction of heat load in a repository thanks to P & T (Pu and MA recycling in FR; Pu and Am recycling in PWR and Cm disposal; Pu recycling in FR or Pu multi-recycling in PWR and MA disposal in comparison with once-through cycle) [8]. (b) Reduction of the radiotoxic inventory of the final waste thanks to P & T of Pu (curve MA + FP), then MA (curve FP).

technological development of the fuels and transmutation targets is a long and extensive process that involves in-depth studies of the materials, not to mention the fine-tuning of the processes for their manufacture. To date, the studies dedicated to fuels and transmutation targets have essentially concentrated on the oxides with a moderate actinides content. Fuels and targets have been prepared, and experimental pins on uranium, plutonium, americium and neptunium basis (containing several hundred grams of MA) are irradiated in different reactors, within the scope of various international co-operations (CEA, ITU, DOE, CRIEPI, JAEA, ...) [9,10]. This problem with the fuels and transmutation targets comes within a different scope than that of the radioactive waste forms, and will therefore not be dealt with further in this contribution.

Within the framework of the French 1991 Act, the studies on P & T have led in parallel to research work on the conditioning matrices for separated elements (in pursuance of the partitioning–conditioning logic). They were notably dedicated to the study and characterisation of the specific ceramic materials of certain radionuclides (e.g. zirconolite, britholite, as concerns the actinides, hollandite as for caesium, apatite as for iodine, etc.). All these mineral phases have been subject to particular developments at CEA and CNRS [11,12]. At the end of the seventies, ANSTO in Australia had also finalised the development of composite ceramics, the 'Synroc', for the conditioning of PF solutions.

However, the power utility does not actually express a real need for the matrices dedicated to the management of its waste, insofar as the glass is an excellent matrix that fills the industrial needs, is suitable for storage and of great durability in a geological repository. The interest of a possible partitioning of the radionuclides is thus solely justified with a view to their possible transmutation. The incorporation of separated radionuclides into a dedicated matrix hardly appears justifiable from a technical and economic standpoint.

In the case of iodine, as a matter of precaution, it would be useful to have an iodine-conditioning matrix available, which would take into account, at the level of its elaboration process, the high volatility of this element. Many studies dedicated to iodine-conditioning are carried out in Japan [13], within the scope of the commissionning of the spent fuel processing plant in Rokkashomura. In France, a number of matrices have been assessed. The matrix now chosen is a lead vanado-iodoapatite $Pb_{10}(VO_4)_{4.8}(PO_4)_{1.2}I_2$. One of the points that remains to be evaluated is the extrapolation to the industrial stage of the synthesis process (reactive pressure-sintering) [14]. Another point to be demonstrated is that the disposal of such a matrix would not lead to an unacceptable dispersion of the iodine in the environment.

3.3. Study of the spent fuel management in the context of a non-renewal of the existing fleet of power plants

The strategy currently implemented in France through the choice of reprocessing–recycling leads one to consider spent fuel not as waste, but as recyclable material; besides, as it was said, the current orientation of France's energy policy does not induce to envisage a stop in nuclear power generation in the short term. However, the validation of the reprocessing–recycling strategy will be definitely obtained only from the moment when the available spent fuel (i.e. the not yet reprocessed UO_2 fuel and MOX type fuel) is effectively used for the production of recyclable materials (uranium and plutonium), and the aforesaid materials will be implemented in the deployment of fast neutron reactors. Insofar as the implementation of this strategy cannot be envisaged from a technological point of view by the year 2040 only, the confirmation of the future of spent fuel put on standby for future reprocessing will probably not be obtained before 2020. Meanwhile, what matters, through the adaptation of a cautious envelope approach, is to maintain the research programmes necessary to (i) gain deeper knowledge of the long-term storage behaviour of spent fuel (in particular MOX), and (ii) ascertain the feasibility of its disposal in deep geological layers.

Nonetheless, whatever their future might be, the power utility must be concerned about the behaviour of the spent fuel it bears responsibility for, throughout its subsequent life stages after use in a reactor (transport, storage). The discussion of all of these topics must account for the evolution of the features of the fuels (fuel type, burn-up increase, new cladding materials, etc.). Section 5 describes the stakes involved in the work carried out in France in this field.

4. Advanced glasses for the future

The increase of burn-up up to 60 GWd t^{-1} for the future UO_2 and the reprocessing of MOX fuels

lead to an increase in the quantity of the FP and MA per fuel ton. In order to minimise the number of packages produced per ton of fuel, the waste incorporation rate will have to be increased.

The limit rate for the incorporation of FP and actinides in the glass is due, on the one hand, to the chemical limits and, on the other hand, to the consequences of the self-irradiation of the glass.

The first limitation to the increase of the incorporation rate is due to the capacity of the glass to 'digest' the increased content of FP and MA (Am, Cm). In the glass grades currently produced by Areva NC, the nominal content of FP and MA oxides is approximately 15 wt%, and the maximum value 18.5 wt%. The means to further increase the incorporation limits of the FP and actinides in the glass are known and studied by Areva NC and CEA. The chemical 'digestibility' of glass grade R7/T7 can be enhanced, if necessary

- through an increase in the glass manufacturing temperature [15] (Fig. 3),
- by making the melt more reducing during glass manufacture [16,17] (Fig. 4).

The second limit is related to the consequences of self-irradiation, notably the one associated with alpha radiation (preponderant after several hundred years), when one increases the actinides content of the glass. Indeed, in the nuclear glass grades, the α self-irradiation is likely to make changes in some properties (vitreous state and mechanical resistance), create stress (swelling, cracking, gas bubbles, etc.), or even change the chemical durability of the glass. Our current standard of knowledge shows that the properties of the confinement glass grades

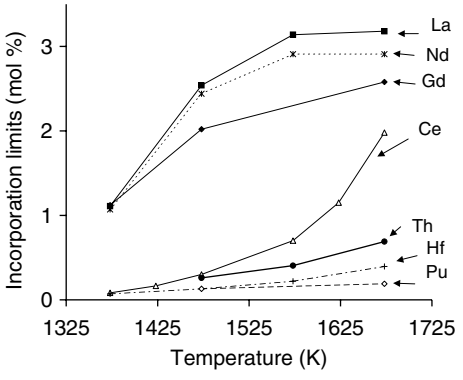

Fig. 3. Solubility limits of different elements in a boro-silicate glass as function of temperature [15].

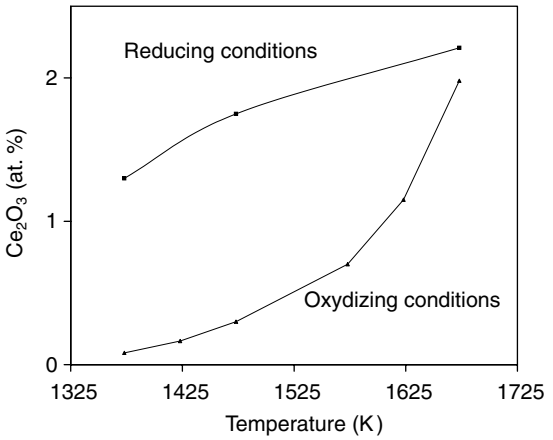

Fig. 4. Cerium solubility in a borosilicate glass as function of redox process conditions [17].

are not altered up to to an alpha disintegrations dose of at least about 10^{19} g^{-1}. The characterisation of $^{244}CmO_2$-doped glass, supported by fundamental and modelling studies, is under way at CEA to bring in supplementary demonstration elements (Fig. 5).

The third limit to the increase in the incorporation rate is due to the capacity of the glass to support the thermal load generated by the disintegration of the radionuclides contained in the glass (in particular fission products and ^{244}Cm). To ensure the stability of the glass, the core temperature of the stored package shall not exceed the vitreous transition temperature. It is precisely for this reason that new formulations of borosilicated, rare earth-enriched glass grades are currently subject to laboratory studies: these glass grades would allow an increase in the vitreous transition temperatures by about 50 K, and thus make possible an increase in the thermal load by 15% with respect to the current limit. However, one may consider that the present incorporation rate of HLW in a matrix, of about 15%, is already high, in the sense that the thermal power of each waste package may be the main constraint, but need to be associated to interim storage or disposal design. A higher density would involve a higher core temperature or could require an adjustment of the waste package size. This consequence is not definitively a limit in volume reduction, but could imply handling an increased number of waste packages to be stored and disposed of. A clear advantage of the (present and) future treatment methods is that it will be possible to adjust this density to the disposal concept. Thus, a maximum waste density may not necessarily be the optimum.

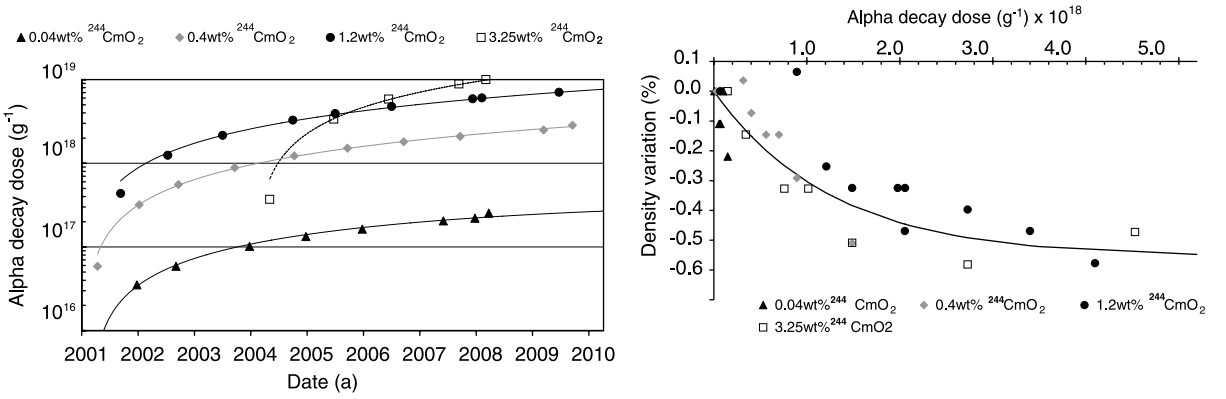

Fig. 5. Effect of an increase in the dose on glass behaviour by means of actinides-doped glass grades (^{244}Cm).

5. Spent nuclear fuel

The operational needs expressed in France as for the transport, storage (wet or dry) and possibly the disposal of spent fuel raise a certain number of major technical and scientific questions. The questions related to the material science concern in particular, are:

- knowledge of the medium and long range intrinsic evolution of the fuel pellet (leaktight fuel rods),
- resistance of the cladding during the initial life stages of the fuel further to its withdrawal from the reactor (transport, storage),
- reprocessing/recycling capacity of the fuel assemblies after a storage period,
- resistance against aqueous corrosion of the fuel pellet under storage conditions.

5.1. Intrinsic evolution of the fuel pellet in the medium and long-term

Spent fuels feature a heterogeneous structure (Fig. 6); unlike the glass, they were not designed from the beginning to ensure a long-term confinement function. Once withdrawn from the reactor, the fuel evolves under the combined effects of its internal physical and chemical unbalances that result from its life phase in the reactor (composition gradients, mechanical stress, etc.) and the driving forces made up by the radioactive and thermal decrease.

Hence, the spent fuel features a heterogeneity of radionuclide localisation in the pellet; in particular, certain radionuclides are not retained by the oxide

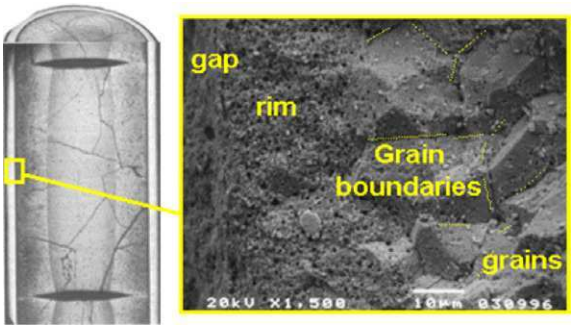

Fig. 6. State of the fuel pellet and fuel microstructure after irradiation in reactor (UO_2 spent fuel, burn-up: 60 GWd t^{-1}).

matrix and are therefore likely to be released to the exterior should the fuel rod leak during the life phases following its reactor life. One major feature of spent fuel is therefore the existence of an activity instantaneously released (IRF) from the fuel at the cladding failure (Fig. 7). This labile activity potentially exists as early as at the time of the withdrawal from the reactor; it then evolves in time under the effect of different processes. A conservative model was developed by CEA to predict the evolution in time of this unsteady fraction [18,19]. Developing a more realistic, but robust model, notably for the MOX, both for the medium-term and the long-term storage represents a scientific challenge.

The irradiated fuel pellet principally evolves due to the effect of the decrease in the radioactivity of the nuclei residing in its core, an evolution that leads to an accumulation of irradiation damages, along with a modification of the chemical inventory and a significant production of helium due to a decrease in the emission of α particles. The evolution of the chemical inventories does not significantly impact

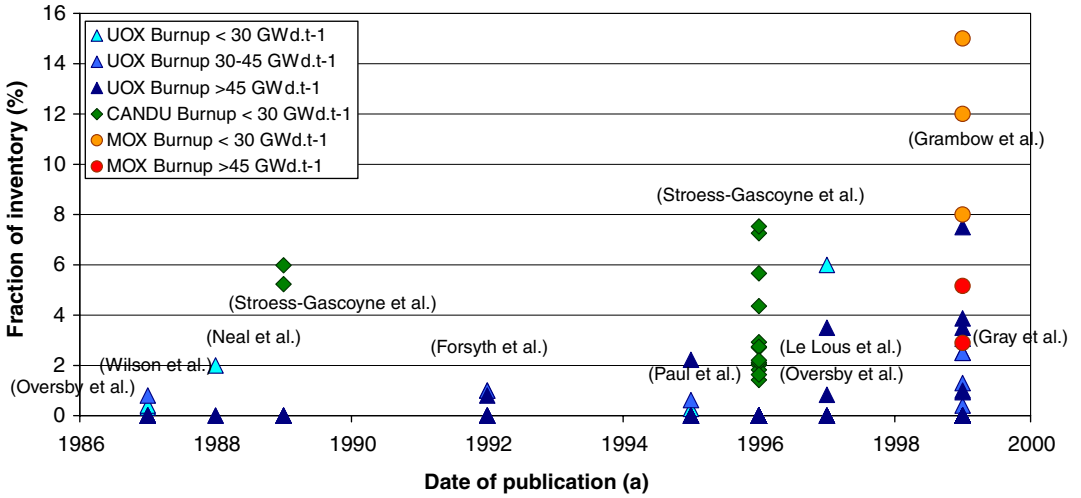

Fig. 7. Distribution of experimental IRF data for Cs during the last years.

the oxidation state of the fuel pellet [20], whereas the decrease in the emission of α particles leads to the accumulation of a large quantity of helium within the pellet, particularly in the case of fuels rich in α emitters, such as MOX (Fig. 8). How does this gas, mainly produced within the grains of the fuel pellets, evolve? Does it remain confined in the oxide $(U,Pu)O_2$ (the solubility of He in UO_2 at temperatures below 573 K is low – <1 at.%), or, on the contrary, diffuse throughout the grains, so that it will be gradually released, at first towards the grain interfaces and then into the free spaces within the rod? The research results available at CEA lead one to assume that at intermediate storage time scales, the α self-irradiation should not cause significant helium diffusion. However, in the long-term, helium should accumulate in intragranular bubbles possibly causing micro-cracks. This phenomenon should accelerate the helium release in grain boundaries and, by increasing the preexisting fission gas bubbles pressure, lead to the opening of grain boundaries as observed in old PuO_2 pellets (Fig. 9). It is hence difficult to currently be conclusive on grain boundaries stability over a long time. The long-term fate of the helium in spent fuel is the major open question concerning the behaviour of the spent fuel in a closed system. Grain boundaries stability is also a major scientific issue to be addressed in

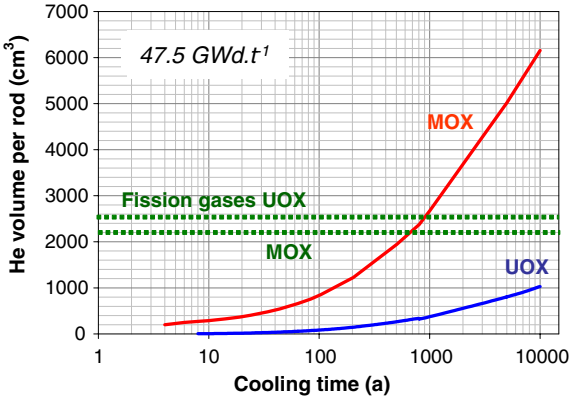

Fig. 8. He produced by radioactive decay in UO_2 and MOX fuels with a burn-up of 47.5 GWd t^{-1} calculated with the CESAR code [21].

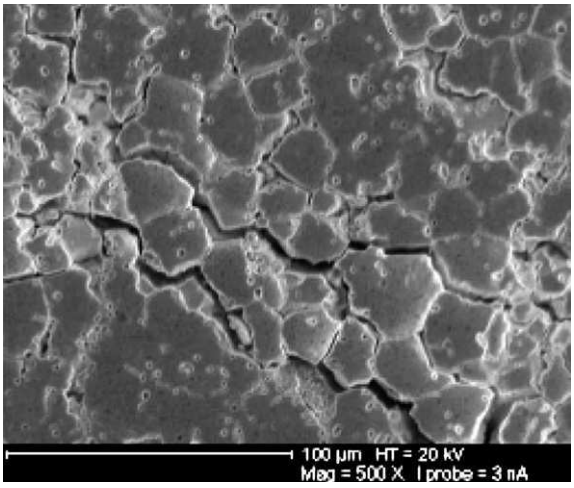

Fig. 9. $^{238}PuO_2$ (90% ^{238}Pu) pellet (after cutting and polishing) stored during 30 years in inert atmosphere [21].

the near-future to better understand the spent fuel long-term evolution and performances.

5.2. Ensuring that the cladding withstands transport and interim storage

End-of-life internal pressure measured after irradiation in reactor ranges from 40 to 60 bars at room temperature. Thus, the cladding will be subjected to significant hoop stresses (from about 70 to 120 MPa) for the fuel rod temperatures as expected during transport or at the beginning of dry storage (up to 673 K). The main mechanism, activated by temperature, which could modify the cladding properties in these conditions is creep. Due to temperature and stress decrease with time, creep is expected to be particularly active during the first stages after irradiation (transport, beginning of dry storage).

Further to the research work already achieved on the Zircaloy-4, creep models have to be developed for the different cladding materials used, taking into account:

- the microstructural state of the alloys (stress-relieved or recrystallised),
- the irradiation defects annealing observed at relatively high temperature (this temperature depends on the duration, stress and hydrogen content).

Another challenge is the definition of a realistic strain criterion of rupture of cladding, taking into account the effects of irradiation defects annealing and recrystallisation.

To prevent any problems that may be posed in the long-term by the thermally activated phenomena, such as creep, wet storage may be preferred to dry storage. Actually, wet storage does not pose any major problem, provided that the fuel assemblies are stored in water, the quality of which is monitored. The zirconium alloys show extremely high resistance to general and localised corrosion under these conditions. The sole issue likely to be encountered is their potential brittleness due to the hydride formation they underwent in the reactor. Indeed, the Zircaloy–hydride precipitates feature low ductility (i.e. brittleness) at low temperatures ($<$373 K) and present, due to this very fact, the risk of cracking under particular mechanical loads (flexion, shocks) in the course of fuel assembly or rod handling operations.

5.3. Ensuring fuel reprocessing capability after interim storage

- To ensure the reprocessability of the fuels after their interim storage implies that one is capable of knowing and accounting for the development of the mechanical and physical properties of the fuel assemblies, i.e. of the cladding that envelopes the fuel itself and the structural components (grid assemblies, guide thimbles, end-pieces), and of the UO_2 matrix. The mechanical integrity of the fuel assembly has to be guaranteed, not only in a normal situation, but also under incident conditions.
- Should the fuels be brought into contact – by incident or accident – with an oxidising atmosphere, the resulting risk will be the swelling of the pellet and its disaggregation during the oxidation process that will transform the UO_2 into U_3O_8. The principal questions concern the evolution of the physical state of the fuel rods and the effects of oxidation on the release of radionuclides.

5.4. Forecasting the evolution of the fuel in geological disposal

The issues related to fuel disposal are twofold:

- characterisation of the fraction of the total inventory not retained by the matrix and being in a 'labile' state of activity (IRF) upon the coming-into-contact of the fuel with water [19];
- determination of the long-term alteration mechanism and kinetics of the matrix, knowing that disposal safety essentially relies on the chemical stability of the matrix itself, which contains the major part of the fission product inventory and virtually all of the heavy nuclei [22].

The first term, which corresponds to a rapid release of radionuclides when water enters the fuel rod (IRF), was already mentioned within the Section 5.1. IRF values have been proposed for geological disposal.

The second term corresponds to the slow release of the radionuclides located within the grains. Up to now, the studies have focused on the effect of the water α and γ radiolysis on the spent fuel matrix dissolution, and models for oxidative dissolution of the matrix governed by α or β/γ radiolysis have been developed. Other models based on the solubility

have also been developed, which involve a less release rate.

The needs in terms of knowledge acquisition and model development require that due consideration will be given to the geochemical environment of the fuel packages (for example, the hydrogen inhibiting effect on matrix dissolution). Various experiments have shown an activity threshold under which the dissolution is not controlled by the water radiolysis but by environmental conditions. This activity threshold must still be defined in conditions representative of the near field conditions. In order to improve the matrix dissolution model, it is necessary to consider these effects (presence of corrosion products, H_2, etc.). The role of secondary phases on the radionuclides release also has to be better studied.

6. Conclusion

The French current reprocessing–recycling strategy allows to use fissile material rationally in the long-term while benefiting from existing industrial tools in the long run. With these existing industrial tools, it brings a robust answer to high-level waste management through vitrification, while meeting the cost effectiveness requirement, respecting the environment and preserving the long-term energy options.

In pursuance of the previously adapted strategic objectives, and while allowing to take lasting advantage of the investments made, this strategy makes it possible to preserve the sustainable nuclear options based on the uranium–plutonium cycle, by way of optimised management of the fissile and fertile materials (depleted uranium). It opens up the possibility of eventually using the plutonium inventory concentrated in the spent MOX fuels to provide for the start-up of the future fast Generation IV reactors.

EDF and its partners are investigating the consequences of this strategy in terms of high-level and long-lived waste management that result from the reprocessing of high burn-up UO_2 fuels and from the future reprocessing of spent MOX fuels. The R&D work under way should make it possible to eventually come up with solutions for the further enhancement of the glass grades, in various aspects: their chemical 'digestibility' that can be increased by a higher manufacturing temperature, the increase of the α-self-irradiation, the thermal disposal conditions.

The strategy currently implemented in France via the choice of the reprocessing–recycling leads to consider the spent fuels as recyclable materials, and not as waste. However, to be on the safe side and to enable to decide with full knowledge of the facts about the future of not-yet-reprocessed spent fuels, further research work has to be undertaken to characterise the alteration modes and behaviour of the spent fuels (UO_2 and MOX) during long-term storage and geological disposal. The most critical issue is probably the long-term stability of the fuel pellet grain boundaries.

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